

Atmospheric aerosols from the Earth's surface to the stratosphere: A global model perspective

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Declaration

I hereby declare that I wrote the dissertation submitted without any unauthorized external assistance and used only sources acknowledged in the work. All textual passages which are appropriated verbatim or paraphrased from published and unpublished texts as well as all information obtained from oral sources are duly indicated and listed in accordance with bibliographical rules. In carrying out this research, I complied with the rules of standard scientific practice as formulated in the statutes of Johannes Gutenberg-University Mainz to insure standard scientific practice.

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Abstract

Atmospheric aerosols play a key role in the Earth’s climate system, influencing the radiative balance, the hydrological cycle, atmospheric chemistry, air quality, and human health. However, their precise effects depend strongly on their spatial and temporal distribution, as well as their properties such as mass, surface area and number concentration, and their chemical composition. Thus, the uncertainties on climate and health effects of atmospheric aerosols remain large. While in-situ and remote sensing observations provide valuable insights, they are often limited by their spatial and temporal coverage or by their inability to directly quantify key aerosol characteristics. To extrapolate these observations to global scales, comprehensive global atmospheric chemistry models are essential.

This thesis presents a series of studies aimed at improving global simulations of atmospheric aerosols from the surface to the stratosphere using the ECHAM5/MESSy2 Atmospheric Chemistry (EMAC) model. The improved model setup forms the basis for a more accurate representation of global aerosol distributions and composition, supporting a better understanding of their climate and health impacts, and advancing the knowledge of key aerosol-related processes.

The first study presents a global EMAC simulation of annually averaged ultrafine particle (UFP) concentrations at the Earth’s surface, evaluated against in-situ observations from field campaigns, the EBAS database, literature and published datasets. The evaluation results are used as guidance for a novel emission-based downscaling of UFP concentrations to a horizontal resolution of $0.1 \times 0.1^\circ$. Recent studies suggest that long-term exposure to high UFP concentrations has a significant, on the global scale mostly unexplored, impact on public health. This first global UFP dataset provides a foundation for global health assessments. Additionally, the model improvements and refinements of primary particle emission size distributions developed in this process are crucial for accurately simulating the horizontal and vertical transport of primary emitted particles.

The second study introduces and evaluates the newly developed MESSy submodel for emissions from Explosive Volcanic ERuptions (EVER), designed to simulate gaseous and aerosol emissions using user-defined vertical distributions. This submodel is complemented by a refined historical volcanic eruption configuration, developed based on an existing emission inventory (1990–2023), with emission parameters optimized through sensitivity analyses conducted in this work. Whereas previous studies on stratospheric volcanic eruptions required manual intervention during simulations, the new submodel automatically integrates volcanic emissions at run-time. This model improvement is essential for aerosol studies in the upper troposphere and stratosphere. Additional use cases for the EVER submodel range from simulations of large-scale wildfire emissions to assessments of aerosol-based solar radiation modification strategies.

The third study presents global distributions of aerosol mass, number concentrations and

chemical composition of atmospheric aerosols from the surface to the stratosphere, based on a newly developed EMAC model setup. This configuration combines elements of previously used tropospheric and stratospheric setups and integrates the developments from the preceding studies. Simulations are evaluated using a comprehensive set of observations, including ground-based measurement stations, aircraft campaigns, and satellite remote sensing. While the previous tropospheric and stratospheric setups were incompatible and unable to consistently reproduce aerosol processes from the surface to the stratosphere, the new setup enables investigations of interactions between the lower troposphere and the stratosphere. This advancement opens the door for future studies on anthropogenic influences on aerosol distributions, long-range aerosol transport, online aerosol-cloud-radiation interactions, and stratospheric ozone chemistry. Moreover, it supports a deeper understanding of observed aerosol processes and their underlying mechanisms.

The three main studies are complemented by a summary of additional research efforts that build directly upon the developments presented in this thesis. First, the improved model setup was applied in two studies investigating the formation of the Asian Tropopause Aerosol Layer (ATAL) during the Asian monsoon season, as well as the subsequent transport and fate of aerosols formed within the ATAL, supporting recent aircraft-based observations. Second, the setup was used in the context of the CAFE-Brazil aircraft campaign over the Amazon basin, exploring the sensitivity of regional atmospheric chemistry and global climate to changes in biogenic volatile organic compound emissions, and analysing aerosol formation and cycling in the region. Third, the influence of non-stratospheric volcanic eruptions on sulfate concentrations in the lowermost stratosphere was examined, focusing on mixing processes in the tropopause region. Fourth, global UFP concentrations were further refined using machine learning to achieve even higher horizontal resolution. This dataset, in combination with the previously developed global UFP map, serves as the basis for a global study on UFP-related public health impacts.

In summary, this work strongly improves the agreement of EMAC simulations with observations across all layers of the lower atmosphere, and provides a solid foundation for model studies extending from the surface to the stratosphere. These advancements may contribute to reducing uncertainties in aerosol-related climate impacts, as emphasized in the latest IPCC reports, as well as in the assessment of health effects associated with aerosol air pollution. The key developments presented here are applied in a range of scientific studies, advancing our understanding of global aerosol distributions, their chemical composition, associated climate and health effects, and the underlying aerosol processes.

Zusammenfassung

Atmosphärische Aerosole spielen eine zentrale Rolle im Klimasystem der Erde, da sie Strahlungsbilanz, Wasserkreislauf, Atmosphärenchemie, Luftqualität sowie menschliche Gesundheit beeinflussen. Ihre genauen Auswirkungen hängen jedoch von ihrer räumlichen und zeitlichen Verteilung sowie ihren Eigenschaften wie Masse, Oberfläche, Anzahlkonzentration und chemischen Zusammensetzung ab. Daher bestehen weiterhin erhebliche Unsicherheiten hinsichtlich der Klima- und Gesundheitseffekte von Aerosolen. In-situ- und Fernerkundungsmessungen liefern zwar wertvolle Erkenntnisse, sind jedoch häufig durch ihre begrenzte räumliche und zeitliche Abdeckung oder durch die fehlende Möglichkeit, Aerosoleigenschaften direkt zu quantifizieren, eingeschränkt. Um Beobachtungen auf globale Ebene zu übertragen, sind umfassende globale Atmosphärenchemie-Modelle notwendig.

In dieser Arbeit wird eine Reihe von Studien mit dem Ziel, globale Simulationen atmosphärischer Aerosole von der Erdoberfläche bis zur Stratosphäre mithilfe des ECHAM5/MESSy2 Atmospheric Chemistry (EMAC) Modells zu verbessern, vorgestellt. Das weiterentwickelte Modell bildet die Grundlage für eine genauere Darstellung globaler Aerosolverteilungen und -zusammensetzung. Damit unterstützt es ein vertieftes Verständnis ihrer Klima- und Gesundheitseffekte sowie zentraler Aerosol-Prozesse.

In der ersten Studie wird eine globale EMAC-Simulation der jährlich gemittelten Konzentrationen ultrafeiner Partikel (UFP) an der Erdoberfläche vorgestellt, validiert anhand von In-situ-Beobachtungen aus Feldkampagnen, der EBAS-Datenbank, Fachliteratur und veröffentlichten Datensätzen. Die Ergebnisse der Evaluierung dienen als Grundlage für ein neuartiges, emissionsbasiertes Downscaling der UFP-Konzentrationen auf eine horizontale Auflösung von $0,1 \times 0,1^\circ$. Neue Studien deuten darauf hin, dass eine langfristige Exposition gegenüber hohen UFP-Konzentrationen erhebliche Auswirkungen auf die Gesundheit haben kann, was auf globaler Ebene jedoch bislang weitestgehend unerforscht ist. Dieser erste globale UFP-Datensatz bietet eine Grundlage für globale Studien zu Gesundheitseffekten von UFP-Exposition. Darüber hinaus sind die im Rahmen dieser Studie entwickelten Modellverbesserungen und Verfeinerungen der Emissionsgrößenverteilungen der primären Partikelemissionen von entscheidender Bedeutung für die realistische Simulation des horizontalen und vertikalen Transports primär emittierter Aerosole.

In der zweiten Studie wird das neu entwickelte MESSy-Submodul für Emissionen explosiver Vulkanausbrüche (EVER), das Gas- und Aerosolemissionen mithilfe benutzerdefinierter vertikaler Verteilungen simulieren kann, vorgestellt und evaluiert. Ergänzt wird dieses Submodul mit einer Modellkonfiguration, die historische Vulkanausbrüche, basierend auf einem bestehenden Emissionsinventar (1990–2023), enthält. Die genauen Emissionsparameter wurden im Rahmen dieser Arbeit durch Sensitivitätsanalysen optimiert. Während frühere Studien zu stratosphärischen Vulkanausbrüchen manuelle Eingriffe während der Simulationen erforderten, integriert das neue Submodul die vulkanischen Emissionen automatisch zur Laufzeit. Diese Modellverbesserung ist von zentraler Bedeutung für Aerosolstudien in der

oberen Troposphäre und Stratosphäre. Weitere Anwendungsfälle reichen von der Simulation großflächiger Waldbrandemissionen bis hin zur Untersuchung aerosolbasierter Strategien zur Veränderung der Sonneneinstrahlung.

Die dritte Studie präsentiert globale Verteilungen der Masse, Anzahlkonzentration und chemischen Zusammensetzung atmosphärischer Aerosole von der Erdoberfläche bis in die Stratosphäre, basierend auf einer neu entwickelten EMAC-Modellkonfiguration. Diese Konfiguration kombiniert Elemente bisher verwendeter troposphärischer und stratosphärischer Setups und integriert die Entwicklungen der vorhergehenden Studien. Die Simulationen werden anhand von umfassenden Beobachtungsdaten aus bodengestützten Messstationen, Flugzeugkampagnen und satellitengestützter Fernerkundung evaluiert. Während die bisherigen troposphärischen und stratosphärischen Modellkonfigurationen unvereinbar waren und Aerosolprozesse von der Erdoberfläche bis in die Stratosphäre nicht konsistent abbilden konnten, ermöglicht das neue Setup nun Untersuchungen zu Wechselwirkungen zwischen unterer Troposphäre und Stratosphäre. Dieser Fortschritt eröffnet Möglichkeiten für künftige Studien zu anthropogenen Einflüssen auf Aerosolverteilungen, zum Transport von Aerosolen, zu Echtzeit-Wechselwirkungen zwischen Aerosolen, Wolken und Strahlung sowie zur Ozonchemie. Darüber hinaus trägt das Modell zu einem tieferen Verständnis beobachteter Aerosolprozesse und ihrer zugrunde liegenden Mechanismen bei.

Die drei Hauptstudien werden durch eine Zusammenfassung zusätzlicher Forschungsarbeiten ergänzt, die unmittelbar auf den in dieser Dissertation vorgestellten Entwicklungen aufbauen. Erstens wurde das verbesserte Modell-Setup in zwei Studien angewandt, in denen die Bildung der asiatischen Tropopausen-Aerosolschicht (ATAL) während der asiatischen Monsunzeit sowie der anschließende Transport und Verbleib der innerhalb der ATAL gebildeten Aerosole untersucht wurden, was neue flugzeug-gestützten Beobachtungen unterstützt. Zweitens fand das Setup Anwendung im Rahmen der CAFE-Brazil-Kampagne über dem Amazonas, zur Untersuchung der Empfindlichkeit der regionalen atmosphärischen Chemie und des globalen Klimas gegenüber Veränderungen in Emissionen biogener flüchtiger organischer Verbindungen sowie zur Analyse der Aerosolbildung in dieser Region. Drittens wurde der Einfluss nicht-stratosphärischer Vulkanausbrüche auf Sulfatkonzentrationen in der unteren Stratosphäre untersucht, mit Fokus auf Mischungsprozesse in der Tropopausenregion. Viertens wurden die globalen UFP-Konzentrationen mithilfe von Methoden des maschinellen Lernens weiter verfeinert, um eine noch höhere horizontale Auflösung zu erzielen. Dieser Datensatz bildet, in Kombination mit der zuvor entwickelten globalen UFP-Verteilung, die Grundlage für eine weltweite Studie zu gesundheitlichen Auswirkungen von UFP-Exposition.

Zusammenfassend verbessert diese Arbeit die Übereinstimmung der EMAC-Simulationen mit Beobachtungsdaten in allen Schichten der unteren Atmosphäre erheblich und schafft eine solide Grundlage für Modellstudien, die sich von der Erdoberfläche bis in die Stratosphäre erstrecken. Diese Fortschritte können dazu beitragen, die Unsicherheiten in Bezug auf aerosolbedingte Klimaeffekte, wie sie in den jüngsten IPCC-Berichten hervorgehoben wurden, sowie bei der Bewertung gesundheitlicher Auswirkungen von Aerosolen zu verringern. Die hier vorgestellten zentralen Entwicklungen finden Anwendung in einer Vielzahl wissenschaftlicher Studien und fördern das Verständnis globaler Aerosolverteilungen, ihrer chemischen Zusammensetzung, der damit verbundenen Klima- und Gesundheitseffekte sowie der zugrunde liegenden Aerosolprozesse.

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1 CHAPTER

Motivation

Particulate matter in the Earth's atmosphere, whether solid or liquid, is a key driver of Earth's climate, the hydrological cycle, and air quality, thus impacting human health and life on the planet in general [1]. The inhalation of aerosol air pollution potentially leads to severe diseases and increased premature mortality [2, 3, 4, 5]. Aerosols as seeds for cloud droplets and ice particles impact cloud formation, lifetime, and precipitation patterns [6, 7, 8], and thereby exert indirect climate effects [6, 7, 9]. Scattering and absorption of radiation by aerosols additionally cause direct radiative forcing [1], while aerosols also influence atmospheric chemistry in general, e.g., by playing a key role in ozone depletion [10, 11]. These effects are well established, yet their magnitude and variability remain among the largest uncertainties in climate and health impact assessments [1, 2, 9, 12].

A major reason for these uncertainties lies in the limited knowledge of global aerosol distributions, composition, and size across all atmospheric layers [1, 12]. Understanding aerosol sources, formation, transport, and removal is inherently complex. Aerosols originate from both natural and anthropogenic sources, undergo chemical transformations, and interact with meteorology [13, 14, 15]. Their impacts depend not only on mass, but also on number concentration, surface area, and chemical composition, properties that are challenging to measure or represent globally.

Observations, though essential, suffer from gaps in spatial and temporal coverage, especially in the logistically challenging free and upper troposphere, and the stratosphere. In-situ measurements are sparse, remote sensing is constrained to the observation of aerosol optical properties, and aircraft campaigns are episodic. At the same time, chemistry-climate models offer the potential to simulate and predict aerosol processes on a global scale. However, they remain limited by horizontal resolution, the parameterization of sub-grid-scale processes, and incomplete knowledge of real-world emissions and chemical pathways. The full potential of both observations and modelling is realized when they complement each other. On the one hand, modelling is crucial for the understanding of processes that produce the observed aerosol properties. On the other hand, model results are only reliable when supported by observational data. Together, they enable the creation of a comprehensive global picture of atmospheric aerosols and their processes.

In recent years, ultrafine particles (UFP) have gained increasing attention in health studies. While aerosol health effects were traditionally linked to $PM_{2.5}$, recent findings suggest that UFP number concentrations may contribute significantly to premature mortality, as they can penetrate deeper into the bloodstream [16, 17, 18, 19]. However, no information on UFP concentrations on a global scale has been available yet. Likewise, information on global

aerosol distributions in the free and upper troposphere, and the stratosphere, is scarce due to observational constraints, leading to large uncertainties in these regions [20, 12].

This thesis aims to advance the global representation and understanding of atmospheric aerosols from the surface to the stratosphere, enhancing the ECHAM5/MESSy2 Atmospheric Chemistry (EMAC) model. Adjusted particle emission size distributions and revisited particle formation pathways enable a first global simulation of UFP particles at the Earth's surface, and improve the representation of primary particle transport to and across the tropopause. The automatic inclusion of stratospheric volcanic eruptions in EMAC simulations better represents the stratospheric aerosol layer. Finally, a new model setup with adjusted aerosol size distributions enables consistent global simulations of key aerosol species across the entire lower atmosphere.

Ultimately, this work shall contribute to reducing uncertainties and support future studies on aerosol-climate-health interactions, as well as supporting detailed process studies, as demonstrated in Chapter 6.

2 CHAPTER

Theory

2.1 The atmosphere of the Earth

The Earth's atmosphere is very thin relative to its radius, yet it is very important for sustaining life by providing breathable air, maintaining liveable ambient temperatures, and shielding organisms from harmful cosmic radiation [15]. For a very long period in the history of humankind, the atmosphere was shaped solely by natural processes, with very limited anthropogenic influence. Since the Industrial Revolution, human activities have increasingly impacted the environment on all scales, and emitted chemical substances into the atmosphere, influencing climate, dynamics, and composition of the atmosphere, leading to the declaration of a new epoch, the *anthropocene* [21], and putting the liveable conditions on Earth at risk [1].

The chemical composition of the atmosphere is predominantly made up of nitrogen (N_2 , 78%), oxygen (O_2 , 21%) and argon (Ar, 0.9%). This basic composition remains fairly constant throughout the atmosphere and over time, with very little variability. However, despite these primary gases already adding up to almost 100%, the atmosphere consists of thousands of additional trace gas and particulate species, that mostly drive climate, atmospheric chemistry and dynamics [15].

Earth's atmosphere is vertically subdivided into different regions, mostly based on the vertical temperature gradient [22, 15]:

Troposphere The troposphere extends from the Earth's surface to an altitude of approximately 8 km at the poles and up to 16 km at the equator, with the tropopause as upper boundary. 80% of the total atmospheric mass are contained in the troposphere, and nearly all weather phenomena occur within this layer. It is characterized by continuous turbulence, convection and mixing, reflected in its name, derived from the Greek “tropos”, meaning “turn” or “change”. Temperatures decrease with height, with a typical lapse rate of about -6.5 K km^{-1} . The troposphere can be further subdivided into the boundary layer, the lowest few kilometres of the troposphere influenced by surface processes, the free troposphere, and the upper troposphere, which is influenced by interactions with the stratosphere.

Stratosphere Above the tropopause, temperatures begin to increase in the stratosphere due to the absorption of shortwave radiation by ozone (O_3). This warming trend continues up to an altitude of 45 to 55 km, where the stratopause is located. The temperature increase leads to stable stratification (hence the name “stratosphere”), resulting in limited vertical mixing. In addition to the ozone layer, the stratosphere also contains the Junge aerosol layer, located between 12 and 28 km, and primarily composed of sulfate aerosols.

Upper atmosphere The upper atmosphere begins above the stratopause and includes several layers. The mesosphere, extending up to about 80 to 90 km, is characterized by decreasing temperatures and rapid vertical mixing. Above the mesosphere lies the thermosphere, where temperatures increase again due to the absorption of short-wave radiation by N_2 and O_2 , extending up to around 500 km. Finally, the exosphere marks the outermost layer, where gas molecules can escape Earth's gravitational field.

2.1.1 Radiation in the atmosphere

The Earth's primary energy source is electromagnetic radiation from the sun [23, 15, 24], which fundamentally drives Earth's energy balance, weather, climate and atmospheric chemistry. The total radiant energy from the sun, reaching the top of Earth's atmosphere per unit time and area, can be expressed via the solar constant S [25]:

$$S = \sigma T_S^4 (a_s/r_0)^2 \approx 1370 \text{ W m}^{-2} \quad (2.1)$$

T_S denotes the temperature of the solar photosphere, a_s the radius of the visible solar disk and r_0 the mean sun-earth distance. $\sigma = 5.67 \times 10^{-8} \text{ W m}^{-2} \text{ K}^{-4}$ is the Stefan-Boltzmann constant. Approximately 30% of the solar short-wave radiation is reflected back to space, while the remaining 70% is absorbed by the Earth's surface [26]. As a consequence, the Earth is heated and re-emits this energy as long-wave radiation, primarily in the infrared spectrum. In equilibrium, Earth's temperature would adjust, such that the incoming solar radiation equals the outgoing terrestrial long-wave radiation, with an equilibrium temperature

$$T_e = \left(\frac{S(1-a)}{4\sigma} \right)^{1/4} \approx 255 \text{ K}, \quad (2.2)$$

where $a = 0.3$ is the Earth's albedo, i.e., the ratio of back-scattered to total incident short-wave radiation. However, while radiation travels through the atmosphere, it interacts with its constituents, such as gases, clouds and aerosols. In general, the light can either be absorbed, i.e., the energy of the beam is converted to thermal energy, or scattered, i.e., the molecule is excited to perform oscillations and re-emits to all directions. A fraction of the long-wave radiation from the Earth is absorbed by greenhouse gases, such as CO_2 , CH_4 and water vapour, and re-emitted. This greenhouse effect is essential for life on planet, as it enables the liveable temperature on Earth's surface that we encounter.

If the atmospheric composition changes, this could lead to a change in the incoming or outgoing radiation and therefore an imbalance between incoming and outgoing radiation, leading to radiative forcing. Radiative forcing quantifies the change in net (down minus up) radiative flux at the top of the atmosphere due to such perturbations. Positive forcing leads to warming, while negative forcing leads to cooling. Since the industrial era, concentrations of greenhouse gases, such as CO_2 and CH_4 , steadily increased, leading to a positive radiative forcing, and thus increasing temperatures, threatening life on Earth [1]. In addition, naturally or anthropogenically induced changes in aerosol concentrations can alter the Earth's albedo, and thereby affecting the incoming short-wave radiation, again resulting in a radiative forcing (see Sect. 2.2.2).

2.1.2 Thermodynamic basics

The atmosphere can, to a very close approximation, be treated as an ideal gas, meaning that its gas molecules can be considered as point particles moving randomly without significant inter-particle forces [27]. An ideal gas is described by the equation of state, also referred to as the ideal gas law:

$$p \cdot V = N \cdot k_B \cdot T \quad (2.3)$$

In this equation, pressure is represented by p , the volume of an air parcel by V , the number of molecules N and the temperature by T . The Boltzmann constant $k_B = 1.381 \times 10^{-23} \text{ J K}^{-1}$ represents the constant of proportionality.

Atmospheric pressure Atmospheric pressure p is one of the most fundamental variables in the atmosphere, and gradients in pressure are the main driver for atmospheric motions. The standard atmosphere at sea level at 45° N is 1013.25 hPa. It is a direct consequence of the weight of the air above the considered air parcel. As a result, pressure decreases with altitude, and can be calculated considering that the gravitational force $F_g = -M_{air} g$ has to be balanced by the vertical pressure gradient force $F_p = dp/dz \cdot V$ [27]:

$$dp/dz = -\rho g \quad (2.4)$$

Integrating this hydrostatic equation, and using the ideal gas law (eq. 2.3), yields the pressure at a given altitude $p(z)$ relative to the pressure p_0 at altitude $z = 0$:

$$p(z) = p_0 \cdot e^{-z/H} \quad (2.5)$$

with $H(z) = RT(z)/(M_{air} g)$ the pressure scale height, and $R = 8.314 \text{ J mol}^{-1} \text{ K}^{-1}$ the universal gas constant. Thus, the atmospheric pressure decreases exponentially with altitude.

Potential temperature and lapse rate If an air parcel rises in the troposphere, it typically expands due to the decreasing pressure profile and cools. Assuming the air parcel is isolated and does not exchange energy with its surroundings, i.e., the process is adiabatic, one can define the potential temperature

$$\theta = T \left(\frac{p_0}{p} \right)^{R/c_p} \quad (2.6)$$

that is conserved during this motion. In other words, the potential temperature represents the temperature a parcel would have if you would bring it adiabatically to the surface or a reference pressure. Using the ideal gas law and the hydrostatic equation and assuming the potential temperature to be constant in the atmosphere, eq. 2.6 can be transformed to:

$$-\frac{dT}{dz} = \frac{g}{c_p} = \Gamma_{ad} = 9.8 \quad (2.7)$$

Γ_{ad} is the dry adiabatic lapse rate, and represents the temperature change of an air parcel if it adiabatically rises or falls, given the potential temperature in the atmosphere was constant. However, in reality θ is not constant, and the actual atmospheric lapse rate in the troposphere averages to $\Gamma_{atmo} = 6.5 \text{ K km}^{-1}$.

Atmospheric stability The relation between the adiabatic lapse rate and the atmospheric lapse rate is essential for atmospheric stability [27]. If $\Gamma_{atmo} < \Gamma_{ad}$, the atmosphere can be considered stably stratified, as rising particles cool faster than the surrounding temperature and face a downward force. This is equivalent to increasing potential temperature with altitude. In contrast, if potential temperature decreases with altitude and $\Gamma_{atmo} > \Gamma_{ad}$, rising air parcels cool slower than the surroundings and the air parcel continues to rise. This phenomenon is known as convection. If the rising air cools enough to reach the saturation mixing ratio for water vapour, the water condenses, releasing latent heat. This process additionally warms the air parcel, reducing its lapse rate, and enables deep convective events, reaching the upper troposphere.

2.1.3 Dynamics & General circulation

The general circulation of the atmosphere is mainly driven by the uneven heating of Earth's surface by the sun, which creates pressure differences [22, 24, 15]. These pressure differences lead to large-scale air movements, in general from high- to low-pressure regions. However, large-scale air movements in the atmosphere are influenced by the Coriolis force, a result of Earth's rotation. This force deflects horizontally moving air to the right in the Northern Hemisphere and to the left in the Southern Hemisphere. The combination of the pressure-driven large-scale air movements and the Coriolis force shape wind patterns and large-scale circulation cells in the Earth's atmosphere.

One such circulation system is the Hadley Cell, which spans both hemispheres from the equator to roughly 30° latitude. In this region, warm air near the equator rises due to intense solar heating, creating strong convective events. Particularly in the Intertropical Convergence Zone (ITCZ), a region of low pressure close to the equator, this rising air causes clouds and thunderstorms to form. The raised air in the ITCZ moves poleward, cools, and sinks at higher latitudes (around 30°), creating a belt of high pressure known as the subtropical highs, before flowing back toward the equator as the trade winds. In addition to the Hadley Cell, the Ferrel and Polar Cell build at higher latitudes, with similar origins, leading to another hotspot of convection around 60° latitude. These cells, in combination with the Coriolis force lead to the formation of the subtropical jetstream and the polar jet, strong zonal wind patterns in the free and upper troposphere.

Atmospheric waves are another important driver of the general circulation [27]. Planetary waves, also known as Rossby waves, are driven by the rotation of the Earth and the variation of the Coriolis force with latitude. Rossby waves oscillate horizontally, especially in mid-latitudes, leading to the meandering of the jetstream, and strongly influencing global weather patterns. In addition, vertical displacements of air, e.g., due to air passing mountain ranges, can lead to vertical gravity waves. Both types of waves can propagate into the stratosphere, where their breaking strongly influences stratospheric dynamics.

This overall general circulation leads to distinct patterns in different layers of the lower atmosphere. These patterns are summarised in the following.

Boundary Layer (BL) The BL is the lowest part of the atmosphere, typically extending up to 1 to 3 km [28, 29]. The actual height can vary strongly due to local conditions, e.g., time of day, local weather and geographic features. The BL is directly influenced by the

Earth’s surface, and thus its dynamics are driven by friction, turbulence, and surface heat exchanges. During the day, the solar heating of Earth’s surface leads to rising warm air and creates a well-mixed BL, with respect to heat, moisture and atmospheric constituents. When the air cools at night, the BL becomes more stable and turbulence is reduced. In general, wind speeds are slowed down by Earth’s friction, so that large-scale movements are suppressed in the BL.

Free Troposphere (FT) The FT starts above the BL and extends to around 8–10 km [29]. The lack of surface friction leads to higher wind speeds compared to the BL, especially in regions dominated by the subtropical and polar jet. Winds approximately follow the geostrophic balance, where the Coriolis force and the pressure gradient force are in equilibrium. As a consequence, the FT is better-mixed zonally with regards to its composition. BL air can enter the FT in convective events, potentially leading to cumulus and cumulonimbus clouds.

Upper Troposphere (UT) & Lowermost Stratosphere (LS) The UT & LS (UTLS) region is defined as the atmospheric layer where isentropic potential temperature surfaces intersect the tropopause, typically spanning altitudes from 8 to 18 km [30, 31]. This region includes the upper troposphere at lower latitudes and the lowermost stratosphere at higher latitudes. It is characterized by complex interactions between tropospheric and stratospheric processes, exchanging air, energy and atmospheric constituents between the troposphere and the stratosphere. The Tropical Tropopause Layer (TTL) is the main source region where tropospheric air, containing long-lived source gases, enters the stratosphere [32]. BL air reaches the TTL through deep convective events. Subsequent transport into the stratosphere occurs either quasi-isentropically into the extratropical lowermost stratosphere, or cross-isentropically into the tropical stratosphere by slow radiatively driven ascent [33]. It is important to note that meridional (latitudinal) transport in the UTLS is inhibited by transport barriers associated with the subtropical jet, creating a “tropical pipe”, and the polar jet [34, 30, 31]. Conversely, transport from the stratosphere to the troposphere also occurs in the UTLS, either by sedimentation from the extratropical LS into the FT, or tropopause foldings, resulting in deep intrusions of stratospheric air into the troposphere [33].

Stratospheric Overworld Above the tropopause, atmospheric temperatures increase with altitude [31]. As a result, the stratosphere is stably stratified, in contrast to the troposphere, and strongly suppresses vertical mixing. Convective events usually stop at the tropopause, and vertical movements are reduced in the stratosphere. The general stratospheric circulation is driven by the Brewer-Dobson circulation [31, 35], which consists of slow rising motion of air in the tropics, poleward transport in the upper stratosphere, and descending motion at higher latitudes. It distributes chemical species in the atmosphere around the globe and influences the temperature structure of the stratosphere. The zonal transport in the tropical stratosphere is dominated by the Quasi-Biennial Oscillation (QBO), an oscillating zonal wind pattern that leads to changing zonal winds from westerlies to easterlies with a mean period of approximately 28 months [36].

2.2 Atmospheric aerosols

The atmosphere consists of chemical species, that can exist in gaseous, liquid, or solid phase. These phases can potentially transition into each other under certain atmospheric conditions. Liquid droplets and solid particles suspended in the atmosphere are collectively referred to as aerosols [13, 15]. Their properties significantly differ from gas phase species.

2.2.1 Sources of atmospheric aerosol

Aerosols in the atmosphere originate from a wide range of natural and anthropogenic sources [13]. These sources can be broadly categorized into primary and secondary aerosols. Primary aerosols are directly emitted into the atmosphere as solid or liquid particles, while secondary aerosols are formed in the atmosphere through gas-to-particle conversion. Thus, aerosol sources combine the emission of solid, liquid and gas phase species.

The vast majority of natural aerosol emissions stem from the following sources:

- **Desert Dust:** One of the largest natural sources of aerosols, desert dust, originates from arid and semi-arid regions, where wind erosion lifts fine mineral particles into the atmosphere.
- **Sea spray:** Aerosols generated by the ocean come from the breaking of waves and the bursting of bubbles at the sea surface, which injects sea spray particles into the atmosphere, mostly consisting of salt, with additions of biological and organic material.
- **Volcanic Eruptions:** Volcanic eruptions inject large quantities of ash and gases like sulfur dioxide (SO_2) into the atmosphere, which can lead to the formation of sulfate aerosols. Smaller eruptions or degassing volcanoes only emit into the troposphere with mostly local impacts, while explosive eruptions potentially directly inject ash and SO_2 into the stratosphere with global impacts.
- **Biogenic Sources:** Natural emissions from plants, such as volatile organic compounds (VOCs) like isoprene and terpenes, can oxidize in the atmosphere to form lower volatile organic species, that can eventually form secondary organic aerosols (SOA). In addition, the terrestrial biosphere also emits biological (such as insect debris, pollen, spores, bacteria and viruses) or Primary Organic Aerosols (POA) into the atmosphere.
- **Wildfires/Biomass Burning:** Biomass burning from wildfires releases large amounts of primary aerosols, including black carbon (soot) and organic carbon, along with precursor gases that can form secondary aerosols. The global influence depends on the strength of the wildfire, being either limited to the troposphere or directly injected into the stratosphere in pyrocumulonimbus clouds. A substantial part of biomass burning emissions can also be classified as anthropogenic origin.
- **Meteoric material:** Small particles from meteors entering Earth's atmosphere contribute to the aerosol burden, particularly in the upper and middle atmosphere [37]. While being a major source of upper stratospheric aerosol, it can be mostly neglected in the troposphere.

Anthropogenic aerosol emissions mostly originate from the following sources:

- **Industrial processes:** Combustion of fossil fuels in power plants, factories, and other industrial processes releases black carbon and POA directly, and precursor gases like sulfur dioxide (SO_2), nitrogen oxides (NO_x) and VOCs, which form sulfate, nitrate and organic aerosols in the atmosphere.
- **Transportation:** Motor vehicles, ships, and aircrafts emit large quantities of aerosols and aerosol precursors. Black carbon (soot) and organic compounds from incomplete combustion of fuels are significant contributors, especially in urban areas and along highways. Again, precursor gases are emitted alongside, potentially forming secondary aerosol.
- **Agriculture:** Agricultural activities, including tilling, harvesting, and burning of crop residues, release among others dust and organic particles into the atmosphere. Livestock farming and the use of fertilizers additionally emit ammonia (NH_3), which can eventually form ammonium aerosol in the presence of acids.
- **Residential combustion:** The burning of biomass, coal, and wood for heating and cooking in many parts of the world is a significant source of primary aerosols like black carbon and organic matter, as well as aerosol precursors.

2.2.2 General properties of aerosol populations

Aerosol populations exhibit distinct properties that vary throughout the atmosphere. These properties can be generally classified by factors such as chemical composition, total mass and number concentrations, size distribution, and interactions with radiation [13].

Aerosol composition A single particle that has travelled through the atmosphere for some time will never be composed of a single pure chemical species. Instead, each particle is a mixture of various species. Moreover, when considering an aerosol population within an air parcel, the composition becomes even more heterogeneous. The composition of an aerosol population at any given time typically reflects the sources and ageing of the particles throughout its journey.

Chemical species in the atmosphere can generally be classified into three categories. First, there are species that exist in the solid phase under all atmospheric conditions, such as desert dust, volcanic ash, meteoric material, and black carbon. Second, some species remain in the gas phase regardless of atmospheric conditions and are not directly relevant to aerosol populations. The third category is the most complex, consisting of low- or semi-volatile species that can exist in the gas, liquid, or particle phase depending on atmospheric conditions. This category includes substances (in the aerosol phase) such as sulfate, ammonium, nitrate, organic aerosols, salts, and metals. These species can undergo phase changes like condensation, evaporation, or crystallization throughout the atmosphere.

Mass and Number concentration One way to describe an aerosol population, is to simply measure total mass and number of aerosol particles within a specific reference volume, known as mass concentration and particle number concentration (PNC). Typically, mass concentrations are reported as $\mu\text{g m}^{-3}$ or ng m^{-3} , while number concentrations are reported as cm^{-3} . In addition, it has proven useful to provide mass and number concentrations only in specific size regions for specific applications. Mass concentrations are often given for particles up to a specific aerodynamic diameter, either $1\ \mu\text{m}$ (PM_1), $2.5\ \mu\text{m}$ ($\text{PM}_{2.5}$) or $10\ \mu\text{m}$

(PM₁₀). PNC is typically further classified into UFP, i.e., number concentration of particles with an aerodynamic diameter below 100 nm.

Size distribution Number and mass concentrations of aerosol populations are linked through the size distribution of the particles. Within a given aerosol population, particles vary in size rather than having a uniform diameter. Freshly formed particles typically range from 1 to 3 nm, while primary particles from desert dust and sea salt can reach sizes of tens of micrometers. Thus, aerosol populations are often characterized by a size distribution, covering the range from 1 nm to 100 μm . These size distributions are typically expressed using differentials dN/dD_p or $dN/d\log(D_p)$, where D_p is the particle diameter.

Ambient aerosols size distributions are typically divided into four log-normal modes. The *nucleation mode* covers freshly formed particles (see Sect. 2.2.3) up to around 20 nm, the *Aitken mode* includes grown nucleation particles and fresh combustion particles between approximately 20 and 100 nm, in the *accumulation mode* particles accumulate due to specific processes (see Sect. 2.2.3) and can grow to micrometer-size, and the *coarse mode* typically contains fresh or aged dust and sea salt particles. Small particles in the nucleation and Aitken modes typically dominate number concentrations, whereas medium-sized particles in the accumulation mode generally dominate mass concentrations.

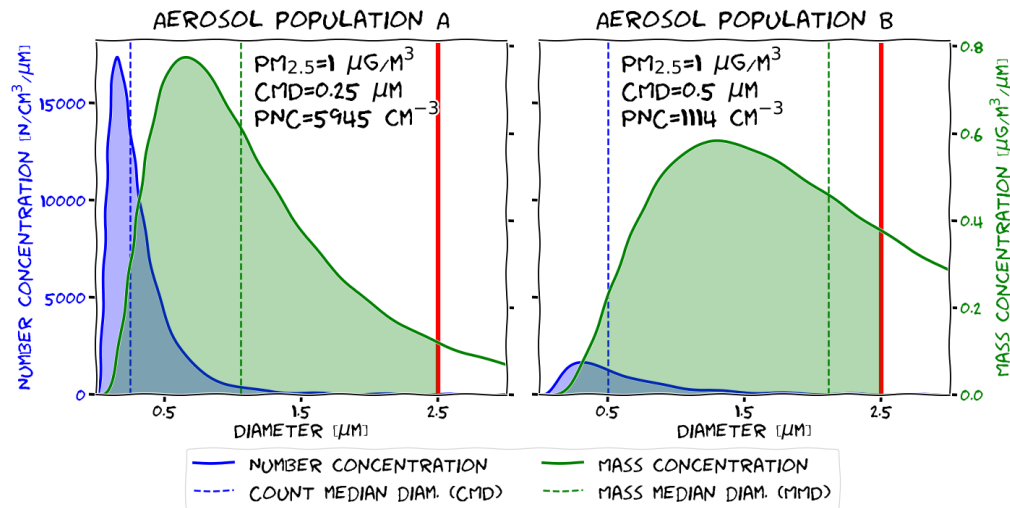


Figure 2.1 Number and PM_{2.5} mass concentration of two aerosol populations with differing size distributions. Both aerosol population have identical PM_{2.5}, despite having differing count median diameter (CMD) and vastly differing PNC. This illustrates the importance of the size distribution in characterizing aerosol populations. The figure is self-created.

Knowledge of the size distributions is crucial in atmospheric processes of aerosols. Merely knowing the number and mass concentrations is insufficient to deduce the size distribution. Figure 2.1 exemplarily illustrates, how log-normal size distributions (identical aerosol density) of two aerosol populations with differing total number concentrations and count median diameter (CMD) can lead to identical mass concentrations. Both aerosol populations show the same PM_{2.5} concentrations of $1 \mu\text{g m}^{-3}$ (integral over green area); however aerosol population A has a CMD of $0.25 \mu\text{m}$ (left panel), while aerosol population B has a CMD of $0.5 \mu\text{m}$. As a consequence, the number concentration of aerosol population A is more

than five times greater (integral over blue area), leading to very different behaviours in the atmosphere.

Interaction with radiation As outlined in Sect. 2.1.1, if a radiation beam is incident on a single aerosol particle, it is either scattered or absorbed [13, 15]. The sum of scattering and absorption as the amount of radiation removed from the beam, is called extinction, and depends on the complex refractive index of the particle. Defining the extinction coefficient σ_{ext} , one can express the Aerosol Optical Depth (AOD) τ of an atmospheric layer ranging from altitude h_0 to h_1 as the integral over the instantaneous extinction at each altitude within the layer:

$$\tau = \int_{h_0}^{h_1} \sigma_{ext}(z) dz \quad (2.8)$$

Beer's Law states that the intensity of the radiation I decreases with $dI = -I\tau$. Thus, after passing an atmospheric layer with the AOD τ , the initial light intensity I_0 will reduce to:

$$I = I_0 \cdot e^{-\tau} \quad (2.9)$$

Thus, a value of $\sigma_{ext} = 1 \text{ km}^{-1}$ can be understood as a weakening of the initial intensity I_0 to $e^{-1} \cdot I_0$ after passing an atmospheric layer of 1 km. AOD can be defined with respect to different atmospheric domains, e.g., total AOD from the surface to the top of the atmosphere or stratospheric AOD (sAOD), only covering the stratosphere.

Absorption and scattering of light by a spherical particle can be described by Mie theory (e.g., [38, 39]), which mostly depends on the wavelength of the radiation, the aerosol size and the aerosol optical properties, strongly depending on its chemical composition and size. By interacting with radiation, aerosols influence the Earth's radiative balance, and can locally warm or cool the atmosphere.

Aerosols & clouds In an atmosphere without aerosols, clouds would only form at very high supersaturations. Thus, aerosols are essential for the formation of clouds in Earth's atmosphere [40, 9, 15]. They can act as cloud condensation nuclei (CCN) or ice-nucleating particles (INP), initiating cloud formation and freezing of liquid water droplets. The ability of aerosols to act as CCN or INP strongly depends on their composition and size, due to varying hygroscopicity and microphysical properties. Thus, the number of CCN and INP are always defined with regard to a specific supersaturation.

In general, at most atmospheric conditions enough CCN or INP are available to form clouds. However, the number of CCN and INP can significantly alter cloud properties [7]. The more CCN are available, the smaller cloud particles form, reflecting more solar radiation (Twomey effect [41]). As a consequence, the cloud appears lighter. In addition, smaller cloud particles lead to drizzle suppression (and thereby increased liquid water content), increased cloud height [42] and longer cloud lifetimes [43].

2.2.3 Atmospheric processes

Once in the atmosphere, aerosols or their precursor gases undergo a number of typical processes, leading to an ageing of the particles [13, 15].

Condensation Condensation in the atmosphere occurs when gas-phase molecules, such as water vapour or low-volatility compounds like sulfuric acid or oxidized organic compounds, transition into the liquid or solid phase on the surface of existing aerosol particles. The rate of condensation depends on the temperature-dependent vapour pressure of the gas phase species relative to its saturation vapour pressure, and the radius and hygroscopicity of the available particles. This process increases the size and mass of the particles, while leaving their number unchanged.

Coagulation Coagulation describes the process of agglomeration of colliding particles to form larger particles. It is typically a combination of Brownian motion, where random particle movements occur due to thermal energy and available aerosol surfaces for collisions. As Brownian velocities decrease with size, coagulation is most efficient for fast-moving small particles with large particles that provide the necessary surface area. This process decreases the number of particles in the atmosphere, and shifts the size distribution upwards. The total mass of the aerosol population does not change.

New particle formation (NPF) In the absence of significant condensational sinks, molecules from very low-volatile vapours can form small clusters, that either evaporate again, or grow via condensation to a critical size. Once, this critical size is reached, new particles are formed that potentially grow. This process is also called nucleation. For a long time, it was believed that sulfuric acid (H_2SO_4) is the solely responsible driver for new particle formation. However, observations showed that other chemical species might also be involved. In fact, the CLOUD experiment at CERN [44, 45] has found a wide range of additional contributors to NPF, such as ammonia (NH_3), ultra-low volatile organics compounds (ULVOCs), nitric acid (HNO_3), and iodic acid (HIO_3). Figure 2.2 gives an overview over a number of observed particle formation channels in the CLOUD experiment, taken from Kirkby et al. (2023) [45]. While NPF can result in significant local increases in PNC, the direct effect on the overall mass of the aerosol population is limited due to the initially small size of these particles. However, newly formed particles can provide surfaces for condensation and grow to CCN size, thereby impacting cloud properties and indirectly contributing to the mass of the aerosol population.

Atmospheric transport Atmospheric aerosols are primarily transported by large-scale atmospheric circulation patterns and local turbulences. The transport efficiency strongly depends on the size of the particles. In the BL, turbulence results in a well-mixed chemical composition, while larger-scale transport occurs via advection. Convection plays a key role in vertically transporting aerosols and their precursors to the FT and UT. In the stratosphere, aerosols are distributed by the Brewer-Dobson circulation and strong zonal winds. These processes distribute aerosols all around the globe.

Sedimentation Aerosols in the atmosphere are subject to the Earth's gravitational field. Thus, they gravitationally settle (sediment) depending on their size, shape, and density. Larger particles sediment faster than smaller ones. In the troposphere, sedimentation is only relevant for coarse-mode particles, as the aerosol lifetime is short compared to sedimentation velocities for smaller particles. However, in the absence of other removal processes in the stratosphere, sedimentation plays an important role in redistributing aerosols of all sizes and transporting them from the stratosphere to the troposphere.

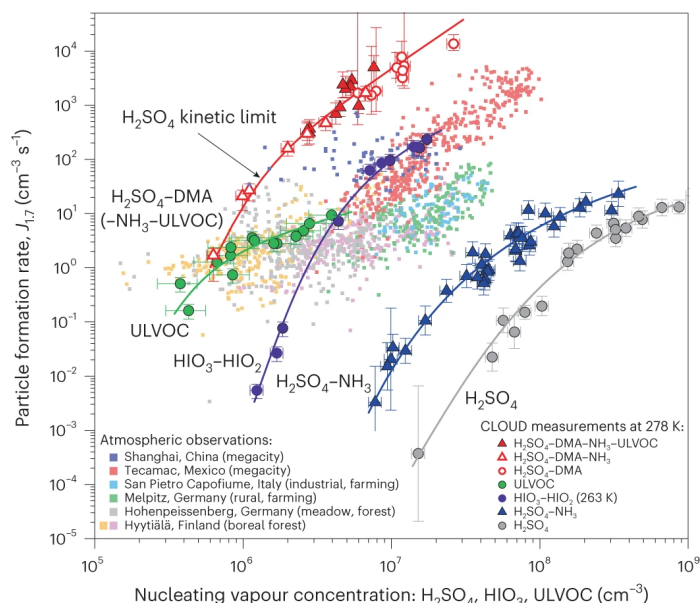


Figure 2.2 Selected nucleation channels observed at the cloud experiment. Displayed are particle formation rates, measured at different conditions in the CLOUD chamber with varying vapour concentrations, compared to atmospheric observations. Figure taken from Kirkby et al. (2023) [45].

Heterogeneous and multiphase chemistry Chemical reactions can be facilitated inside liquid aerosols or on the surface of liquid and solid aerosols, referred to as multiphase or heterogeneous reactions, respectively [46, 47, 48]. They depend on the collision frequency between gas phase molecules and aerosol, determined by the aerosol surface area and atmospheric pressure, the uptake efficiency of the particle and the reaction rate itself. Via heterogeneous chemistry aerosols influence gas phase chemistry. Multiphase reactions in cloud droplets can lead to the formation of sulfuric and nitric acid in acid rain [47]. In the stratosphere, the production and loss of HO_x and NO_x , as well as the activation of halogen species on aerosol surfaces (especially on polar stratospheric clouds forming in the polar winter at low temperatures) can affect the ozone layer, leading to a depletion of stratospheric ozone [10, 11] (more details in Sect. 2.6).

2.2.4 Chemistry of aerosol precursor gases

Secondary aerosol forms through the conversion of gas phase species to the solid or liquid phase. The main quantity determining the efficiency of secondary aerosol formation is the volatility of the gas phase species, i.e., the tendency of condensed species to evaporate, respectively the tendency of gas phase species to remain in the gas phase [49]. High-volatile gas phase species tend to stay in the gas phase, while low-volatile species prefer to transition to the liquid or particle phase. Volatility is usually provided as saturation vapor pressure or saturation mass concentrations, i.e., the amount of the substance in the gas phase in equilibrium with the condensed phase.

In general, ageing of gaseous compounds, specifically oxidation processes by reaction with the OH radical, O_3 , NO_3 or halogens, decrease their volatility. Nitrogen- and sulfur-containing

species mostly end up as nitric and sulfuric acid upon oxidation, with a higher oxidation state and much lower volatility than the original species. Nitric and sulfuric acid can readily dissolve in water droplets, and form sulfate and nitrate aerosol. In the presence of ammonia, the salts ammonium sulfate and ammonium nitrate can form, while ammonium sulfate is preferred if sufficient sulfuric acid is available [15].

A large number of different VOCs are emitted from natural and anthropogenic processes. Upon different oxidation pathways they can form thousands of organic species with differing volatility [50]. It is analytically and computationally not feasible to keep track of all these species. To account for this complexity, Volatility Basis Sets (VBS) were introduced [51], providing logarithmically equally spaced saturation bins, covering several orders of magnitudes in which organic tracers are lumped for the calculations. A simple approximation of the saturation mass concentration C^0 in $\mu\text{g m}^{-3}$ of organic species can be derived by only considering the number of carbon (n_C) and oxygen (n_O),

$$\log_{10} C^0 = (n_C^0 - n_C)b_c - n_O b_O - 2 \frac{n_C n_O}{n_C + n_O} b_{CO}, \quad (2.10)$$

with the coefficients $n_C^0 = 25$ (carbon number of pentacosane -C_{25} [51]), $b_C = 0.475$, $b_O = 2.3$ and $b_{CO} = -0.3$. This expression can be extended to additionally contain nitrogen and sulfur species, that influence the volatility of the organic compound [52]. Based on the value of $C^* = \gamma C^0$ with γ the activity coefficient, organic compounds can then be classified into different volatility regimes [53]:

- **Extremely Low-Volatility Organic Compounds (ELVOC, $C^* < 3.2 \times 10^{-4} \mu\text{g m}^{-3}$)** strongly tend to the condensed phase at all atmospheric conditions, and can potentially form new particles in the absence of condensational sinks.
- **Low-Volatility Organic Compounds (LVOC, $3.2 \times 10^{-4} < C^* < 0.32 \mu\text{g m}^{-3}$)** tend to the condensed phase under most atmospheric conditions.
- **SemiVolatile Organic Compounds (SVOC, $0.32 < C^* < 320 \mu\text{g m}^{-3}$)** can either reside in the gas or condensed phase at typical ambient conditions
- **Intermediate Volatility Organic Compounds (IVOC, $320 < C^* < 3.2 \times 10^6 \mu\text{g m}^{-3}$)** reside in the gas phase under typical atmospheric conditions, but can be oxidized to form organic compounds with lower volatility.
- **Volatile Organic Compounds (VOC, $C^* > 3.2 \times 10^6 \mu\text{g m}^{-3}$)** reside in the gas phase under all ambient atmospheric conditions. Freshly emitted gas phase organics are usually VOCs, but can be oxidized to form lower volatile compounds.

2.2.5 Aerosol removal

At some stage in the lifecycle of an atmospheric aerosol particle, it is removed from the atmosphere. In general, this occurs through two main processes: wet deposition and dry deposition [13].

Wet deposition, also referred to as scavenging, is the primary removal pathway for soluble species in the troposphere. During cloud formation, aerosol particles can dissolve into

the cloud droplets. If the cloud precipitates, these aerosol particles are removed from the atmosphere through in-cloud scavenging. As raindrops fall, they may collide with particles in the vertical column below the cloud, and the particles can dissolve in the droplet. This is referred to as impact scavenging. However, these process does not necessarily lead to a removal of particles. Under certain conditions, raindrops can re-evaporate at lower altitudes, releasing the scavenged aerosol particles back into the atmosphere, leading to a redistribution of the particles rather than their removal.

Dry deposition is only relevant close to surfaces, and describes the direct removal of particles by collision with non-atmospheric surfaces, such as the Earth's surface, trees, or buildings. Dry deposition can be realised via three different mechanisms. First, aerosol particles can be deposited to the Earth's surface due to turbulence. Second, large particles can directly sediment onto the Earth's surface. And third, aerosols can collide with obstacles above the Earth's surface. Dry deposition can be neglected outside the BL. However, sedimentation can transport coarse particles into the BL, where they are subject to dry deposition.

Overall, these removal processes in combination with the aerosol size determine aerosol lifetime across the atmospheric layers. In the BL, the aerosol lifetime is rather short, mostly below one week, as particles are subject to dry and wet deposition. Once in the free and upper troposphere, aerosol lifetime can reach several months, depending on relative humidity and cloud formation. Aerosols reaching the stratosphere, can reside there two years or more, depending on latitude and altitude. The only significant removal process in the stratosphere is sedimentation back into the troposphere, which can be slowed down by radiative heating and subsequent uplift.

2.3 Observations of atmospheric aerosols

Atmospheric aerosols were first recognized through observations of natural phenomena like haze, fog, and volcanic eruptions. In the late 19th century, John Aitken built the first instrument to measure the number of particles in the atmosphere [54, 55]. This laid the foundation for in-situ measurements of aerosols, directly capturing and investigating aerosol populations. During a similar time, Lord Rayleigh (John William Strutt) [56, 57] worked on the understanding of aerosol optical properties by studying scattering and absorption of light by particles, providing the basis for remote sensing of atmospheric aerosols, i.e., characterizing aerosol populations by their interaction with light.

2.3.1 In-situ measurements

In-situ aerosol measurements provide detailed information on aerosol populations by directly sampling the air at the measurement location [13]. Meanwhile, a wide range of instruments has been developed, serving different purposes. They are mostly used to measure aerosol parameters such as total mass, chemical composition, particle number concentration, particle size distribution, and optical properties.

In-situ measurements can be performed in various settings. Long-term measurements are mostly performed at the Earth's surface on fixed measurement sites. Measurements can also be performed in moving platforms on the surface, such as ships or cars. Additionally,

aircrafts and balloons can be equipped with in-situ measurement devices to sample air in the FT, UTLS and the stratosphere.

2.3.2 Remote sensing

Measurements that are performed at some distance to the actually measured aerosol populations are referred to as remote sensing [13]. The information is retrieved by electromagnetic waves, i.e., radiation. If this electromagnetic radiation is emitted by a natural source (short-wave radiation from the sun or long-wave radiation from the Earth), and the instrument solely measures the scattered radiation, the remote sensing is referred to as passive. Active remote sensing instrument emit radiation and measure the back-scattered fraction of it.

Passive remote sensing can be realized using different techniques and geometries. Extinction can either be measured from the ground (solar radiation) providing information on the column composition or from space, using occultation methods, observing radiation from the sun, the moon or stars through the Earth's atmosphere, providing 3-dimensional information on the atmosphere's composition. Other instruments measure scattering or direct emission of solar or infrared radiation in different geometries, i.e., at the limb (the edge of the atmosphere with satellites), in nadir geometry (satellites observing vertically down), or from the Earth's surface.

Active remote sensing is mostly realised via lidar measurements, emitting short laser pulses, and observing the back-scattered fraction. Lidar observations can be performed from the surface, satellites, aircrafts or balloons.

2.4 Modelling of atmospheric aerosols

2.4.1 Atmospheric modelling

In 1904, Vilhelm Bjerknes formulated the idea that weather could be predicted by solving a set of fundamental physical equations [58], including the Navier-Stokes equations, which describe fluid motion, and equations of thermodynamics and mass conservation. This idea is the foundation of all atmospheric modelling. Up to this day, most atmospheric models still rely on the solution of these fundamental equations. However, the computational effort to solve these equations were impractical at that time. Lewis Fry Richardson first attempted to perform these calculations in 1922 [59], however unsuccessful. As a consequence, he envisioned a “forecast factory” of human calculators to collectively perform weather forecasts.

With the development of non-human computers between 1940 and 1950, first numerical weather predictions became possible, applying strict simplifications [60]. From this start, the atmospheric model development accelerated. In the 1960s, the first General Circulation Models (GCMs) appeared, based on fundamental physical laws, as envisioned by Vilhelm Bjerknes. They represented the atmosphere in simplified forms, usually in two-dimensional models, but included important concepts like heat transfer, the hydrological cycle, and radiation. This allowed for first basic climate simulations. In the 1970s, three-dimensional GCMs arose with increasing complexity, higher resolution and better representation of processes like cloud formation, ocean circulation, and interactions between the atmosphere

and the biosphere.

In the following decades, more and more chemical processes were added to atmospheric models, resulting in the creation of Chemistry Climate Models (CCMs) [60]. The combination of atmospheric dynamics with chemical reactions allowed for simulations of phenomena like ozone depletion, aerosol-cloud interactions and the greenhouse effect, and thus leading to a better understanding of air pollution, greenhouse gases, and their impact on climate. Since the early 2000s, Earth System Models arose, coupling the atmosphere with other Earth components, such as oceans, land surfaces, ice sheets, and ecosystems, helping to understand feedbacks within the Earth System.

Today, a large number of atmospheric models exist, that serve a lot of different purposes. Box models help understanding specific processes with fixed boundary condition, 2D- models enable the study of transport processes within the atmosphere. 3D-models can represent the global or regional atmosphere from the Earth’s surface to the Upper Atmosphere. Eulerian models simulate the state of Earth’s atmosphere at fixed coordinates, while Lagrangian models follow individual air parcels. Chemical processes and additional Earth components can be included for specific studies. However, atmospheric models are among the most complex model systems, and thus are still limited by computational power. As a consequence, there always has to be a trade-off between scientific detail and computational cost, especially with regards to long-term simulations.

2.4.2 EMAC model

The numerical simulations presented in this thesis have been produced using the ECHAM5/MESSy2 Atmospheric Chemistry Model (EMAC) [61, 62]. MESSy [63] stands for Modular Earth Submodel System, which enables the coupling of a large number of physical, chemical, and diagnostic submodels to different GCMs. Specifically, for this work, the GCM ECHAM5 has been used as the base model. ECHAM5 is a further development of the ECMWF operational forecast model cycle 36 (EC), combined with a comprehensive parameterization developed in Hamburg (HAM) [64]. The MESSy infrastructure allows for flexible atmospheric simulations with varying complexity, depending on the submodels used and their configuration.

EMAC simulations can be performed at different resolutions, both horizontally and vertically. The dynamical calculations in ECHAM5 are carried out in spectral space to reduce computational costs. Consequently, the horizontal resolution is defined by the triangular truncation of the waves. Most simulations in this thesis are performed at T63 horizontal resolution, corresponding to $1.875 \times 1.875^\circ$ grid box size, or T106 ($1.1 \times 1.1^\circ$) and T255 ($0.47 \times 0.47^\circ$) for short-term high-resolution studies. In tropospheric simulations, 31 vertical levels are used up to a height of approximately 30 km or 10 hPa, while simulations including the stratosphere use 90 vertical levels, extending up to 0.01 hPa (or 80 km).

The MESSy infrastructure consists of four layers of different abstraction: 1) the Base Model Core Layer (**BMCL**), which includes the base model (in this case ECHAM5) and is responsible for simulating the general circulation, with implemented calls to 2) the base model interface layer (**BMIL**), which manages the activation of and communication with the various submodels, thus responsible for the MESSy infrastructure, 3) the SubModel Interface Layer

(**SMIL**), which handles the submodel communication with the base model and the other submodels, and 4) the SubModel Core Layer (**SMCL**), which contains the core routines of the submodels and is independent of both the base model and other submodels.

Within the BMIL, a range of submodels manages the model infrastructure, strictly separated from process-related and diagnostic submodels. These generic submodels are active in all simulations and cannot be deactivated. The **SWITCH** submodel enables the user to select specific submodels for a simulation, allowing for flexibility and control of resource usage. The chosen submodels are then called by the **CONTROL** submodel. The **CHANNEL** submodel is responsible for the storage, exchange and information of data [62]. All prognostic and diagnostic variables are stored in channel objects, which can have individual attributes, dimensions, and representations. Model output can be controlled through a namelist, determining which channel objects are output and at which time intervals. Additionally, both the base model and all submodels have access to all channel objects in the interface layer, facilitating communication between them.

Information on all trace gases and aerosol species, hereafter referred to as tracers, is handled by the **TRACER** submodel [65]. The total abundance of each tracer is stored separately from the static metadata. The tendencies of all prognostic and diagnostic variables, including tracers, are globally managed by the **TENDENCY** submodel [66], individually accounting for each process' contribution to the total tendency. Tendencies are applied following the operator splitting concept [63], where the individual tendencies of each process are added based on the variable state from the preceding time step, along with all tendencies from previous processes in the sequence.

Initial and boundary conditions are essential for numerical simulations. In MESSy, these can be provided with the **IMPORT** submodel, which allows the specification of input files via namelist. The imported objects can then be used by the respective process submodels. Four-dimensional (space and time) input data is usually provided on a grid differing from the model grid. The **GRID** submodel [67] handles the definition and transformation methods to align different grid specifications. The **TIMER** submodel manages the time control of the simulation, while the **QTIMER** submodel optimizes the use of queue limits and provides runtime diagnostics. Finally, the **TOOLS** submodel [62] offers shared tools for use across submodels.

Process related and diagnostic submodels, distinct from the model infrastructure submodels, can be flexibly activated or deactivated for each simulation. These submodels are called at each timestep to perform prognostic and diagnostic calculations for each grid box. Some process submodels are essential for every chemistry simulation. These submodels will shortly be introduced in the following. Submodels relevant for the simulation of atmospheric aerosols are discussed in the following section, while all other submodels are summarized in Sect. 2.4.4.

Chemical reactions are managed by the **MECCA** submodel [68], which includes predefined chemical mechanisms that can be extended or modified. Photochemical reactions depend on the photolysis rate (denoted as j values) which are computed online by the **JVAL** submodel [69]. Emissions can either be provided using offline emission inventories through the **OFFEMIS** submodel or calculated online via the **ONEMIS** submodel [70]. In this

thesis, sea salt and dust emissions are calculated online, with the wind speed provided by the base model.

2.4.3 Aerosols in EMAC

This thesis primarily focuses on the simulation of atmospheric aerosols. Within the MESSy framework, various submodels are responsible for handling aerosol processes. Aerosol microphysics can be managed by the MADE3, GMXe, or PTRAC submodels for passive tracers. For this work, the aerosol microphysics and gas aerosol partitioning submodel (Global Modal-aerosol eXtension, GMXe) [71] is used, which is based on the M7 aerosol microphysics model [72].

Interactive log-normal size modes form the core of GMXe’s microphysics. Each mode is defined by an upper and lower diameter boundary, and a given constant sigma σ_m (log-normal distribution width). A detailed investigation of these parameters’ influence on global aerosol distributions is presented in Chapter 5. Within each mode, the median diameter D_m can adjust freely in response to the relevant processes. The prognostic variables for each mode are the number of particles (N_m), and the corresponding mass of each constituent. Other diagnostic variables, such as density and diameter, can be derived from these quantities. In general, all species within each mode are assumed to be internally mixed, meaning that particles within a mode have the same composition, determined by the mass of the respective species. However, the modes are externally mixed with respect to each other. Although all particles are assumed to be spherical for computational simplicity, it is important to note that their shapes can vary significantly in reality.

The original GMXe setup includes seven log-normal modes: four soluble (nucleation, Aitken, accumulation, and coarse) and three insoluble (Aitken, accumulation, and coarse). However, the number of modes in GMXe can be adjusted flexibly, while a higher number of aerosol modes results in higher computational costs. The size distribution for each mode, given the particle diameter D_p , follows a log-normal distribution (with width σ_m), which is defined as [15]:

$$\frac{dN_m}{dD_p} = \frac{N_m}{(2\pi)^{1/2} D_p \ln \sigma_m} \exp \left\{ -\frac{(\ln D_p - \ln D_m)^2}{2 \ln^2 \sigma_m} \right\} \quad (2.11)$$

$$\frac{dN_m}{d \log D_p} = D_p \cdot \ln 10 \cdot \frac{dN_m}{dD_p} \quad (2.12)$$

The total PSD of the aerosol population is then the sum of the size distributions of each mode. To quantify PNC within a specific size segment, it is necessary to integrate the size distribution functions of each mode, using the error function:

$$\text{erf}(z) = \frac{2}{\pi} \int_0^z e^{-x^2} dx. \quad (2.13)$$

The total number of particles with a diameter up to D_{max} , e. g., relevant for the calculation of UFP with a diameter up to 100 nm, can then be calculated as follows:

$$N_{\text{cum}}(D_p) = \frac{N_m}{2} \left(1 + \text{erf} \left(\frac{\ln D_{max} - \ln D_m}{\sqrt{2} \ln \sigma_m} \right) \right) \quad (2.14)$$

For comparison with observations, it is important to evaluate the PNC within the same size segment, $N_{\text{frac}}(D_{\text{min}}, D_{\text{max}})$, where D_{min} to D_{max} , correspond to the measurement size limits. This can be calculated by subtracting the two cumulative PNC at the two size boundaries:

$$N_{\text{frac}}(D_{\text{min}}, D_{\text{max}}) = N_{\text{cum}}(D_{\text{max}}) - N_{\text{cum}}(D_{\text{min}}) \quad (2.15)$$

Similarly, the cumulative aerosol mass up to a specific diameter $M_{\text{cum}}(D_p)$ can be calculated by multiplying the cumulative volume $V_{\text{cum}}(D_p)$ by the average density of the mode, ρ_m :

$$V_{\text{cum}}(D_{\text{max}}) = \frac{N_m \pi}{12} \exp \left\{ 3 \ln D_m + \frac{9}{2} \ln^2 \sigma_m \right\} \left(1 + \operatorname{erf} \left(\frac{\ln (D_{\text{max}}/D_m) - 3 \ln^2 \sigma_m}{\sqrt{2} \ln \sigma_m} \right) \right) \quad (2.16)$$

$$M_{\text{cum}}(D_{\text{max}}) = \rho_m \cdot V_{\text{cum}}(D_{\text{max}}) \quad (2.17)$$

The total mass between D_{min} and D_{max} for comparison with observations can be calculated with the approach outlined in Eq. 2.15.

Within GMXe, the following aerosol processes are considered:

- **Primary emissions:** Based on the emission fluxes calculated in OFFEMIS and ONEMIS, primary particles are emitted in GMXe. In addition to the mass flux (M_{emis}), the emission median diameter d_{emis} , aerosol density ρ_{emis} and the emission mode must be provided. Using this information, the number of primary emitted particles N_{emis} that match the mass flux can be calculated.

$$N_{\text{emis}} = \frac{6 \cdot M_{\text{emis}}}{\pi \cdot \rho_{\text{emis}} \cdot d_{\text{emis}}^3} \cdot \exp \left\{ -4.5 \ln^2 \sigma_m \right\} \quad (2.18)$$

σ_m represents the width of the log-normal mode. If the emission diameter is larger (smaller) than the median mode diameter, the median mode diameter will increase (decrease) accordingly.

- **Inorganic aerosol thermodynamics:** GMXe offers several thermodynamic models for calculating gas-aerosol partitioning. In this work, the thermodynamic equilibrium model ISORROPIA-II [73] is used. This model can treat the $\text{K}^+ - \text{Ca}^{2+} - \text{Mg}^{2+} - \text{NH}_4^+ - \text{Na}^+ - \text{SO}_4^{2-} - \text{NO}_3^- - \text{Cl}^- - \text{H}_2\text{O}$ system by identifying the relevant subset of thermodynamic equations and solving for the equilibrium state using chemical potential. As a result, gas phase species can condense on existing particles or liquid species can evaporate, depending on the equilibrium state.
- **Coagulation:** The description of coagulation in GMXe is adopted from the M7 model [72], with the coagulation coefficients calculated for Brownian Motion. During coagulation, the mass of the smaller particle is transferred to the mode of the larger particle, and insoluble particles are transferred to the soluble modes when they collide with soluble particles.
- **Mode redistribution:** The processes described above can cause a shift in the median diameter of the aerosol modes. As a result, after each time step, aerosol mass and number is redistributed between the size modes. The part of the size distribution that

overlaps into neighbouring boundaries is transferred to the next higher or lower bin. This approach prevents unphysical results and effectively simulates observed growth and shrinkage processes.

Additionally, GMXe includes a simplified mechanism for the formation of SOA. However, in this work the submodel ORACLE [74] is used for organic gas-aerosol partitioning. ORACLE introduces a range of organic gas phase species, classified into different volatility bins and source sectors. The volatility classification follows the VBS approach (see Chapter 2.2.4) [51].

While GMXe includes a parameterization for nucleation processes, it relies on older parameterizations of the sulfuric acid-water nucleation system [75]. Therefore, a New Aerosol Nucleation (NAN) submodel [76] was recently developed, providing a flexible interface for implementing new particle formation rates, primarily based on the results of the CLOUD experiment (see Sect. 2.2.3). In standard simulations conducted in this work, the following NPF channels are included: binary $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ [77], ternary $\text{H}_2\text{SO}_4\text{-NH}_3\text{-H}_2\text{O}$ [77], synergistic $\text{H}_2\text{SO}_4\text{-NH}_3\text{-HNO}_3$, pure organic [78], and nucleation from oxidized organic species with sulfuric acid [79]. The CLOUD experiment also found that the presence of ions can enhance NPF [77, 78]. Within NAN, NPF can be distinguished between ion-induced and neutral processes. The formation of ions is calculated using the IONS submodel [76].

As discussed in Sect. 2.2.5, aerosols are removed from the atmosphere through either wet or dry deposition. Wet deposition is handled by the SCAV submodel [80], which differentiates between in-cloud scavenging (mostly dominant for particles with a diameter above $0.1\text{ }\mu\text{m}$) and impact scavenging, that relies on Brownian motion (dominant for small, fast-moving particles). The efficiency of wet deposition depends on the solubility of the aerosol population. Dry deposition of aerosols is managed by the DDEP submodel [81] and occurs only in the lowermost model layer, where the atmosphere interacts with the Earth’s surface. The dry deposition rate depends on several factors, including the aerodynamic resistance of the surface, the particle size, the molar mass of the aerosol relative to dry air, and the thickness of the layer. Larger particles tend to deposit due to the gravitational acceleration, while smaller particles are removed through turbulence. Both SCAV and DDEP also handle the removal of gas phase species. Sedimentation of aerosol particles is treated by the SEDI submodel [81], which primarily depends on the density and size of the particle, and is calculated for all model layers.

AERosol OPTical properties are calculated online with the AEROPT submodel [82] or the new AOP submodel, either based on climatological data or coupled to GMXe using online aerosol populations. These calculations rely on precomputed lookup tables that provide Mie size parameters for each aerosol mode on 32 predefined wavelength bands, 16 in the shortwave and 16 in the longwave spectrum. The output can either be used diagnostically for comparison to satellite observations, or used for radiative transfer calculations via the RAD submodel [82]. RAD is the MESSy implementation of the original ECHAM5 radiation scheme. It uses aerosol and cloud optical properties from AEROPT and CLOUDOPT (described below), along with greenhouse gas concentrations, to simulate radiative transfer processes based on incoming solar radiation.

2.4.4 Additional MESSy submodels

In addition to the MESSy submodels discussed above, this work also uses the following submodels for physical parameterizations and diagnostics:

- **AIRSEA** [83] treats the exchange of gas phase species between the atmosphere and the ocean, i.e., emission and deposition, with a focus on organic species, such as DMS, isoprene and acetic acid.
- **BIOBURN** calculates emissions from Biomass Burning (BB) and Agricultural Waste and residue Burning (AWB) based on observed dry matter burned and fire type [84, 85].
- **CLOUD** is responsible for the calculation of cloud microphysics based on different cloud schemes. Simulations in this thesis use the standard cloud scheme from ECHAM5 [64].
- **CLOUDOPT** [82] calculates cloud optical properties, based on cloud cover, liquid and ice water, and cloud nuclei concentration, that can be used for radiative considerations.
- **CONVECT** [80] handles convective processes based on different convection schemes. In this thesis, the Tiedtke scheme [86] is used.
- **CVTRANS** [87] is responsible for the convective transport of trace gases and aerosols based on the chosen convection scheme (see above)
- **GWAVE** treats non-orographic gravity waves from the original ECHAM5 code [64] based on parameterizations from Hines [88, 89].
- **LNOX** [80] includes parameterizations of NO_x (NO and NO_2) from lightning. This work uses the parameterizations from Grewe et al. [90], scaled to 2.6 Tg N per year.
- **MEGAN** (Model of Emissions of Gases from Nature [91]) estimates biogenic emissions from terrestrial ecosystems.
- **MSBM** simulates the formation of Polar Stratospheric Clouds and handles stratospheric heterogeneous reactions [62].
- **ORBIT** [82] calculates orbital parameters of the Earth around the sun to support calculations on solar radiations incident on Earth.
- **OROGW** includes parameterizations of subgrid scale orography drag based on the original ECHAM5 code [64]
- **QBO** can be used for the nudging of the model to an assimilation of QBO zonal wind observations (as done in this work), or for self-consistent free-running simulations including the QBO.
- **S4D** [62] samples model data along the track of a moving observatory platform, such as aircrafts, balloons or ships. Calculations are performed online, and output can be produced at each timestep.

- **SORBIT** [62] samples model data along sun-synchronous satellite orbits, synchronising satellite overpasses with the simulation time. This facilitates comparison of model calculations with satellite observations.
- **SURFACE** is based on the original ECHAM implementation [64], and calculates properties of the Earth’s surface and the soil.
- **TNUDGE** [70] calculates pseudo-emissions (or sinks) of tracers with unknown emissions, but well-known surface mixing ratios
- **TROPOP** [61] provides the height of the tropopause, which can be defined either according to the WMO criteria based on the lapse rate or using potential vorticity. In addition to computing potential vorticity, it also determines the BL height.

2.5 Ambient aerosol populations

Due to the advancing technologies in both measurement techniques and atmospheric modelling, the knowledge on global ambient aerosol populations has rapidly increased over the last decades. Observations from ground-based stations, aircrafts, balloons, ships and satellites have been complemented with modelling results to shape a picture of global aerosol distributions. Nevertheless, substantial uncertainties remain, particularly due to limited measurements in certain regions of the atmosphere. This section summarizes the current state of knowledge regarding aerosol populations across different atmospheric layers.

2.5.1 BL

Due to surface friction, wind speeds at the Earth’s surface are decelerated, and large-scale transport is suppressed in the BL. Simultaneously, turbulent mixing leads to high dry deposition rates at the Earth’s surface. As a result, aerosol populations in the BL are strongly shaped by local sources and sinks, meteorology and orography, leading to a highly heterogeneous distribution of surface aerosol.

An important measure for aerosol populations is $\text{PM}_{2.5}$ concentration. Numerous studies have combined in-situ measurements, satellite observations and atmospheric modelling [92, 93, 94, 4] to estimate global $\text{PM}_{2.5}$ concentrations, exhibiting hotspots in densely populated regions, such as East and South Asia, and over desert regions (see Fig. 2.3).

Mass spectrometers all over the globe provide valuable insights into the composition of atmospheric aerosols in the BL [95, 96, 97, 98]. While elevated $\text{PM}_{2.5}$ concentrations in desert regions are clearly dominated by mineral dust [99], aerosol composition in other regions of the world is more diverse. In urban areas, POA and SOA, black carbon, and sulfate typically dominate, with notable contributions from nitrate, ammonium, and various metals. The primary source of these particles are combustion and general industrial processes.

Aerosol populations in remote and rural regions are shaped by local land use and long-range transport patterns. Over the ocean, sea salt particles become very important, while organic aerosol is dominating in forest regions. Non-marine arctic and antarctic regions usually

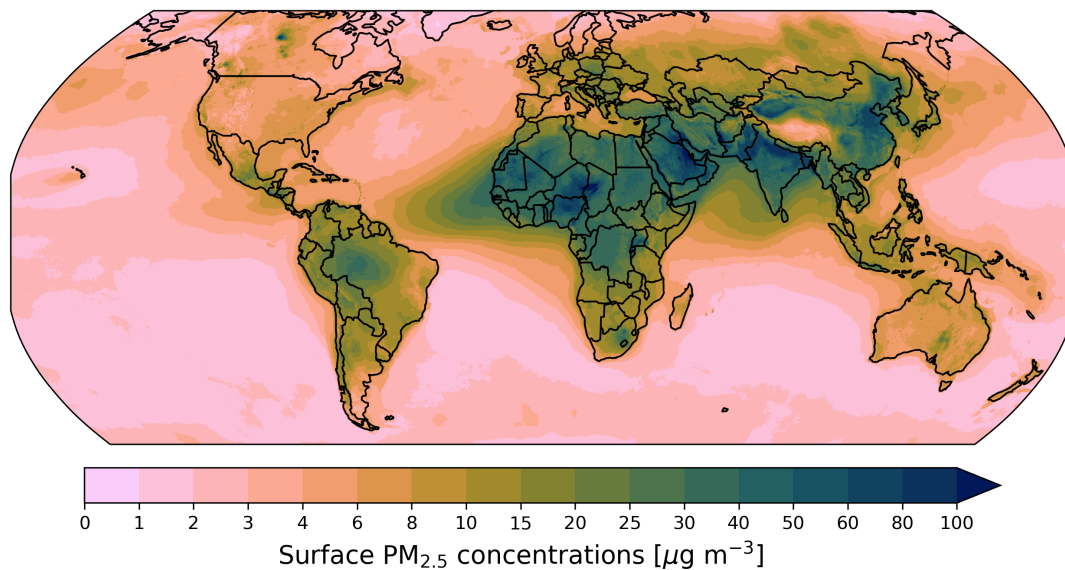


Figure 2.3 Surface PM_{2.5} concentrations, self-plotted based on pseudo-observations from van Donkelaar et al. [92].

exhibit very low PM_{2.5} concentrations, reflecting minimal local emissions and limited long-range transport.

New particle formation in the BL typically occurs under conditions of low condensational sink and high availability of precursor gases, such as low-volatility organic compounds, sulfuric acid, or iodic acid. Gordon et al. (2017) [100] estimated that approximately 50% of surface-level aerosols originate from NPF. PNCs vary by several orders of magnitudes across the BL, with strong hotspots due to local emissions [101], and quick relaxation to background levels with distance from the source due to the short lifetime of freshly formed particles [102]. Numerous studies have provided local high-resolution datasets of PNC and UFP concentrations using land use regression models [101, 103, 104, 105]. However, global-scale information on UFP remains limited.

2.5.2 FT

Knowledge of aerosol populations in the FT is primarily derived from aircraft and balloon observations, as well as stationary measurements at high-altitude mountain sites. Aerosols in the FT are mainly influenced by upward transport from the BL, with transport efficiency depending on particle size, or by direct injections from elevated sources such as industrial stacks, wildfires or volcanic eruptions. In contrast to the BL, the absence of surface friction in the FT allows for more efficient zonal long-range transport, resulting in a more homogenized aerosol distribution relative to local sources. The FT usually contributes less to tropospheric AOD than the BL, with studies finding around 31% of the tropospheric attributable to the FT globally [106] and 13-31% locally over a Chinese city (Wuhan) [107]. This contribution is also influenced by variations in BL height, which differ between day and night.

As aerosol mass and surface concentrations decrease in the FT, the condensational and

coagulation sinks also decrease. Consequently, if precursor gas concentrations are sufficiently high, conditions can favour NPF, as reported in various field campaigns [108, 109, 110]. These precursor gases can originate from the surface [111, 112] transporting sulfuric acid and Highly Oxidized Organic Molecules (HOMs) from urban or forest regions to the FT, from the outflow of convective clouds in marine regions [113, 114], or from the stratosphere via stratospheric intrusions introducing high sulfuric acid concentrations in the FT [115].

2.5.3 Stratosphere (Overworld)

In 1960, Christian Junge and coworkers conducted balloon measurements in the stratosphere to search for meteoric and cosmic dust, as well as debris from nuclear bomb tests. Instead, they primarily found $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ droplets, distributing in the altitude range between 12 and 28 km [116]. Subsequent studies showed that they form a global stratospheric layer, referred to as the Junge layer [117, 118, 119]. The lower boundary of the Junge layer is defined by the tropopause, below which cloud processes significantly limit the lifetime of sulfate aerosols. Above 30 km, stratospheric temperatures rise, leading to the evaporation of sulfate droplets.

The Junge layer is believed to primarily form through natural processes. In 1976, Paul Crutzen proposed that the transport of long-lived carbonyl sulfide (OCS) to the stratosphere, followed by its photochemical destruction, is the dominant source of background stratospheric sulfate [120]. This theory has been supported by model studies, which show that OCS alone is sufficient to sustain the Junge Layer [121, 122], with other short-lived sulfur compounds contributing only minor amounts [122]. Additionally, volcanic eruptions can enhance the Junge Layer by injecting SO_2 directly into the stratosphere, forming sulfate droplets after oxidation. Very strong volcanic events can potentially cause substantial negative radiative forcing, up to -3 to -5 W m^{-2} [123, 124].

While sulfate aerosols dominate the stratospheric aerosol burden, recent studies have shown that other compounds can also be present in varying amounts [20]. Meteoric and cosmic dust, primarily observed in the upper stratosphere and mesosphere, also influence the lower stratosphere [125, 126, 127]. Additionally, direct injections of primary particles are observed during volcanic eruptions, where volcanic ash is released, and after intense wildfires, which introduce black and organic carbon through pyroconvection.

Additionally, anthropogenic and natural precursor gases and primary particles emitted at the Earth's surface can reach the upper troposphere through strong convective events, followed by quasi-isentropic transport to the lowermost stratosphere or cross-isentropic into the tropical stratosphere (see Sect. 2.1.3). This process typically occurs on local and seasonal timescales, but can significantly enhance stratospheric burdens of ammonium, nitrate, dust and organic carbon [20]. More detail will be given below.

2.5.4 UTLS

The UTLS marks the transition region between the troposphere and the stratosphere, containing the tropopause. Thus, the UTLS aerosol burden is influenced by tropospheric and stratospheric processes.

The UT can be penetrated by deep convective events, which transport precursor gases and primary aerosols, particularly in the tropics. Larger particles are generally scavenged in deep convective clouds, resulting in a high amount of aerosol precursor gases at simultaneous low condensational sink [128]. As a result, bursts of new particles are observed in the outflow of these deep convective clouds [128, 129, 130, 131, 132]. This process varies across different regions of the globe, leading to distinct aerosol patterns in the UT:

- **Asian Summer Monsoon (ASM):** The South and East Asian Summer Monsoon initiates strong convective updrafts over India and China [133, 134], transporting large concentrations of anthropogenic pollutants from densely populated regions to the UT. In 2011, the Asian Tropopause Aerosol Layer (ATAL) was discovered with the help of the CALIOP instrument onboard the CALIPSO satellite, occurring seasonally from June to August [135]. Recent aircraft campaigns have revealed, that primary particles are mostly scavenged during the convective uplift, while new particles form and grow within the Asian Monsoon Anticyclone [132]. It has been shown that the ATAL primarily consists of ammonium nitrate and SOA [136, 137]. The fate and global impact of ATAL aerosol remain unclear.
- **Amazon basin:** With the movement of the Intertropical Convergence Zone (ITCZ), different areas of the Amazon basin experience strong convective events at different times of the year. During these events, large numbers of particles are created in the Upper Troposphere. The exact mechanisms behind the particle production remain unknown. [138, 139]

Sedimentation of aerosols from the stratospheric overworld can penetrate into the UTL region. While this process competes with the slow adiabatic ascent in the tropics, large scale subsidence in the extratropics leads to transport into the LS. These particles can be further transported into the UT and vice versa. Additionally, stratospheric intrusions influence the tropical UT in a similar way as the FT, particularly in mid-latitudes.

2.6 Implications of (anthropogenic) atmospheric aerosols

The discussed properties of atmospheric aerosols have significant implications for the atmosphere. Most importantly, atmospheric aerosols, whether natural or anthropogenic, influence local and global climate, public health, the ozone layer, and the hydrological cycle. The effects are often contradicting, making policy on atmospheric aerosols particularly challenging.

In Sect. 2.2.2, the interaction of aerosols with radiation and clouds were discussed. First, aerosol scattering and absorption of light directly affects Earth’s radiative balance, and anthropogenically induced enhancements of the atmospheric aerosol burden exert a **direct radiative forcing**. Second, cloud albedo is very sensitive to increasing CCN concentrations, leading to a longer cloud lifetime and brighter clouds. This results in an **indirect radiative forcing**. The latest IPCC report from Working Group I [1] calculated a total aerosol radiative forcing from both direct and indirect effects between 1750 and 2019 of -1.06 W m^{-2} , while the respective uncertainties exceed the uncertainties of all other contributors and dominate the uncertainties of the total anthropogenic radiative forcing. This translates to a cooling of the atmosphere of 0.51°C due to anthropogenic aerosol. Similarly, Lelieveld et al. [140] concluded, that a reduction in fossil fuel emissions would lead to a

2.6 Implications of (anthropogenic) atmospheric aerosols

warming of 0.51°C , while reducing all pollution particles would result in a warming of 0.73°C .

In general, aerosols have a much shorter lifetime compared to greenhouse gases [1]. As a result, the reduction of combustion processes and fossil fuel emissions to reduce anthropogenic CO_2 emissions could lead to an additional short-term warming of the atmosphere, as the provided "cooling shield" of anthropogenic aerosol will diminish before the onset of the beneficial effects of the reduced greenhouse gas concentrations.

However, the overall positive climate effects of anthropogenic aerosol is only one side of the coin. Enhanced aerosol concentrations lead to a wide range of additional consequences for the atmosphere, biosphere and humanity in general, that call for a reduction of emissions of aerosol and precursor gas emissions.

Fine particulate matter ($\text{PM}_{2.5}$) and UFP exposure are strongly associated with respiratory and cardiovascular diseases, neurodegenerative conditions, and premature mortality [2, 3, 4, 5]. Urban centres, where aerosol emissions are concentrated, face disproportionate health burdens. Furthermore, heterogeneous reactions on aerosol surfaces influence the concentrations of reactive gases such as ozone (O_3) and nitrogen oxides (NO_x) with additional health effects [141, 142, 143].

Additionally, stratospheric aerosol both, from natural and anthropogenic sources, provides surfaces for heterogeneous reactions. Most prominently, this impacts catalytic reactions, leading to depletion or enhancement of the stratospheric ozone layer, depending on atmospheric chlorine concentrations [10, 11]. Volcanic eruptions in the recent past (including Pinatubo) have led to a depletion of ozone owing to the activation of chlorine species on the surfaces of volcanic aerosols. However, as atmospheric chlorine concentrations decrease, increased aerosol surfaces likely exert a positive influence on the ozone layer. This anticipated shift arises because, at sufficiently low halogen concentrations, the effect of removing ozone-destroying NO_x species on aerosol surfaces can outweigh the catalytic ozone depletion effect associated with chlorine species [10], depending on latitude and altitude.

Additional consequences of anthropogenically enhanced aerosol include:

- Aerosols modify cloud microphysics, suppressing precipitation in some regions while intensifying storms in others [144, 145].
- High aerosol concentrations result in haze, reducing visibility and affecting both aesthetics and transportation [146, 147].
- Aerosols deposit harmful substances, such as heavy metals and acidifying compounds, onto land and water, disrupting ecosystems and biodiversity [148, 149].

One proposed solution to this dilemma is solar radiation modification [150], injecting aerosols or their precursors into the stratosphere to reflect incoming short-wave radiation back into space. While this could potentially mitigate some of the negative consequences of global warming, it also carries significant risks [151, 152]. These radiation modifications would affect the Earth's radiative balance through a different mechanism than greenhouse gases, and would additionally perturb the Earth's atmosphere with unforeseeable consequences for the hydrological cycle, stratospheric ozone, the biosphere and humanity in general. Moreover,

such an approach would require global cooperation and long-term planning, with drastic consequences of sudden modifications.

A comprehensive understanding of global aerosol distributions in the lower atmosphere and the processes that govern these distributions is essential for mitigating anthropogenic impacts on Earth's atmosphere. The work presented here aims to contribute to this understanding.

2.7 Structure & goal of the thesis

This thesis is primarily based on 3 studies: one published, one accepted for publication, and one to be submitted, presented in Chapters 3, 4 and 5. I am the first author of these studies. Chapter 6 outlines additional work I significantly contributed to. All of the presented work involves numerical atmospheric chemistry simulations, with a particular focus on global aerosol distributions.

The overarching goal of the thesis is the improvement of aerosol simulations from the Earth's surface to the stratosphere using the EMAC model, addressing the research gaps and uncertainties outlined earlier. The key outcome is one comprehensively evaluated model setup that focuses on global aerosol distributions across all layers of the Lower Atmosphere. This setup serves as the basis for both current and future studies that contribute to our understanding of global aerosol distributions and their implications for climate, public health, the hydrological cycle and atmospheric chemistry.

Chapter 3 presents the first global simulation of UFP concentrations, downscaled to a horizontal resolution of 10×10 km. To achieve this, particle emission distributions have been optimized, and nucleation channels evaluated. Global data on UFPs are crucial for cohort studies investigating the relationship between diseases and UFP exposure, as well as for estimating the health effects on a global scale, which can inform policy decisions. Additionally, improvements in modelling nucleation and primary emissions at the Earth's surface are essential for accurately simulating the transport of both primary and secondary particles to higher altitudes that is strongly size dependent.

Another key component missing for atmospheric chemistry simulations from the surface to the stratosphere was the consistent inclusion of emissions from volcanic eruptions in standard model setups. While a comprehensive amount of volcanic studies with EMAC have been conducted, these simulations typically relied on manual adjustments to account for volcanic events, and a standardized treatment of stratospheric SO₂ emissions was lacking. Chapter 4 addresses this gap by presenting the implementation and evaluation of the new MESSy submodel for Explosive Volcanic ERuptions (EVER). The new submodel handles emissions of gas and aerosol species with varying vertical shapes. A standard setup has been developed and evaluated, covering the period from 1991 to the end of 2023, which automatically includes all stratospheric SO₂ emissions based on the emission inventory by Schallcock et al. [123].

By combining the tropospheric model configuration used for simulating surface UFP and PM_{2.5} concentrations with the stratospheric setup previously applied to volcanic eruptions, and integrating the newly developed EVER submodel, a unified model configuration for

atmospheric aerosols from the surface to the stratosphere has been established. The aerosol size distribution was adjusted to represent all layers in the lower atmosphere, and the evaporation of sulfate particles has been included. The model setup is presented and evaluated in Chapter 5, demonstrating reasonable agreement with observations across all layers of the lower atmosphere. Based on this setup, global aerosol distributions of aerosol mass, number concentrations, and chemical composition are presented in the BL, the FT, the UTLS, and the stratospheric overworld.

The newly developed model setup enables the investigation of global aerosol distributions, as well as studies on specific atmospheric processes with global implications. Chapter 6 provides an overview of additional studies in which I participated as a co-author, primarily in collaboration with observational research groups. These studies focused particularly on the Amazon Basin, the South Asian monsoon system, the impacts of volcanic eruptions, and further applications related to UFP.

2.8 List of Publications

2.8.1 Published

- [P.1] M. Kuntic, I. Kuntic, D. Cleppien, A. Pozzer, D. Nußbaum, M. Oelze, T. Junglas, L. Strohm, H. Ubbens, S. Daub, M. T. Bayo Jimenez, S. Danckwardt, T. Berkemeier, O. Hahad, **M. Kohl**, S. Steven, A. Stroh, J. Lelieveld, T. Münzel, and A. Daiber. “Differential inflammation, oxidative stress and cardiovascular damage markers of nano- and micro-particle exposure in mice: Implications for human disease burden”. *Redox Biology* (2025), p. 103644. ISSN: 2213-2317. DOI: <https://doi.org/10.1016/j.redox.2025.103644>.
- [P.2] C. M. Nussbaumer, **M. Kohl**, A. Pozzer, I. Tadic, R. Rohloff, D. Marno, H. Harder, H. Ziereis, A. Zahn, F. Obersteiner, A. Hofzumahaus, H. Fuchs, C. Künstler, W. H. Brune, T. B. Ryerson, J. Peischl, C. R. Thompson, I. Bourgeois, J. Lelieveld, and H. Fischer. “Ozone Formation Sensitivity to Precursors and Lightning in the Tropical Troposphere Based on Airborne Observations”. *Journal of Geophysical Research: Atmospheres* 129.14 (2024), e2024JD041168. DOI: <https://doi.org/10.1029/2024JD041168>.
- [P.3] C. Xenofontos, **M. Kohl**, S. Ruhl, J. Almeida, H. M. Beckmann, L. Caudillo-Plath, S. Ehrhart, K. Höhler, M. Kaniyodical Sebastian, W. Kong, et al. “The impact of ammonia on particle formation in the Asian Tropopause Aerosol Layer”. *npj Climate and Atmospheric Science* 7.1 (2024), p. 215. DOI: [10.1038/s41612-024-00758-3](https://doi.org/10.1038/s41612-024-00758-3).
- [P.4] **M. Kohl**, J. Lelieveld, S. Chowdhury, S. Ehrhart, D. Sharma, Y. Cheng, S. N. Tripathi, M. Sebastian, G. Pandithurai, H. Wang, and A. Pozzer. “Numerical simulation and evaluation of global ultrafine particle concentrations at the Earth’s surface”. *Atmospheric Chemistry and Physics* 23.20 (2023), pp. 13191–13215. DOI: [10.5194/acp-23-13191-2023](https://doi.org/10.5194/acp-23-13191-2023).
- [P.5] A. Pozzer, S. F. Reifenberg, V. Kumar, B. Franco, **M. Kohl**, D. Taraborrelli, S. Gromov, S. Ehrhart, P. Jöckel, R. Sander, V. Fall, S. Rosanka, V. Karydis, D. Akritidis, T. Emmerichs, M. Crippa, D. Guizzardi, J. W. Kaiser, L. Clarisse, A. Kiendler-Scharr, H. Tost, and A. Tsimpidi. “Simulation of organics in the atmosphere: evaluation of

EMACv2.54 with the Mainz Organic Mechanism (MOM) coupled to the ORACLE (v1.0) submodel”. *Geoscientific Model Development* 15.6 (2022), pp. 2673–2710. DOI: 10.5194/gmd-15-2673-2022.

- [P.6] S. F. Reifenberg, A. Martin, **M. Kohl**, S. Bacer, Z. Hamryszczak, I. Tadic, L. Röder, D. J. Crowley, H. Fischer, K. Kaiser, J. Schneider, R. Dörich, J. N. Crowley, L. Tomsche, A. Marsing, C. Voigt, A. Zahn, C. Pöhlker, B. A. Holanda, O. Krüger, U. Pöschl, M. Pöhlker, P. Jöckel, M. Dorf, U. Schumann, J. Williams, B. Bohn, J. Curtius, H. Harder, H. Schlager, J. Lelieveld, and A. Pozzer. “Numerical simulation of the impact of COVID-19 lockdown on tropospheric composition and aerosol radiative forcing in Europe”. *Atmospheric Chemistry and Physics* 22.16 (2022), pp. 10901–10917. DOI: 10.5194/acp-22-10901-2022.

2.8.2 Accepted for publication

- **M. Kohl**, C. Brühl, J. Schallock, H. Tost, P. Jöckel, A. Jost, S. Beirle, M. Höpfner, and A. Pozzer, “New submodel for emissions from Explosive Volcanic ERuptions (EVER v1.1) within the Modular Earth Submodel System (MESSy, version 2.55.1)”. *Accepted for publication in Geoscientific Model Development (GMD) on April 9, 2025*, <https://doi.org/10.5194/egusphere-2024-2200>
- N. Tripathi, B. Krumm, A. Edtbauer, A. Ringsdorf, N. Wang, **M. Kohl**, R. Vella, L. Machado, A. Pozzer, J. Lelieveld, and J. Williams, “Impacts of convection, chemistry, and clearing on biogenic volatile organic compounds over the Amazon”. *Accepted for publication in Nature Communications on May 6, 2025*

2.8.3 In Review

- F. Köllner, **M. Kohl**, F. Ekinici, O. Eppers, J. Schneider, A. Dragoneas, S. Molleker, S. Johansson, J. Ungermann, E. Kretschmer, M. Höpfner, V. Lauther, C. Volk, O. Appel, P. Brauner, D. Kunkel, H.-C. Lachnitt, H. Bozem, F. Weyland, N. Emig, L. Ort, A. Zahn, L. Tomsche, A. Pozzer, J. Lelieveld, Y. Cheng, M. Riese, P. Hoor, and S. Borrmann, “Asian monsoon as a major source for ammonium nitrate and organic aerosol in the extratropical stratosphere”. *In Review for Nature Communications Earth & Environment*
- P. Georgiades, **M. Kohl**, M. Nicolaou, T. Christoudias, A. Pozzer, C. Dovrolis, and J. Lelieveld, “High-resolution global particle number concentration through a machine learning model and Earth observations”. *In Review for Nature Scientific Data*
- C. Xenofontos, **M. Kohl**, S. Ruhl, J. Almeida, L. Caudillo-Plath, R. Cruz-Simbron, L. Dada, J. Duplissy, S. Ehrhart, H. Finkenzeller, K. Höhler, W. Kong, F. Kunkler, C. J. Lietzke, B. Mentler, A. Morawiec, A. Onnela, P. Rato, B. Rorup, D. M. Russell, M. Schervish, W. Scholz, M. K. Sebastian, M. Simon, E. Sommer, Y. Tong, N. S. Umo, G. R. Unfer, L. Vettikkat, B. Yang, W. Yu, I. Zgheib, Z. Zheng, J. Curtius, N. M. Donahue, R. C. Flagan, H. Gordon, I. El Haddad, A. Hansel, H. Harder, X. He, J. Kirkby, M. Kulmala, K. Lehtipalo, O. Möhler, T. Petäjä, M. Pöhlker, S. Schobesberger, D. Stolzenburg, M. Wang, P. M. Winkler, D. R. Worsnop, M. Höpfner, R. Volkamer, A. Pozzer, J. Lelieveld, and T. Christoudias, “Global Impact of Anthropogenic NH₃ Emissions on Upper Tropospheric Aerosol Formation.” *In Review for PNAS*

- J. Lelieveld, P. Georgiades, **M. Kohl**, Y. Cheng, T. Christoudias, J. Sciare, M. Nicolaou, C. Dovolis, M. Fnais, M. Kulmala, A. Daiber, O. Hahad, M. Kuntic, T. Munzel, and A. Pozzer, “Global health implications of exposure to ultrafine particle air pollution.” *In Review for Science*
- D. Russell, F. Kunkler, J. Shen, **M. Kohl**, J. DeVivo, N. Bhattacharyya, C. Xenofontos, H. Klebach, L. Caudillo, M. Simon, E. Ahongshangbam, J. Almeida, A. Amorim, H. Beckmann, M. Busato, M. Canagaratna, A. Chassaing, R. Cruz-Simbron, L. Dada, P. Holzbeck, B. Judmaier, M. K. Sebastian, P. Koemets, T. Krüger, L. Liu, M. Martinez, B. Mentler, A. Morawiec, A. Onnela, T. Petäjä, P. Rato, M. Reza, S. Ruhl, W. Scholz, E. Sommer, A. Tomé, Y. Tong, J. Top, N. Umo, G. Unfer, R. Ward, J. Weissbacher, B. Yang, W. Yu, M. Zauner-Wieczorek, I. Zgheib, J. Zhang, Z. Zheng, I. El Haddad, R. Flagan, A. Hansel, H. Junninen, M. Kulmala, K. Lehtipalo, J. Lelieveld, O. Möhler, S. Schobesberger, R. Volkamer, P. Winkler, D. Worsnop, T. Christoudias, A. Pozzer, N. Donahue, H. Harder, J. Kirkby, X. He, and J. Curtius, “Isoprene chemistry under upper-tropospheric conditions.” *In Review for Nature Communications*
- R. Vella, S. Gromov, C. Nussbaumer, L. Stecher, **M. Kohl**, S. Ruhl, H. Tost, J. Lelieveld, and A. Pozzer, “Shifts in global atmospheric oxidant chemistry from land cover change.” *In Review for Atmospheric Chemistry & Physics*

3 CHAPTER

Global ultrafine particle concentrations at the Earth's surface

This chapter presents a research article published in the journal *Atmospheric Chemistry & Physics*. I am the first author of and the main contributor to this paper. I performed the model simulations with Andrea Pozzer and Sourangsu Chowdhury. I analysed the model results, evaluated the simulation and developed the downscaling procedure. I prepared the manuscript and made all figures. More detailed information on the Author contributions are provided at the end of the paper.

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Numerical simulation and evaluation of global ultrafine particle concentrations at the Earth's surface

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Abstract. A new global dataset of annually averaged ultrafine particle (UFP) concentrations at the Earth's surface for the years 2015–2017 has been developed through numerical simulations using the ECHAM/MESSy Atmospheric Chemistry model (EMAC). We present total and size-resolved concentrations along with their inter-annual variability. Size distributions of emitted particles from the contributing source sectors have been derived based on literature reports. The model results of UFP concentrations are evaluated using particle size distribution and particle number concentration measurements from available datasets and the literature. While we obtain reasonable agreement between the model results and observations (logarithmic-scale correlation of $r = 0.76$ for non-remote, polluted regions), the highest values of observed, street-level UFP concentrations are systematically underestimated, whereas in rural environments close to urban areas the model generally overestimates observed UFP concentrations. As the relatively coarse global model does not resolve concentration gradients in urban centres and industrial UFP hotspots, high-resolution data of anthropogenic emissions are used to account for such differences in each model grid box, obtaining UFP concentrations with unprecedented $0.1^\circ \times 0.1^\circ$ horizontal resolution at the Earth's surface. This observation-guided downscaling further improves the agreement with observations, leading to an increase in the logarithmic-scale correlation between observed and simulated UFP concentrations to $r = 0.84$ in polluted environments (and 0.95 in all regions), a decrease in the root mean squared logarithmic error (from 0.57 to 0.43), and removal of discrepancies associated with air quality and population density gradients within the model grid boxes. The model results are made publicly available for studies on public health and other impacts of atmospheric UFPs, as well as for intercomparison with other regional and global models and datasets.

1 Introduction

Atmospheric aerosols in various size ranges have a significant impact on public health, the hydrological cycle and climate. Close to the Earth's surface, aerosol particles are among the main pollutants and drivers of atmospheric chemistry in the boundary layer, being directly relevant for human health (Burnett et al., 2014; Pope and Dockery, 2006; Cohen et al., 2005). At the same time, aerosols can directly scatter and absorb solar and thermal radiation, altering the radiative balance of the Earth's atmosphere (e.g. Bellouin et al., 2020). Furthermore, atmospheric aerosols act as cloud condensation nuclei (CCN) and thus influence cloud formation processes and cloud properties. Consequently, changes in CCN concentrations may affect the hydrological cycle and indirectly the radiative balance of the Earth's atmosphere by altering cloud cover and albedo (e.g. Christensen et al., 2020; Lohmann and Feichter, 2005; Twomey, 1959).

Recently, multiple studies have concluded that the exposure to particulate matter air pollution from a variety of sources has major implications for public health (Lelieveld et al., 2020, 2015; Chowdhury et al., 2022). The latest Global Burden of Disease study has associated 4.2 million deaths globally with the exposure to ambient particulate matter with an aerodynamic diameter smaller than $2.5\ \mu\text{m}$ ($\text{PM}_{2.5}$) and 0.37 million deaths to ambient ozone pollution (Murray et al., 2020). Evidence presented in recent studies indicates that the long-time exposure to high concentrations of ultrafine particles (UFPs), i.e. particles with an aerodynamic diameter smaller than 100 nm (WHO, 2006) which barely contribute to $\text{PM}_{2.5}$ mass, significantly impacts human health, leading to the increased incidence of cardiovascular and cerebrovascular diseases (Downward et al., 2018; Delfino et al., 2005; Stone et al., 2017). The health impacts of UFPs may be attributed to their high potential to penetrate more deeply into the lungs and potentially into the bloodstream compared to coarser size particles (Schraufnagel et al., 2019; Schraufnagel, 2020; Hong and Jee, 2020).

Reflection and absorption of solar radiation can be neglected for UFPs as the scattering and absorption coefficients typically peak at particle diameters between 400 and 1000 nm. However, aerosol particles with a minimum diameter of approximately 40 nm and larger can serve as CCN, while particle numbers strongly decrease for sizes greater than 200–300 nm in general (Andreae, 2009). Thus, UFPs significantly contribute to CCN concentrations.

Atmospheric aerosols can either be directly emitted into the atmosphere (primary) by natural or anthropogenic processes or be nucleated from precursor gases (secondary), with the latter being considered to be the largest source of atmospheric aerosols (Gordon et al., 2017). Freshly nucleated secondary particles usually have a diameter between 1 and 20 nm (Curtius, 2006) and can grow by coagulation and condensation of trace gases (Kulmala et al., 2004). While primary aerosols from natural sources (e.g. desert dust and sea

salt) are emitted at diameters predominantly in the micrometre range and larger (Dentener et al., 2006), anthropogenic particles (e.g. from combustion processes) are usually emitted at much smaller sizes, contributing to UFPs (Kwon et al., 2020; Paasonen et al., 2016).

Despite the importance of UFPs for atmospheric processes and human health, very little is known about their global distribution at the Earth's surface. A simulation of global particle number concentrations at the Earth's surface was performed by Gordon et al. (2017), however focusing on new particle formation (NPF) and CCN up to an altitude of 460 m above the surface, and by Chen et al. (2021) with a nested high-resolution simulation over eastern Asia ($0.33^\circ \times 0.33^\circ$) and a particular focus on organic aerosols. Local (surface or vertical) distributions of particle number concentrations and size distributions were measured and modelled by Ketzel et al. (2021) and Frohn et al. (2021, both street- and address-level UFP concentrations in Denmark), Fountoukis et al. (2012) and Saha et al. (2021, high-resolution UFP concentrations over Europe and the United States, respectively), Kukkonen et al. (2016, dispersion of particle numbers in European cities), Franco et al. (2022, NPF and growth in the Amazon rainforest), Williamson et al. (2019) and Liu and Matsui (2022, both NPF and the contribution of organic aerosols in the remote atmosphere from the surface to the upper troposphere), and Weigel et al. (2021, NPF in the South Asian monsoon).

Thus, present knowledge on global surface UFP concentrations is mostly limited to local model studies and in situ measurements of particle size distributions (PSDs), from which UFP concentrations can be inferred. While the number of PSD measurements is increasing (Wu and Boor, 2021; Rose et al., 2021), they are still sparse and mostly not continuous over time. Furthermore, there are no clear methodological guidelines for measuring PSDs or particle number concentrations (PNCs) (Trecheta et al., 2023), and measurement size limits vary greatly. However, new recommendations have recently been suggested (CEN/TS 17434:2020, 2020; CEN/TS 16976:2016, 2016; ACTRIS, 2021).

To generate a first, global, annually averaged UFP dataset for the years 2015–2017 we used the ECHAM/MESSy Atmospheric Chemistry model (EMAC; Jöckel et al., 2006), including gas-phase and heterogeneous chemistry with comprehensive chemical mechanisms, aerosol microphysics with size-resolved particulate matter and cloud interactions. Such data can be applied to investigate the impact of UFPs on public health, CCN formation and cloud properties, as well as for intercomparison studies with other regional and global models and datasets.

Several emission inventories describe the total emitted mass of aerosol and gas species on a global grid (e.g. Hoesly et al., 2018; Crippa et al., 2020; Granier et al., 2019). However, the inference of the number of emitted particles is very sensitive to their size distribution as small changes can lead to large deviations in the resulting particle number concen-

trations. Information on these size distributions is rare, and the uncertainties are high (Paasonen et al., 2016). For that reason we use existing information on PSDs of emitted particles from the literature and evaluate our results against measurement data in China, India, Europe, the United States and various remote regions.

Another challenge in the global modelling of UFPs is the limited model resolution. Studies showed that UFP concentrations return to background levels within a distance of about 1000 m from the source (e.g. Karner et al., 2010, for roadways). Thus, UFP concentrations can show sharp urban to rural gradients (e.g. Saha et al., 2021) that cannot be captured efficiently by a global model that is limited in the horizontal grid size. As a result, high local UFP concentrations in densely populated urban areas may be artificially diluted by the surrounding regions in the grid boxes. This may result in an underestimation of UFP concentrations in city centres, while concentrations in remote regions close to urban areas may be overestimated. This shortcoming in the evaluation of model results is studied in depth as the inability of localized measurements to represent the grid box environment (representation error) in Schutgens et al. (2016a, b, 2017).

The correlation between the underestimation of the model on the one hand and local high-resolution anthropogenic emissions at the measurement sites relative to the average anthropogenic emissions at model resolution on the other hand is used here to derive UFP concentrations at $0.1^\circ \times 0.1^\circ$ horizontal resolution. We show that this improves the agreement between observations and simulations, reduces the spatial representation error, and decreases inconsistencies introduced by the difficulties of the coarse model resolution to capture population density and air pollution gradients.

2 Global model and methods

The ECHAM/MESSy Atmospheric Chemistry (EMAC) model (Jöckel et al., 2006) is a combination of the fifth-generation European Centre Hamburg general circulation model, ECHAM5 (Roeckner et al., 2003), which serves as the dynamical base model, and the second version of the Modular Earth Submodel System, MESSy2 (Jöckel et al., 2010), comprising various submodels that describe the chemistry and physics of the atmosphere.

The simulation used for this study is performed at a spectral, horizontal resolution of T63 ($1.875^\circ \times 1.875^\circ$ or approx. 180×180 km at the Equator) with 31 vertical, hybrid terrain-following and pressure levels up to 10 hPa altitude and the surface level extending up to approximately 45–70 m above the surface, depending on latitude and season. The model simulation is “nudged” (Jeuken et al., 1996; Jöckel et al., 2006) towards meteorological reanalysis data of the years 2014–2017 (ERA-Interim; Berrisford et al., 2011) from the European Centre for Medium-Range Weather Forecasts (ECMWF).

Global anthropogenic emissions of reactive gases and aerosols at the surface from the simulated years are taken from the Community Emissions Data System (CEDS; McDuffie et al., 2020a, b). We consider primary emitted black carbon (BC), organic carbon (OC) and sulfate (SO_4 ; 2.5 % of SO_2 emissions according to Dentener et al., 2006) as direct aerosol sources at the surface. The CEDS anthropogenic emissions from the sectors energy generation (ENE), industries (IND), land transportation (TRA), domestic energy use (DOM), waste (WST), agricultural soils (AGR), solvent production and application (SLV; no primary particle emissions), and ship and other navigation (SHIP) are considered. Emissions from biomass burning (BB) and agricultural waste and residue burning (AWB) were calculated daily using the BIOBURN submodel based on observed dry matter burned and fire type compiled by Andreae (2019). The biomass burning emissions for OC and BC were increased by a factor of 4.48 and 2.8, respectively, based on the work of Pan et al. (2020) and a comparison with observations in the Amazon Basin (Holanda Bruna, personal communication, 2023). Aircraft emissions of reactive gases, BC and OC were taken from the CAMS global aviation emissions (CAMS-GLOB-AIR; Granier et al., 2019). Sea salt (see algorithm from Guelle et al., 2001) and dust emissions (see algorithm from Klingmüller et al., 2018) are calculated online using the submodel ONEMIS (Kerkweg et al., 2006). All emissions are distributed to six different emission height levels based on the description by Pozzer et al. (2009).

Aerosols are treated using the MESSy submodel GMXe (Pringle et al., 2010). Aerosol microphysics are based on aerosol size distributions with currently seven interactive lognormal modes that cover the typical size spectrum of aerosol species and differentiate into four hydrophilic (nucleation, Aitken, accumulation and coarse) and three hydrophobic (Aitken, accumulation and coarse) aerosol modes. All aerosols are approximated as spherical particles. The properties of aerosols in each mode are completely defined by the total mass (internal mixture of contributing species), density, number concentration, median radius and width of the lognormal distribution. After each simulation step aerosols may transfer between modes depending on size changes. Organic aerosol species are additionally described by the Organic Aerosol Composition and Evolution (ORACLE; Tsimpidi et al., 2018) submodel, taking into account the partitioning between aerosols and the gas phase. ORACLE distinguishes between primary and secondary organic aerosols from different sources and volatilities (in up to five logarithmically spaced saturation concentration bins, ranging from 10^{-2} to $10^6 \mu\text{g m}^{-3}$, depending on the emission sector).

Heterogeneous and gas-phase chemistry are treated with the submodel MECCA (Sander et al., 2019) with the Mainz Isoprene Mechanism (MIM1; Pöschl et al., 2000; Jöckel et al., 2006) as the chemical mechanism, comprising more than 100 gas-phase species and more than 250 reactions. Dry deposition, sedimentation and wet deposition are simu-

lated with the submodels DDEP, SEDI (both Kerkweg et al., 2006) and SCAV (Tost et al., 2006), respectively. The submodel NAN (Ehrhart et al., 2018) is used to estimate binary and ternary nucleation following Dunne et al. (2016), pure organic nucleation (Kirkby et al., 2016), and nucleation from oxidized organic species and sulfuric acid (Riccobono et al., 2014). The parameterization of ion-induced nucleation is included in NAN, using ion pair production and steady-state ion concentrations from the submodel IONS (Ehrhart et al., 2018). IONS calculates the ion pair production from galactic cosmic rays and from radon decay. The latter is obtained from the diagnostic radon (DRADON) submodel (Jöckel et al., 2010).

Aerosols at the surface simulated with the EMAC model have been extensively evaluated in many publications, with a focus on either PM_{2.5} mass or aerosol optical depth (Pozzer et al., 2012, 2015; Lelieveld et al., 2019; Pozzer et al., 2022). Chowdhury et al. (2022) conducted an evaluation for aerosol optical depth, PM_{2.5}, and black carbon and organic aerosols in PM_{2.5} using a similar setup as used in this study.

The number of emitted aerosols (N_{aer}) is calculated as

$$N_{\text{aer}} = \frac{6 \cdot M_{\text{aer}}}{\pi \cdot \rho_{\text{aer}} \cdot d_{\text{med}}^3} \cdot \exp(-4.5 \ln^2 \sigma_{\text{ln}}), \quad (1)$$

where the emitted aerosol mass M_{aer} is given by the respective emission dataset. ρ_{aer} is the density of the considered aerosol species, and σ_{ln} is the width of the lognormal mode in the model. The fraction is just the geometrical derivation of the number of spherical particles from total mass, given a diameter d_{med} , while the exponential function corrects for the lognormal volume distribution (compare Eqs. 8.34 and 8.51 from Seinfeld and Pandis, 2016). The median diameter of the emitted particles (d_{med}) depends on the considered sector and species. As $N_{\text{aer}} \propto 1/d_{\text{med}}^3$, the number of emitted particles is highly sensitive to the emission median diameter; i.e. a doubling of the diameter leads to a reduction in the number of emitted particles by a factor of 8.

In order to emit the aerosols with a realistic size distribution for each sector, a detailed investigation of the emission size distributions is performed based on the findings of Paasonen et al. (2016). Paasonen et al. (2016) used an emission model (Amann et al., 2011) in combination with emission factors and size distributions from the literature in order to obtain global emission particle size distributions for different sectors. As the median emission diameter d_{med} is a global quantity in the EMAC model, we derived the median of the present global size distributions from Paasonen et al. (2016) and used them as d_{med} for the corresponding sectors in our model. The distributions along with their median diameters are depicted in Fig. 1.

Additionally, the aerosol median emission diameter from biomass burning and AWB is estimated to be 130 nm based on the average of multiple studies on biomass burning emissions summarized in a review (Reid et al., 2005), confirmed

by Janhäll et al. (2010, who measured 120 nm) and the respective size distribution from Paasonen et al. (2016) for AWB (median diameter of 126 nm). Ship aerosol emissions are assumed to be represented by a median diameter of 40 nm based on studies by Kasper et al. (2007, who found 20–40 nm for low-speed marine diesel engines), Diesch et al. (2013, small nucleation mode from 10–20 nm and Aitken mode around 35 nm) and Petzold et al. (2008, 52 nm in fresh plume and up to 100 nm in aged plume). Aerosols from aircraft emissions are emitted at a diameter of 40 nm as well, based on studies from Petzold and Schröder (1998) and Petzold et al. (2003), who distinguished a mode between 30 and 45 nm and an additional smaller accumulation mode around 180 nm. Dust particles are emitted only in the accumulation and coarse modes according to Klingmüller et al. (2018) and play a negligible role in UFP concentrations (d'Almeida, 1987). For the emissions of sea salt we use the diameters from the algorithm of Monahan (1986), also only emitting particles in the accumulation and coarse modes.

As only a few measurements of PSDs are available, which can be used to directly infer UFP concentrations, measurements of total PNCs are additionally used for the model evaluation as they are often dominated by UFPs, especially close to UFP sources (e.g. Baldauf et al., 2016; Kumar et al., 2014). Figure 2 shows a typical simulated PSD (blue line) as the sum of the four soluble and three insoluble modes. The solid red line shows the upper bound for UFPs at 100 nm. The dashed red line is the variable lower bound, used exclusively for comparison to observations, as measurement devices entail a cut-off particle diameter below which no particles are detected. The published global UFP dataset includes arbitrarily small particles and does not use a lower cut-off value.

The UFP concentration in the simulation N_{UFP} is calculated using the particle number concentration $N_{\text{tot},i}$, the width σ_i and the median diameter $D_{\text{m},i}$ of each mode i :

$$N_{\text{UFP}} = \sum_{i=1}^7 \frac{N_{\text{tot},i}}{2} \cdot \left\{ \operatorname{erf} \left(\frac{\ln(D_{\text{up}}/D_{\text{m},i})}{\sqrt{2} \ln(\sigma_i)} \right) - \operatorname{erf} \left(\frac{\ln(D_{\text{low}}/D_{\text{m},i})}{\sqrt{2} \ln(\sigma_i)} \right) \right\}, \quad (2)$$

where D_{up} is the fixed upper bound of 100 nm, and D_{low} is the variable lower detection limit associated with the measurement device below which no particles can be detected. The cut on the detection limit is applied on the simulation for comparisons with observations. For the final global dataset we report the total number of UFPs, and thus D_{low} is set to 0 nm, and the second error function in Eq. (2) equates to -1 .

The total PNC is the sum of the particle number concentrations $N_{\text{tot},i}$ for each mode (i.e. $D_{\text{up}} = +\infty$, and the terms in the curly brackets in Eq. (2) add up to $+2$ for $D_{\text{low}} = 0$). However, PNC measurements also have a lower cut-off value, which has to be considered for model evaluation when comparing simulated and measured PNCs.

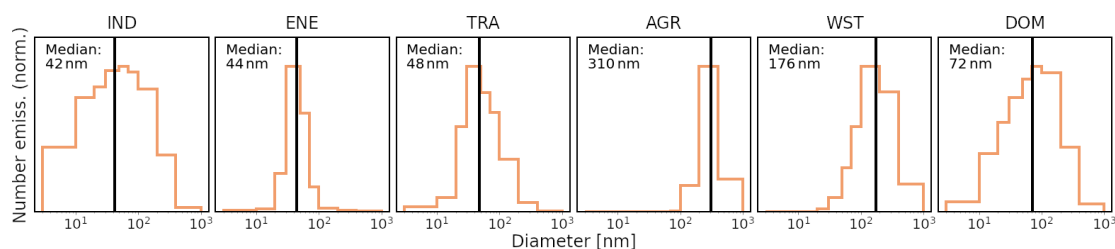


Figure 1. Normalized size distributions of primary emitted particles from different sectors (IND: industries, ENE: energy generation, TRA: land transportation, AGR: agricultural soils, WST: waste, DOM: domestic energy use) taken from Paasonen et al. (2016) with corresponding median diameters derived from these distributions (black line). The median diameters are taken as median diameters for the sector emissions in our simulation.

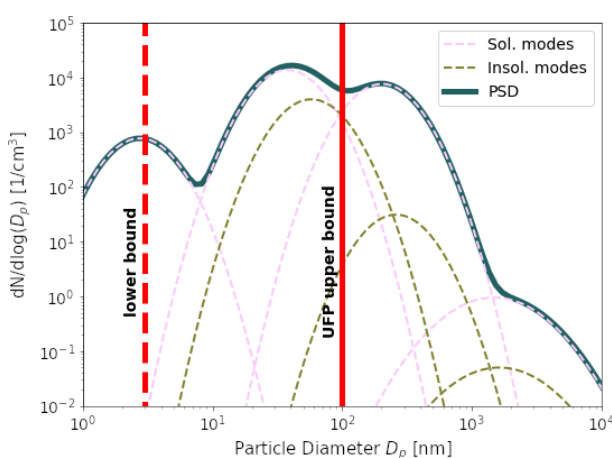


Figure 2. Typical particle size distribution (PSD) taken from the simulation in an urban region. The dashed lines represent the soluble and insoluble aerosol modes. The PSD is the sum of all these modes (blue line), typically dominated by the soluble modes. UFPs are defined as all particles with a diameter below 100 nm (right red line), while the total particle number concentration is the full integral over the PSD. For comparison between observed and simulated concentrations, the lower bound (dashed red line) is considered a cut-off, which depends on the detection limit of the measurement device. The final dataset includes all UFPs without lower bounds.

3 Observations

In order to evaluate the simulated UFP concentrations from the EMAC model, observations were collected from different stationary measurement sites at the Earth's surface with a focus on polluted regions. The simulation has been performed for the years 2015–2017 with 1 year of spin-up (2014), whereas we evaluated the simulation for the year 2015 only. Thus, if available, the simulations were compared to observations in 2015. However, to increase the number of available datasets to compare with simulation results, we additionally used annual averages of observed UFP concentrations and PNCs from all available years for the evaluation. This is noted accordingly in the following sections. Observational

data were collected for UFP concentrations and PNCs and were compared to the respective calculated simulated values according to the lower cut-off value of the respective measurement devices. The sources of the observational data of UFPs (derived from PSDs) and PNCs are listed below.

3.1 EBAS

EBAS is a database for atmospheric measurement data operated by the Norwegian Institute for Air Research (NILU) and contains measurements for different programmes of which we used the following:

- The European Monitoring and Evaluation Programme (EMEP) monitors air pollutants in Europe.
- The Global Atmosphere Watch – World Data Centre for Aerosols (GAW-WDCA) is a data repository for microphysical, optical and chemical properties of atmospheric aerosol.
- The Aerosol, Clouds and Trace Gases Research Infrastructure (ACTRIS) contains long-term atmospheric measurement data.

The data were obtained from the EBAS database (<http://ebas.nilu.no/>, last access: 17 February 2022). We analysed all available PSDs (mostly in Europe and remote regions) and PNC measurements (Europe, North America and remote regions) taken in 2015.

3.2 Field measurements, literature and published datasets

We derived UFP concentrations from PSDs measured by groups involved in the present study in India, China and the Amazon rainforest. In India PSDs were measured in Delhi (Thamban et al., 2021), Mahabaleshwar and Hyderabad (both Sebastian et al., 2022). Measurements in China were taken in Shanghai (unpublished), Beijing (Liu et al., 2020), Lin'an (Shen et al., 2022) and Gucheng (Li et al., 2021). Additionally, we used measurements from the Amazon Tall Tower Observatory (ATTO) centrally located in

the rainforest in Brazil, about 150 km northeast of Manaus (Franco et al., 2022). The observational datasets are complemented by including literature and published datasets from China and India (Gani et al., 2020; Wu et al., 2008), as well as from the Atmospheric Tomography mission (ATom) aircraft campaign (Brock et al., 2019).

4 Results

Annual averages of modelled UFP (UFP_M) concentrations in 2015 on a global scale are displayed in Fig. 3, ranging from about 40 cm^{-3} over parts of the ocean to up to more than $12\,000 \text{ cm}^{-3}$ in China, India, Indonesia and Papua New Guinea (considering averages over model grid boxes). UFP_M concentrations are generally high across Europe, ranging from 2000 to 6000 cm^{-3} ; however, they are lower than in several Asian hotspot areas. In North America, UFP_M concentrations are simulated to be higher along the East Coast, with additional hotspots in and around the big cities along the West Coast, reaching up to 8000 cm^{-3} in and around San Francisco and Los Angeles. We also simulate high UFP_M concentrations close to $10\,000 \text{ cm}^{-3}$ in the West Coast of South America and regional enhancements along the East Coast. Southern Australia, northern Africa and the Middle East (the Persian Gulf region) also show enhanced UFP_M concentrations exceeding 8000 cm^{-3} .

Continental polar regions in North America, Europe, Asia and Antarctica are simulated to have UFP_M concentrations mostly below 500 cm^{-3} due to the absence of pollution particle sources. Low UFP_M concentrations (down to 150 cm^{-3}) are also simulated across the Amazon rainforest and in major desert regions (e.g. the Sahara). However, there is a lack of measurement data of PNCs and UFP concentrations in desert regions that hinders the evaluation of the model results. Low UFP concentrations in the boundary layer over the Amazon forest have been reported previously and have been attributed to missing sources of primary UFPs and the growth of secondary particles during transport from the upper troposphere through the condensation of oxidized organic species, reaching the boundary layer through convective downdrafts (Zhao et al., 2020; Wang et al., 2016; Andreae et al., 2018).

UFP_M concentrations over the oceans are highly variable, with relatively high values exceeding 2500 cm^{-3} over the Pacific and Indian oceans downwind of pollution sources on land and very low values below 50 cm^{-3} over the Southern Ocean and tropical Atlantic and Pacific oceans. Low UFP concentrations in the tropical ocean environment have been observed by the ATom aircraft campaign as well, potentially caused by the efficient removal of small particles by coagulation and again the downward transport of aged secondary particles from the upper troposphere (Williamson et al., 2019).

PNCs were previously modelled by Gordon et al. (2017) at relatively low global resolution, Saha et al. (2021) for

the United States only and Chen et al. (2021) with a focus on eastern Asia. Gordon et al. (2017) concentrated on cloud condensation nuclei and averaged over a vertical column of 460 m. We generally simulate higher concentrations compared to Gordon et al. (2017), mostly capturing UFP hotspots, in accordance with observations, especially after the redistribution based on anthropogenic emissions (see Sect. 4 and 4.2). The comparably lower concentrations simulated by Gordon et al. (2017) might partially result from the larger vertical column in the simulation, the coarser resolution or the different emission diameters (globally 60 nm for all fossil-fuel-related emissions). Our simulated UFP concentrations agree very well with the PNC simulation from Chen et al. (2021) in most continental regions, especially in Europe, North and South America, Africa, Australia, and eastern Asia (which they focused on). However, simulated PNCs in India are considerably lower than our UFP concentrations (supported by observations; see Sect. 4.1.3). Additionally, we simulate higher UFP concentrations at marine southern mid-latitudes (in reasonable agreement with measurements taken during the ATom campaign; see Sect. 4.1.5) than Chen et al. (2021).

The simulated spatial distribution of UFPs over the contiguous United States matches that of Saha et al. (2021). They used a land use regression model to produce a high-resolution ($200 \times 200 \text{ m}$) product of UFP concentrations. We reach similar values in the urban regions in the United States after the redistribution based on anthropogenic emissions as described in Sect. 4.2; however the values are still lower than the highest values simulated by Saha et al. (2021). The difference might be due to the fact that UFP concentrations are a subset of PNCs. Ketzel et al. (2021) simulated PNCs at the street- and address-level on the same order of magnitude as our simulation up to the urban background level but with much higher peak values, reaching more than $30\,000 \text{ cm}^{-3}$ in traffic hotspots, which we cannot resolve. Our simulated UFP concentrations are much lower than the modelled results from Fountoukis et al. (2012), which reach around $20\,000 \text{ cm}^{-3}$, even for background regions in Europe in May 2008, and up to more than $100\,000 \text{ cm}^{-3}$ in local hotspots in southeastern Europe. However, the general increasing trends from western to eastern Europe agree.

Trechera et al. (2023) analysed observations of PNCs and UFP concentrations in Europe from 2017 to 2019, focusing on daily and seasonal patterns, UFP drivers, and regional trends. They again find increasing UFP concentrations from northern to southern and from western to eastern Europe. Our simulation also exhibits the west–east tendency in Europe, however no clear pattern from north to south. Seasonality and drivers of UFP concentrations and composition will be the subject of follow-up studies.

UFPs have widely differing size distributions across the globe. Figure 4 illustrates this based on three size bins, i.e. 1.7–20, 20–50 and 50–100 nm. Freshly nucleated particles between 1.7 and 20 nm mostly dominate in remote environ-

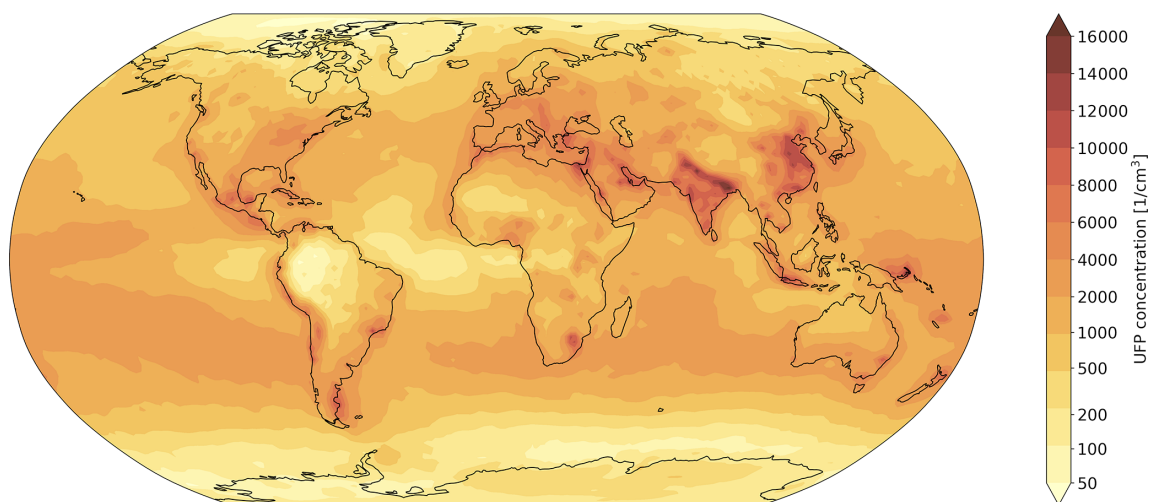


Figure 3. Annual average of global UFP concentrations simulated with EMAC for the year 2015 at model resolution of $1.875^\circ \times 1.875^\circ$ and from the lowest vertical model level (surface level).

ments, especially over oceans. PSDs usually peak at diameters between 20 and 50 nm in polluted regions, representing a mixture of freshly emitted primary particles from fossil fuel combustion and rapidly grown secondary particles. Biomass burning particles are typically emitted at larger diameters (see Fig. 1). Additionally, high concentrations of organic vapours lead to strong condensational growth in forest regions. Thus, UFPs are dominated by 50–100 nm particles there.

The interannual variability in UFP concentrations is presented in Fig. 5. We observe strong interannual variation, exceeding 1000 cm^{-3} and (up to 50 %) along the Intertropical Convergence Zone (ITCZ), which is mostly caused by meteorological differences in the different years, potentially caused by the strong El Niño in 2015, followed by weaker La Niña and El Niño events in 2016 and 2017. The meteorological influence on UFP concentrations could be the basis for future studies. We additionally find absolute interannual variation exceeding 1000 cm^{-3} over polluted regions, although below 20 % in relative terms. This is mostly due to a decreasing trend in emissions over southern and eastern Asia, Europe, and the United States. However, we observe an increasing trend in UFP concentrations over parts of Africa, especially over Nigeria and South Africa.

4.1 Evaluation

In the following, we present the evaluation of the simulated UFP_M concentrations based on observational data. Section 4.1.1 to 4.1.4 evaluate urban and rural regions in four comparably highly polluted regions for which measurement data are available, namely Europe, North America, India and China. In Sect. 4.1.5 remote regions in polar, forest, mountain and ocean environments with a lower population den-

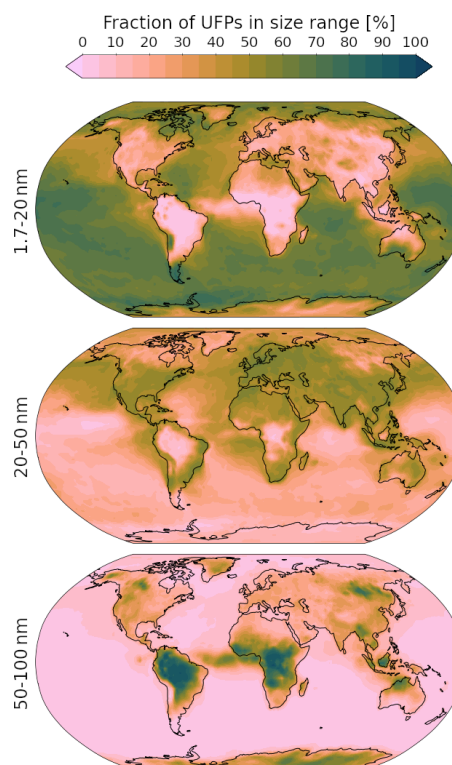


Figure 4. Fraction of UFPs in different size ranges, representing freshly nucleated particles (1.7–20 nm), fossil fuel emissions or grown nucleation particles (20–50 nm), and further grown or larger emitted particles (50–100 nm).

sity are evaluated. Mountain environments are defined covering altitudes from 1500 to 3000 m a.s.l., while measurements

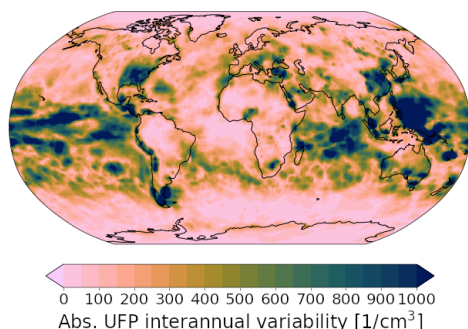


Figure 5. Absolute interannual variation in UFP concentrations calculated based on the years 2015–2017. It is defined by the maximum minus the minimum annually averaged UFP concentrations.

above 3000 m a.s.l. were excluded as they are mostly located in the free troposphere, and this study concentrates on boundary layer processes and the respective evaluation.

Annual averages of observed UFP (UFP_O) concentrations and PNCs (PNC_O) were either derived from daily PSDs (by integrating the number of aerosol particles per size bin from the lowest measurement bin up to the bin crossing the upper size threshold of 100 nm) and PNCs – removing outliers differing by a factor greater than 10 from the median value and subsequently calculating the annual average – or directly provided. The daily averaged number concentrations of the model aerosol modes were integrated for the same size region as the observations (from the lower detection limit of the measurement device up to the highest measurement bin with a mean diameter below 100 nm) according to Eq. (2). UFP_M concentrations were sampled at the vertical grid box covering the measurement site altitude.

The evaluation in this section concentrates on the modelled (UFP_M or PNC_M) and observed (UFP_O or PNC_O) concentrations. The downscaled (i.e. redistributed within each grid box) concentrations (UFP_R or PNC_R) are the subject of Sect. 4.2. We used three different statistical measures for the evaluation based on daily model output and observations if available:

- PF2, percentage of modelled values that are simulated within the range of a factor of 2 of the observed values;
- NRMSE, root mean squared error (RMSE) normalized by the range of the observations; and
- $\overline{M/O}$, geometric mean of the ratio between the modelled (M) and observed daily mean (O).

4.1.1 Europe

UFP_O concentrations in Europe were derived from daily PSDs provided by the EBAS database. An overview of the different measurements in Europe and a comparison to

UFP_M are presented in Table 1. We used all available observations from the EBAS database from 2015 in Europe from rural and urban stations. The remote stations as defined above are excluded here and are discussed separately in Sect. 4.1.5. Additionally, we excluded measurements in Athens (GR) and Preila (LT) due to apparent inconsistencies in the observational datasets.

The simulation shows reasonable agreement with the observations at most measurement stations with $\text{PF2} \geq 40\%$, $\text{NRMSE} \leq 0.4$ and $0.5 \leq \overline{M/O} \leq 2.0$. However, there are some exceptions. At the measurement stations of Madrid, Leipzig-Eisenbahnstrasse, Leipzig-Mitte and Dresden-Nord, the model strongly underestimates the UFP concentrations. This is due to a combination of two effects. Firstly, UFP concentrations are typically related to anthropogenic emissions (see Sect. 4.2 for details). The high UFP concentrations in the densely populated urban centres are artificially diluted in the simulation by lower concentrations in the surroundings covered by the grid box. This spatial representation error after Schutgens et al. (2016a) can partly be corrected for by redistributing UFP concentrations using high-resolution anthropogenic emission datasets, discussed in detail in Sect. 4.2. Secondly, the measurements were performed close to busy streets (the Leipzig-Eisenbahnstrasse, Leipzig-Mitte and Dresden-Nord sites particularly measure traffic emissions) with high primary UFP emissions from vehicles, which would require impracticable street-level horizontal resolution to resolve.

The model overestimates UFP_M concentrations at the stations of L'Observatoire pérenne de l'environnement (OPE; France) and Finokalia (GR). OPE is located at a remote region close to Nancy, and the grid box is thereby highly influenced by the urban region. Finokalia on the other hand is located in a rural environment close to the coast of the island Crete. The interference of the oceanic influence and surrounding enhanced anthropogenic emissions from the island leads to enhanced UFP concentrations within the grid box.

Multiple measurements in one city provide an interesting case of the variability in UFPs within one modelled grid box. Measurements were performed at four different locations in Leipzig (shown in Fig. 6), varying by up to a factor of 3, while simulated UFP_M concentrations (dashed grey line) approximately coincide with the observations taken at some distance from heavy traffic locations (Leipzig). The station at Leipzig-Eisenbahnstrasse is at a curbside and measures very high UFP_O concentrations. It can be noted that the model is capable of reproducing urban background conditions, i.e. the average over urban regions and their surroundings in the grid box, while not replicating local UFP hotspots, for example roadsides or near industrial emitters, due to limited horizontal resolution. It is expected that dilution and coagulation during atmospheric transport from these hotspots to the surroundings will quickly reduce the UFP concentrations to levels that are realistically represented by the urban back-

Table 1. Summary of observations of UFP (UFP_O) concentrations (cm^{-3}) derived from PSDs across Europe for 2015, with data archived in the EBAS database. The geographical location is indicated by latitude (Lat) and longitude (Long), and the elevation of the site above the sea surface is indicated by the altitude a.s.l. in metres (Alt). The lower cut-off value (Cut) from the measurement device (also applied on the simulation) is expressed in nanometres. The annual averages of the UFP_O and UFP_M concentrations for the grid box encompassing the station are compared, and different measures of agreement are listed. The same comparisons are performed after the redistribution of the model (Redistributed model, UFP_R), which are discussed in detail in Sect. 4.2. All included measurement stations contained at least 200 d of valid measurements in 2015. Abbreviations: DD – Dresden, NOAK – National Atmospheric Observatory Košice, ECO – Environmental Climate Observatory, LE – Leipzig, OPE – Observatoire pérenne de l'environnement.

Station	Observations					Model results				Redistributed model			
	Lat	Long	Alt	Cut	UFP_O	UFP_M	PF2	NRMSE	$\overline{M/O}$	UFP_R	PF2	NRMSE	$\overline{M/O}$
Annaberg-Buchholz (DE)	50.57	13.0	545	9.4	6649	3597	55	0.21	0.55	3317	49	0.22	0.51
Cabauw Zijdweg (NL)	51.97	4.93	1	9.1	6324	4539	77	0.22	0.72	3828	62	0.26	0.6
DD-Nord (DE)	51.06	13.74	120	4.8	10 526	4289	29	0.34	0.39	7690	67	0.25	0.7
DD-Winckelmannstr (DE)	51.04	13.73	112	9.4	5474	4218	85	0.16	0.78	7562	77	0.22	1.39
ECO Lecce (IT)	40.34	18.12	36	9.4	5372	3741	59	0.36	0.62	4821	62	0.37	0.8
Finokalia (GR)	35.34	25.67	250	8.6	1421	3628	34	0.6	2.3	3917	28	0.67	2.48
Hohenpeissenberg (DE)	47.8	11.01	985	9.4	2302	2479	78	0.25	1.08	2234	76	0.23	0.97
K-puszt (HU)	46.58	19.35	125	6.0	4040	5127	67	0.27	1.32	4966	66	0.26	1.28
LE-Eisenbahnstr (DE)	51.35	12.41	120	4.8	14 410	4358	15	0.37	0.3	5020	23	0.35	0.34
LE-Mitte (DE)	51.34	12.38	111	4.8	10 624	4251	27	0.34	0.39	5521	47	0.3	0.5
LE-West (DE)	51.32	12.3	122	4.8	6222	4451	74	0.21	0.69	4686	77	0.21	0.73
Leipzig (DE)	51.35	12.43	118	4.8	4882	4300	88	0.17	0.86	4953	91	0.17	0.99
Madrid (ES)	40.46	−3.73	669	14.4	11 311	2916	22	0.17	0.28	6192	59	0.15	0.59
Melpitz (DE)	51.53	12.93	87	4.8	7018	4343	72	0.19	0.62	4400	72	0.19	0.63
Montseny (ES)	41.78	2.36	700	8.9	3056	2375	69	0.21	0.78	2371	69	0.21	0.78
NOAK Kosetice (CZ)	49.58	15.08	534	8.6	2569	4332	57	0.3	1.79	3701	66	0.24	1.53
Neuglobsow (DE)	53.14	13.03	62	9.4	2808	3515	69	0.17	1.3	2986	74	0.15	1.11
OPE (FR)	48.56	5.5	392	9.7	1834	2865	63	0.32	1.6	2786	64	0.31	1.55
Prague-Suchdol (CZ)	50.12	14.38	277	5.6	6421	4412	74	0.15	0.7	6124	89	0.13	0.97
SIRTA Palaiseau (FR)	48.71	2.16	162	10.0	4239	3441	82	0.2	0.84	5391	79	0.24	1.31
Schauinsland (DE)	47.9	7.92	1205	9.4	1545	1833	78	0.19	1.22	1783	80	0.19	1.19
Vielsalm (BE)	50.3	6.0	496	8.8	2082	2596	75	0.22	1.26	2628	73	0.23	1.27

ground (e.g. Karner et al., 2010, finding a distance of 1000 m for roadways). This is in agreement with a study from Salma et al. (2014), showing measured UFP concentrations at different locations across Budapest, Hungary. They measured on average a factor of 3 higher UFP concentrations in the city centre compared to the urban background and an additional factor of 2 higher concentrations in street canyons.

4.1.2 North America

In North America only measurements of PNCs are available. Annual averages of observed PNCs (PNC_O) at urban sites in the United States are taken from a study by Saha et al. (2021). The measurements were performed in different years, from 2009 to 2016. The results are summarized in Table 2. We note that the model again underestimates PNC_O at central urban stations, mostly by a factor ranging from 1.5 to 2.5.

Additional observations of PNC_O in the United States and Canada were obtained from the EBAS database providing daily measurements. They are compared to annual averages of the simulation for days with valid measurements, shown in Table 3. PNC_M in Bondville, Egbert and at the Appalachian State University (ASU) agree reasonably well with the ob-

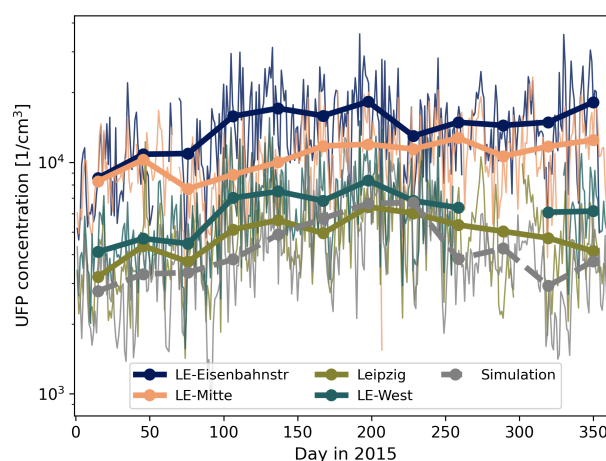


Figure 6. Monthly averaged UFP concentrations in the simulation (grey, dashed) and measured at different stations in Leipzig, Germany (thick, solid lines). The daily fluctuating UFP concentrations are shown with thin, transparent lines in the same colour.

Table 2. Summary of observations of PNCs (annual averages, cm^{-3}) in urban regions across the United States for different years taken from a study by Saha et al. (2021). The geographical location is indicated by latitude (Lat) and longitude (Long). The annual averages of observed (PNC_O) and modelled (PNC_M) PNCs (2015 for PNC_M , variable years for the observations) for the grid box encompassing the station are compared. The measurements applied a cut-off minimum diameter (Cut, in nanometres). The same cut-off was used to calculate simulated PNC_M . Simulated PNCs after the emission-sector-based redistribution are displayed as well (PNC_R), discussed in Sect. 4.2.

Location	Lat	Long	Year	Cut	PNC_O	PNC_M	PNC_R
Rochester (NY)	43.14	−77.54	2013	10	4450	3799	6294
Boston (MA)	42.33	−71.10	2016	10	12 200	4614	9169
Somerville (MA)	42.40	−71.09	2009	6	10 000	4687	8640
Queens (NY)	40.73	−73.82	2009	20	8210	4787	12 722
Long Island (NY)	40.74	−73.58	2009	20	7600	4787	9975
Livermore (CA)	37.69	−121.78	2012	7	8220	6090	6553
Red Wood (CA)	37.48	−122.20	2015	7	11 910	6090	9757
San Pablo (CA)	37.96	−122.36	2012	7	10 480	6090	12 119
Santa Rosa (CA)	38.44	−122.71	2012	7	8660	6090	11 526
Anaheim (CA)	33.83	−117.94	2016	7	13 950	8019	12 100
Central LA (CA)	34.07	−118.23	2016	7	17 780	8019	14 929
Compton (CA)	33.90	−118.21	2012	7	14 000	8019	14 916
Rubidoux (CA)	34.00	−117.42	2016	10	12 930	7975	11 858

Table 3. Summary of observations of PNC_O (cm^{-3}) across the United States and Canada in 2015, with data archived in the EBAS database. The geographical location is indicated by latitude (Lat) and longitude (Long), and the elevation of the site above the sea surface is indicated by the altitude a.s.l. in metres (Alt). The annual averages of PNC_O and PNC_M for the grid box encompassing the station are compared, and different measures of agreement are listed. The same comparisons are performed after the redistribution of the model (Redistributed model), which are discussed in Sect. 4.2. All included measurement stations contained at least 250 d of valid measurements in 2015. There is no particle size cut-off value given in the datasets, and thus none is applied on the simulation. Abbreviations: ASU – Appalachian State University, SGP – Southern Great Plains observatory.

Station	Observations				Model results				Redistributed model			
	Lat	Long	Alt	PNC_O	PNC_M	PF2	NRMSE	$\overline{M/O}$	PNC_R	PF2	NRMSE	$\overline{M/O}$
ASU, Boone (NC, US)	36.21	−81.69	1076	2930	2471	58	0.26	0.81	3317	55	0.3	1.09
Bondville (IL, US)	40.05	−88.37	213	4095	4130	55	0.26	0.96	4343	55	0.27	1.01
Egbert (ON, CA)	44.23	−79.78	255	4962	3238	53	0.26	0.68	2775	49	0.27	0.58
SGP E13 (OK, US)	36.60	−97.48	318	3600	6966	40	0.61	1.82	7457	38	0.67	1.95
Trinidad Head (CA, US)	41.05	−124.15	107	1526	2882	48	0.59	1.8	2800	49	0.57	1.75

servations with respect to the aforementioned criteria (see Sect. 4.1.1), while at the Southern Great Plains (SGP) observatory and at Trinidad Head the concentrations are overestimated by the model. The SGP site is located in the middle of wheat fields and pastures and is thereby efficiently shielded from major UFP sources in the encompassing grid box. The Trinidad Head measurement was performed directly at the Californian coast, and the corresponding grid box is influenced by a mixture of anthropogenic, rural and oceanic influences, leading to strong UFP gradients (comparable to Fionokalia in Europe).

4.1.3 India

Annually averaged UFP_O concentrations in Delhi (Thamban et al., 2021), Mahabaleshwar and Hyderabad (both from Sebastian et al., 2022) were obtained from a collaboration

with groups performing field measurements. Additionally, we adopted daily measurements of PSDs from Gani et al. (2020) for the Indian Institute of Technology Delhi (IITD), which we converted to UFP concentrations and combined with the measurements of Thamban et al. (2021). The evaluation for India is summarized in Table 4.

The simulation grid cell covering Delhi underestimates UFP_O concentrations at all three urban measurement stations in Delhi. The higher UFP_O concentrations over the IITD compared to the two other measurements may be due to its proximity to a major highway. In contrast, our simulations are biased high over Hyderabad. The measurement station in Hyderabad is a suburban university campus, approximately 15 km from the city centre, where UFP concentrations are expected to be significantly reduced compared to the downtown regions with strong traffic emissions. The consequent UFP gradients may not be adequately resolved by our model cal-

Table 4. Summary of UFP_O concentrations (cm⁻³) derived from PSDs in India for different years. The geographical location is indicated by latitude (Lat) and longitude (Long). The annual averages of UFP_O and UFP_M concentrations (2015 for UFP_M, variable years for the observations) for the grid box encompassing the station are compared. The measurements applied a cut-off minimum diameter (Cut, in nanometres). The same cut-off was used to calculate simulated UFP_M. Simulated UFP concentrations after the emission-sector-based redistribution are displayed as well (UFP_R), which are discussed in Sect. 4.2. Abbreviations: IITM – Institute of Information Technology & Management, IITD – Indian Institute of Technology Delhi, MRIU – Manav Rachna International University.

Station	Lat	Long	Year	Cut	UFP _O	UFP _M	UFP _R	Reference
Delhi IITM	28.61	77.1	2018/2019	14.1	19 750	15 339	28 789	Thamban et al. (2021)
Delhi IITD	28.55	77.19	2017/2018/2019	14.1	42 972	15 339	24 917	Thamban et al. (2021), Gani et al. (2020)
Delhi MRIU	28.45	77.28	2018/2019	14.1	17 350	15 339	15 959	Thamban et al. (2021)
Mahabaleshwar	17.92	73.66	2015	5.5	2441	2248	2316	Sebastian et al. (2022)
Hyderabad	17.46	78.32	2019/2020/2021	10	4680	6350	11 131	Sebastian et al. (2022)

culations (spatial representation error after Schutgens et al., 2016a).

4.1.4 China

Observations of PSDs in China were obtained through collaboration with relevant groups for Shanghai (unpublished), Beijing in 2018/19 (Liu et al., 2020), Lin'an (Shen et al., 2022) and Gucheng (Li et al., 2021). Additionally, we included observations from Beijing from 2004–2006 (Wu et al., 2008). We compare the UFP concentrations derived from our simulation to the PSD measurements from these sites. The results are summarized in Table 5.

UFP_M concentrations in Shanghai and Beijing are slightly lower (approximately 20 %) than UFP_O concentrations for the years closest to 2015 (2014 for Shanghai, 2018/19 for Beijing), unlike other urban locations discussed above. However, UFP_O concentrations in Beijing from 2004–2006 are underestimated by more than a factor of 2 by UFP_M. A probable reason for this is the various air pollution reduction measures in place in China, especially in Beijing. In Beijing, PM_{2.5} concentrations decreased from 89 µg cm⁻³ in 2013 to 58 µg cm⁻³ in 2017 and 42 µg cm⁻³ in 2019 (Zeng et al., 2019; Lu et al., 2020), implying a simultaneous reduction in UFP sources. Due to the air pollution reduction measures, industrial areas are also moved further away from city centres, potentially decreasing the association with UFP concentrations (see also Sect. 4.2).

UFP_M overestimates UFP_O strongly in Lin'an and slightly in Beijing's suburban Gucheng. Lin'an is influenced by the neighbouring city of Hangzhou with approximately 10 million inhabitants, Gucheng by sharing the grid box with central Beijing.

4.1.5 Remote regions

We considered remote measurements over the open ocean, in forests, on mountains and at polar sites, of both PNCs and UFP concentrations. The simulation and measurement results are summarized in Table 6. Simulated average con-

centrations are all within a factor of 2 of the observations. In particular, the northern hemispheric polar regions show good agreement, while there is a stronger overestimation and underestimation of UFP_O at the two measurement stations in Antarctica, both at elevated altitudes.

The annual average forest UFP_O concentrations at ATTO in Brazil (Franco et al., 2022) and Hyytiälä (EBAS database) are both underestimated by the simulation. The timeline analysis of the measurements in the Amazon forest (ATTO) shows this pronounced underestimation only in winter; i.e. UFP_M shows a stronger seasonality than UFP_O. The potential causes will be the subject of future studies, also considering data from an upcoming measurement campaign.

Measurements over the open ocean were taken from the ATom aircraft campaign (Brock et al., 2019), conducted in different seasons from 2016 to 2018. We collected all measurements that were performed below 200 m over the ocean and compared them to daily averaged UFP_M concentrations in the lowest vertical model level of the corresponding horizontal grid cell at the respective day of the year. UFP_O concentrations over the Southern Ocean are mostly underestimated by the simulation, while concentrations over the Pacific are overestimated. Considering generally higher UFP_M concentrations over the Pacific Ocean and lower UFP_M concentrations over the Southern Ocean (compare Fig. 3), the open-ocean UFP variance seems to be overestimated by the model.

4.1.6 Global

Figure 7 summarizes the evaluation results of all measurement stations presented and compares the annual averages of the observed to the simulated UFP concentrations in a scatterplot. The shapes of the symbols represent the five studied regions. Population density data (represented by the colour scheme in Fig. 7) are taken from CIESIN (2018), sampled at the measurement stations with a resolution of 0.1° × 0.1°.

The logarithms of UFP_O and UFP_M concentrations are correlated by $r = 0.93$ ($r = 0.76$ excluding remote regions), while the RMSLE (root mean square logarithmic error) is

Table 5. Summary of UFP_O concentrations (cm^{-3}) derived from PSDs in China for different years. The geographical location is indicated by latitude (Lat) and longitude (Long). The annual averages of UFP_O and UFP_M concentrations (2015 for the simulation, variable years for the observations) for the grid box encompassing the station are compared. The measurements applied a cut-off minimum diameter (Cut, in nanometres). The same cut-off was used to calculate simulated UFP_M . Simulated UFP concentrations after the emission-sector-based redistribution are displayed as well (UFP_R), which are discussed in Sect. 4.2.

Station	Lat	Long	Year	Cut	UFP_O	UFP_M	UFP_R	Reference
Shanghai	31.17	121.43	2014	13.4	12 800	10 850	23 623	Unpublished
Beijing	39.94	116.29	2018/2019	5.6	14 812	11 489	20 123	Liu et al. (2020)
Beijing	39.9	116.38	2004–2006	3.0	24 900	11 408	21 041	Wu et al. (2008)
Lin'an	30.28	119.75	2015	3.8	4928	11 815	12 447	Shen et al. (2022)
Gucheng	39.15	115.73	2018/2019	12.9	8284	10 678	9680	Li et al. (2021)

Table 6. Summary of UFP_O concentrations (cm^{-3}) and PNC_O in remote regions. Four different settings are distinguished, namely ocean, forest, mountain and polar. The geographical location is indicated by latitude (Lat) and longitude (Long), and the elevation of the site above the sea surface is indicated by the altitude a.s.l. in metres (Alt). The annual averages of the observed (Obs) and simulated (Mod) concentrations for the grid box encompassing the station are compared, and different measures of agreement are listed (see Table 6). We applied the measurement detection limit on the simulation for all UFP concentrations, while information on the detection limit was not available for the PNC measurements.

Station	Setting	Lat	Long	Alt	Meas	Obs	Mod	PF2	NRMSE	$\overline{M/O}$	Reference
ATom campaign	Ocean	–	–	< 200	UFP	403	800	–	–	–	Brock et al. (2019)
ATTO (Brazil)	Forest	–2.14	–59.0	120	UFP	376	199	–	–	–	Franco et al. (2022)
BEO Moussala (BG)	Mountain	42.18	23.59	2925	UFP	542	940	45	0.51	1.58	EBAS
Hyytiälä (FI)	Forest	61.85	24.3	179	UFP	1430	1140	59	0.21	0.77	EBAS
Sammaltunturi Pallas (FI)	Polar	67.97	24.12	565	UFP	556	489	43	0.24	0.71	EBAS
Trollhaugen (NO)	Polar	–72.02	2.53	1309	UFP	154	62	31	0.23	0.36	EBAS
Värriö (FI)	Polar	67.75	29.61	390	UFP	697	800	44	0.34	0.93	EBAS
Zeppelin mountain (NO)	Polar	79.9	11.86	473	UFP	161	185	40	0.42	0.55	EBAS
Zugspitze (DE)	Mountain	47.41	10.98	2650	UFP	944	655	57	0.22	0.65	EBAS
Alert (NU, CA)	Polar	82.50	–62.34	2182	PNC	205	279	47	0.26	1.48	EBAS
Barrow (AK, US)	Polar	71.32	–156.61	11	PNC	277	417	33	0.44	0.98	EBAS
South Pole	Polar	–90.00	–24.80	2841	PNC	192	238	56	0.21	1.5	EBAS

0.55.¹ The geometric mean of the ratio between the modelled and observed mean values is $\overline{M/O} = 0.82$.

In remote areas all simulated concentrations are within a factor of 2 of the observed ones. For these areas the population density (and respective anthropogenic emissions) within the encompassing grid box is mostly uniform at low numbers of inhabitants; i.e. the grid cells only cover remote areas. Thus, the grid cell average of UFP concentrations is not influenced by densely populated and typically much more polluted regions.

All other measurement stations considered have in common that the encompassing grid boxes include an urban city centre with its surroundings and thus have high variance in anthropogenic emissions within the grid box. Figure 7 indicates a link between local population density and UFP concentrations. In fact, the logarithm of the population density is positively correlated to the logarithm of the UFP_O con-

centrations in non-remote regions with $r = 0.80$. Thus, it can be expected that the actual UFP concentrations in grid boxes encompassing urban regions and its surroundings are non-uniform, with higher UFP concentrations at higher population density and lower UFP concentrations at lower population density. This inability of in situ observations to represent the grid box environment is defined as the spatial representation error and is studied by Schutgens et al. (2016a) and was found to be the strongest close to sources, in agreement with our evaluation results.

The spatial representation error is illustrated in Fig. 7. At low UFP_O concentrations in non-remote regions the local population density is lower (background, suburban and rural stations in grid boxes including urban centres), while UFP_M concentrations are higher than UFP_O concentrations ($\overline{M/O} = 1.32$ for a population density smaller than $100 \text{ individuals km}^{-2}$), as the model grid boxes are influenced by the urban regions. At higher UFP_O concentrations population density increases as well, and the simulation underestimates UFP_O concentrations on average by a factor of almost 2 ($\overline{M/O} = 0.62$ for a population density in excess

¹We use the correlation and RMSE of the logarithmic values here as several orders of magnitudes are covered, and the influence of the lower UFP concentrations would be negligible otherwise.

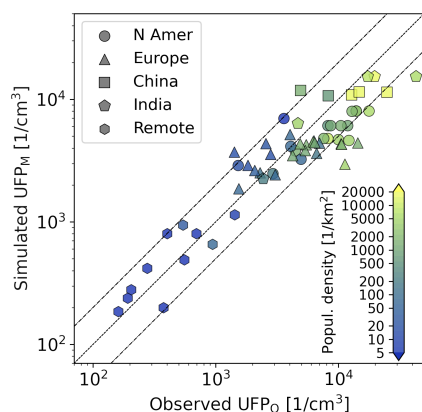


Figure 7. Summary of all measurement stations in a scatterplot showing observed UFP_O concentrations on the x axis and simulated UFP_M concentrations on the y axis. Different symbols indicate the different regions. The colour represents the local population density, sampled at the measurement stations with a resolution of $0.1^\circ \times 0.1^\circ$.

of 1000 individuals km^{-2}), as these urban stations are surrounded by suburban and rural regions that lower the simulation grid cell average of UFP_M concentrations.

The main cause of the spatial representation error is the limited model resolution of approx. 180×180 km (at the Equator). Consequently, the correlation between the logarithm of the local population density and UFP_M concentrations in non-remote regions is only $r = 0.57$, thus much lower than the correlation to UFP_O concentrations. To overcome this, an increase in model resolution of at least a factor of 3 (in one dimension) would be necessary, which would lead to an unreasonably high demand of computing time. Thus, an alternative approach was developed to mimic a resolution increase, by retroactively (after the simulation) redistributing UFP_M concentrations per grid box based on local high-resolution primary anthropogenic emissions relative to the grid box averaged primary anthropogenic emissions, guided by observations. This is discussed in the following section.

4.2 Downscaling based on primary anthropogenic emissions

Our results, in line with the observations, corroborate that UFP concentrations at the Earth's surface are strongly influenced by anthropogenic activity. Although our access to long-term measurements is limited, the data available from several urban stations show a high variance in UFP_O concentrations among different sites in a city (e.g. for Leipzig, Germany, and Los Angeles, the United States), being enhanced when model grid boxes contain urban and rural environments (e.g. Salma et al., 2014). Population density and UFP_O concentrations are highly correlated, while the eval-

uation of UFP_M concentrations indicates discrepancies due to sharp gradients of anthropogenic emissions within areas covered by the model grids.

Primary anthropogenic emissions (PAEs) from the CEDS database are available at a resolution (that is higher than the model resolution) of $0.5^\circ \times 0.5^\circ$ (hereafter referred to as $\text{PAE}_{\text{CEDS};0.5}$) and were regridded to the simulation mesh (resulting in grid box averaged emissions $\text{PAE}_{\text{CEDS};\text{GB}}$). Primary anthropogenic emissions from the Emissions Database for Global Atmospheric Research (EDGAR) v6.1 (Crippa et al., 2022) are available at an even higher resolution of $0.1^\circ \times 0.1^\circ$ (hereafter referred to as $\text{PAE}_{\text{EDGAR};0.1}$). Studies showed that locally enhanced UFP concentrations usually reach (urban) background levels within 1000 m from sources (e.g. Karner et al., 2010), and the curbside UFP concentrations are highly localized. Hence, this section aims to use local $\text{PAE}_{\text{CEDS};0.5}$ and $\text{PAE}_{\text{EDGAR};0.1}$ and the relation to the respective grid box average at coarser resolution to fine-tune the simulation results guided by the observations, gaining improved resolution (downscaling) and closer agreement with observations (reducing the spatial representation error), especially in urban centres and their surroundings. It is important to mention that we do not modify the total number of UFP_M concentrations per grid box in the following but that UFP_M concentrations are merely redistributed within each grid box.

Figure 9 illustrates the two-step downscaling procedure by the example of New Delhi and surroundings, using $\text{PAE}_{\text{CEDS};0.5}$ to downscale to $0.5^\circ \times 0.5^\circ$ and $\text{PAE}_{\text{EDGAR};0.1}$ for further downscaling, obtaining a UFP dataset with $0.1^\circ \times 0.1^\circ$ horizontal resolution. As a basis we calculate *relative anthropogenic emissions* (RAEs), i.e. the local anthropogenic particle number emissions relative to the average emissions of the grid box at coarser resolution:

$$\text{RAE}_{\text{CEDS}} = \text{PAE}_{\text{CEDS};0.5} / \text{PAE}_{\text{CEDS};\text{GB}},$$

$$\text{RAE}_{\text{EDGAR}} = \text{PAE}_{\text{EDGAR};0.1} / \text{PAE}_{\text{EDGAR};0.5}.$$

RAE_{CEDS} (top left in Fig. 9) and $\text{RAE}_{\text{EDGAR}}$ (top right in Fig. 9) can be interpreted as the local excess or deficit of $\text{PAE}_{\text{CEDS};0.5}$ ($\text{PAE}_{\text{EDGAR};0.1}$) over $\text{PAE}_{\text{CEDS};\text{GB}}$ ($\text{PAE}_{\text{EDGAR};0.5}$), respectively. We limit RAE_{CEDS} to a maximum value of 10 and $\text{RAE}_{\text{EDGAR}}$ to 7 due to missing observational datasets at locations with higher RAE.

In the first step we investigated the relationship between RAE_{CEDS} and the underestimation of UFP_O concentrations and PNC_O at each station ($\text{UFP}_O / \text{UFP}_M$ or $\text{PNC}_O / \text{PNC}_M$) for all evaluation results in grid boxes with anthropogenic emissions exceeding two million particles per square metre and second.² The relationship is displayed in Fig. 8a. As expected, there is a logarithmic correlation ($r = 0.42$) between the two quantities. We perform a linear fit that crosses the

²All high-resolution pixels in the remaining (less anthropogenically influenced) grid boxes are bilinearly interpolated with respect to the coarser resolution.

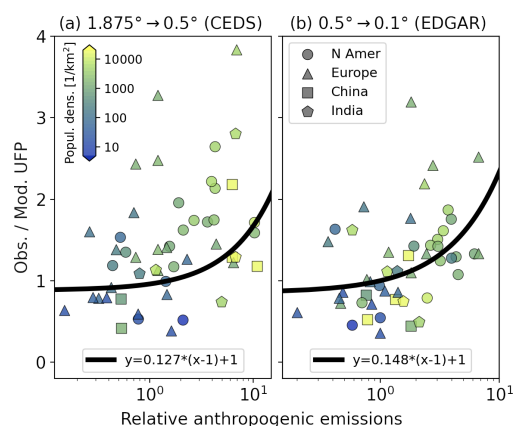


Figure 8. Scatterplot of the relative anthropogenic emissions (RAEs) against the model–measurement discrepancy (observed and simulated UFP concentrations). The RAEs of the CEDS database relative to the model grid box are directly compared to the model–measurement discrepancy (a), while the RAEs of CEDS relative to the CEDS grid box are compared to the remaining discrepancy after the first downscaling (b). We use the same colour code for the population density and shapes for the regions as in Fig. 7. The black lines show a linear fit crossing the (1,1) point. Higher RAEs correspond to a more pronounced underestimation of the observations.

point (1,1); i.e. $y = c_{\text{CEDS}} \cdot (x - 1) + 1$, where y is the ratio $N_{\text{obs}}/N_{\text{mod}}$ and x is the RAE_{CEDS} . This function was chosen as it is the only global function that conserves the average grid box UFP_{M} concentrations after applying it to the model results. The fit parameter c was determined to be $c_{\text{CEDS}} = 0.127$ using a logarithmic least squares fit. Thus, if RAE_{CEDS} increases by 1, the model underestimation increases by 0.127. The fit function is shown as a black line in Fig. 8a. This relationship is used to downscale simulated UFP_{M} concentrations, yielding redistributed UFP ($\text{UFP}_{0.5}$) concentrations.

This procedure is repeated for the relationship between $\text{RAE}_{\text{EDGAR}}$ and the remaining model–observation discrepancies after the first downscaling (Fig. 8b). We determine c_{EDGAR} to be $c = 0.148$, and thus $\text{UFP}_{0.5}$ can be further downscaled to the urban and industrial environments, yielding redistributed $\text{UFP}_{0.1}$ (hereafter referred to as UFP_{R}) concentrations (rightmost plot in Fig. 9). Summarized, we obtain two downscaling functions that are applied sequentially to simulated UFP_{M} concentrations:

$$\text{UFP}_{0.5} = \text{UFP}_{\text{M}} \cdot (0.127 \cdot (\text{RAE}_{\text{CEDS}} - 1) + 1),$$

$$\text{UFP}_{\text{R}} = \text{UFP}_{0.5} \cdot (0.148 \cdot (\text{RAE}_{\text{EDGAR}} - 1) + 1).$$

To demonstrate the independence of the downscaling procedure from the data used for the fit, we randomly subdivided the non-remote measurement stations into a training and test dataset (25 for training, 24 for testing) in 5000 different random ways, derived the fit parameters from the training dataset only, and subsequently applied them to the test

dataset. The results are displayed in Fig. 10. The fit parameters c_{CEDS} and c_{EDGAR} range between 0.07 and 0.23 in 90 % of the runs, while the average values are very close to the values derived from the complete dataset, and all derived fit parameters are greater than 0. The RMSLE consistently decreases with rising order of downscaling, with an average improvement of 0.11 in total (95.6 % of the time RMSLE decreases), while the logarithmic correlation increases on average by 0.07 (improves 95.1 % of the time). The bias $\overline{M/O}$ evolves from 0.78 on average in the model output resolution (2.4 % of the time between 0.9 and 1.1) to 0.92 after the CEDS downscaling (55.3 % of the time between 0.9 and 1.1) to 1.04 (66.2 % of the time between 0.9 and 1.1) after the final downscaling, slightly overestimating the observed values. We conclude that the fit parameters can be applied to data points outside the training dataset. However, as we want to maximize the number of measurement stations to use for the global downscaling, we use the complete evaluation results for the downscaling as described before.

The resulting UFP_{R} concentrations are included in Tables 1–5 in the last columns. At measurement sites in Europe, urban regions in the United States, India and China the agreement with measurements generally improves. For instance, UFP_{R} concentrations in Madrid, the measurement station with the strongest underestimation in this analysis (triangle in the top-right corner in Fig. 8a), are increased by a factor of 2.1, strongly improving the agreement (see Table 1: PF2 from 22 % to 59 %, $\overline{M/O}$ from 0.28 to 0.59). The remaining underestimation is likely caused by the influence of nearby roads with heavy traffic, which are of localized relevance only.

UFP_{R} in Beijing and Shanghai is strongly increased by the redistribution as well, which can even lead to an overestimation of the observed UFP_{O} concentrations (see Table 5). However, for Beijing the redistributed UFP_{R} falls between the high UFP_{O} from 2004–2006 and the lower UFP_{O} from 2018/19. This is in line with the emission reduction in China (see Sect. 4.1.4). UFP_{R} concentrations over Beijing’s suburban Gucheng, sharing the model grid box with Beijing, are reduced, approaching UFP_{O} concentrations.

Similarly, UFP_{R} concentrations are strongly increased at the measurement stations Delhi IITM and Delhi IITD, resulting in UFP_{R} concentrations in between the two UFP_{O} concentrations. Moreover, UFP_{R} is only slightly increased at Delhi MRIU due to the lower local PAE_{HR} , remaining similar to UFP_{O} .

The left panel of Fig. 11 shows the comparison between UFP_{R} and UFP_{O} concentrations (analogously to Fig. 7) after the applied redistribution. In spite of the redistribution of UFPs within grid cells, there is still an overestimation of $\overline{M/O} = 1.27$ (1.32 before) at low population density below 100 individuals km^{-2} . On the other hand, in densely populated regions (more than 1000 individuals km^{-2}), simulated and observed UFP concentrations are of the same magnitude ($\overline{M/O} = 1.01$). Thus, the biases described in Sect. 4.1.6 are

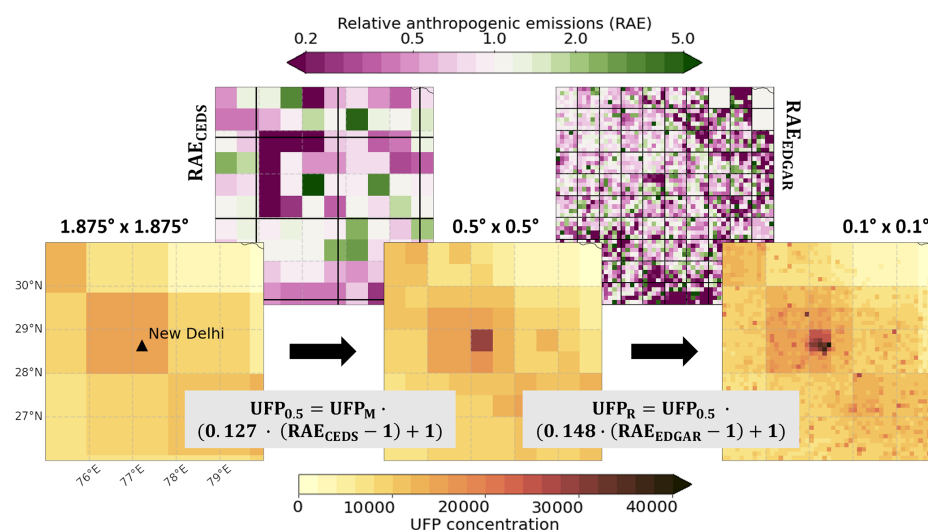


Figure 9. Illustration of the downscaling procedure using anthropogenic particle number emissions from the CEDS (Hoesly et al., 2018) and EDGAR (Crippa et al., 2022) emission databases, using the example of New Delhi and surroundings. Relative anthropogenic emissions (RAEs; local anthropogenic particle number emissions relative to the average over the initial grid box, displayed on the top) are set in relation to the model–observation discrepancy (observed and modelled UFP concentrations), displayed in Fig. 8. The resulting relation is used for downscaling UFP concentrations from the model resolution ($1.875^\circ \times 1.875^\circ$) to $0.5^\circ \times 0.5^\circ$ (CEDS) and $0.1^\circ \times 0.1^\circ$ (EDGAR) horizontal resolutions in two steps. The average of the RAE over each grid box of the coarser resolution (black boxes) equals 1, and thus the respective average UFP concentrations per grid box remain unchanged. Note that the colour scale for UFP concentrations differs from Fig. 3.

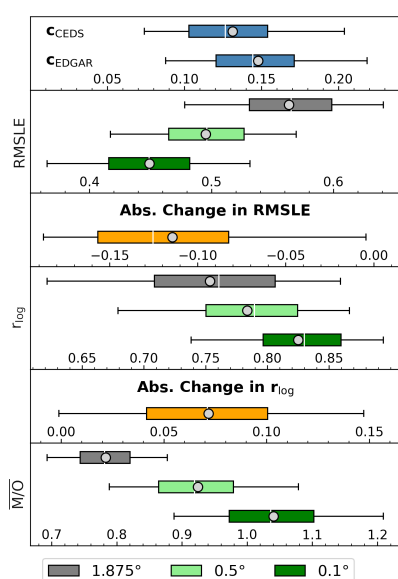


Figure 10. Results of 5000 independent training and test runs for the fit parameter estimation (25 stations in training dataset, 24 in test dataset). The fit parameters are displayed in the top panel, while the statistical measures are displayed below at different orders of downscaling, along with the absolute change in RMSLE and the logarithmic correlation. We show box-and-whisker plots, where the boxes illustrate the 25 %–75 % interval and the whiskers the 5 %–95 % interval. The median values are marked by the vertical white line and mean values by the light grey circles.

strongly reduced though not fully eliminated, especially for scarcely populated regions.

Evaluation results before and after the downscaling are displayed in the right panel of Fig. 11. The strong improvement in municipal regions is apparent when considering the urban measurement stations in the United States (top right), aligning closely around the centre line. An increase in correlation is also clearly visible in China, Europe and India.

All UFP_R concentrations differing by more than a factor of 2 from UFP_O can be attributed to nearby major traffic nodes and motorways (Leipzig-Eisenbahnstrasse) or alternatively coastal sites (Finokalia), a shielded location in a wheat field (Southern Great Plains E13) and the influence of a neighbouring megacity (Lin'an and Hyderabad). These discrepancies between UFP_R and UFP_O may perhaps be resolved with very high (< 1 km) horizontal resolution, which is computationally impracticable with a global model and cannot be achieved with downscaling due to missing emission datasets with respective horizontal resolution.

Finally, the logarithmic correlation between population density and UFP_R concentrations increases to $r = 0.77$ ($r = 0.57$ for UFP_M) after redistribution, which is similar to the logarithmic correlation between population density and UFP_O concentrations ($r = 0.80$). This indicates an improved representation of the anthropogenic influence in the down-scaled dataset.

Figure 12 shows UFP_R concentrations in Asia after the applied downscaling, revealing more detailed features in

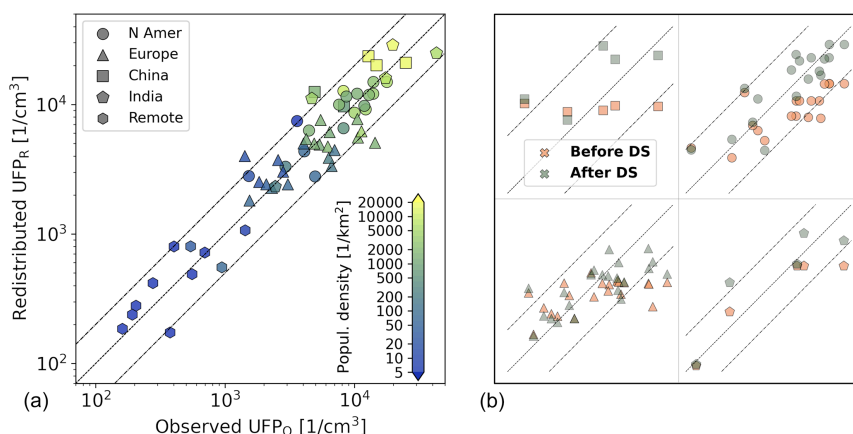


Figure 11. (a) The same as Fig. 7 but after the redistribution of UFP_M concentrations (downscaling) based on the relative anthropogenic emissions per grid box (more details in the text). (b) Evaluation results before and after the downscaling (DS) for China, North America, Europe and India (from top left to bottom right). Note that the value range differs in all subpanels.

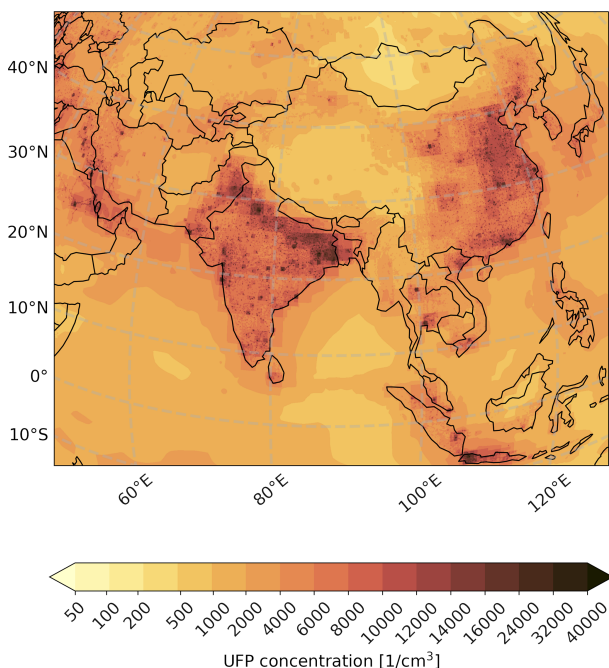


Figure 12. Annual average of UFP_R concentrations in Asia simulated with EMAC for the year 2015 after the downscaling based on primary anthropogenic emissions at a resolution of $0.1^\circ \times 0.1^\circ$. Note that the colour scale from Fig. 7 is extended as UFP_R concentrations can exceed $40\,000\text{ cm}^{-3}$.

anthropogenically influenced regions, especially in eastern China and northern India. Note that the colour scale is extended as UFP_R concentrations can exceed $40\,000\text{ cm}^{-3}$ in Indian industrial regions, as well as in parts of urban environments in Mumbai, New Delhi, Shanghai, Riyadh, Kuwait and Cairo.

5 Limitations and uncertainties

As the resulting UFP_M and UFP_R concentrations are an intricate interplay of emission parameters, numerical simulation, evaluation and observation-guided redistribution, it is not possible to directly infer quantitative uncertainties for the provided datasets. However, the final results in Fig. 11 show that 90 % of the annually averaged UFP_R concentrations are within a factor of 2 of UFP_O concentrations (84 % for UFP_M), and all simulated UFP_R concentrations are within a factor of 3 of the observations after the emission-grid-based redistribution. Additional limitations and uncertainties are qualitatively discussed next.

Median diameters of directly emitted particles per sector were estimated based on the emission size distributions from Paasonen et al. (2016) and measurement reports in the literature with associated uncertainties. Additionally, we assumed the diameters to be globally identical for each sector and used the same diameters for all species. These simplifications lead to limitations in the precision by which the different sectors and species contribute to the total UFP_M and UFP_R number, respectively. Anthropogenic emissions are distributed in each time step based on monthly averages from the emission datasets. This potentially introduces bias due to the non-linearity of chemical and microphysical processes. Future studies will include emission time factors to improve the time resolution of emissions.

Apart from UFP concentrations, we also used PNCs for the evaluation. PNCs are a proxy for UFP concentrations, as UFPs tend to dominate the total particle number (e.g. Baldauf et al., 2016; Kumar et al., 2014). However, locally there can be deviations. To increase the number of measurements for the evaluation we additionally used observations from years that differ from the simulation year (2015). Different meteorological conditions and emissions potentially lead to

biases in the evaluation. The downscaling was performed using evaluation results from differing years as well, while the simulation results were only taken from 2015. Thus, potential errors arise from the interference of the downscaling and general temporal trends, as seen from the analysis of the interannual variability in Sect. 4. The interannual variability (up to 50 % at low absolute UFP concentrations and below 20 % over polluted regions over a time span of 3 years) has been attributed to meteorological variations (along the ITCZ and mostly over the oceans) and decreasing (southern and eastern Asia, Europe, and the United States) or increasing (Nigeria and South Africa) long-term trends.

We define UFPs as all particles with a diameter smaller than 100 nm in the model without a lower cut on the particle diameter. Particles with a diameter below 1.7 nm are typically regarded as molecular clusters, and nucleation rates are reported as the frequency that clusters reach this threshold (e.g. by the CLOUD experiment; Kirkby et al., 2011). As a consequence, freshly formed particles are introduced to the model with a diameter of 1.7 nm. However, due to the lognormal modal aerosol distribution (Pringle et al., 2010), smaller particles down to infinitesimal small diameters are automatically created by definition. As a result, small fractions (up to around 2 % in polluted regions, with maximum values of up to 20 % locally over the oceans) of the presented UFPs have a diameter smaller than 1.7 nm. However, these particles are not considered to be molecular clusters as the low diameter is a result of computational effects.

In addition to the uncertainties introduced by the horizontal resolution of the model, the vertical resolution of the model, ranging from 45–70 m for the surface layer, adds additional uncertainties. While the surface layer is mostly entirely located in the boundary layer, decreases in UFP concentrations are still observed in general, as reported by Du et al. (2017), Harrison et al. (2019), Heintzenberg et al. (2011) and Villa et al. (2017), with the strongest decrease with altitude being observed close to UFP sources. We analysed the relative change in UFP concentrations from the surface layer to the next highest model level, exhibiting a decrease of 20 %–30 % in UFP concentrations on average over polluted regions, with the reduction slightly exceeding 50 % over central Africa. At higher latitudes, we even see increasing UFP concentrations at the second lowest model level. The decrease over polluted regions is in line with the observations from Du et al. (2017), Harrison et al. (2019) and Heintzenberg et al. (2011), who observe a decrease of up to 30 % at comparable altitudes.³ Considering a linear decrease and increase within the range of 30 % within the model sur-

face layer, this adds an additional uncertainty of ± 15 % to the simulated UFP concentrations.

Finally, an important uncertainty is the spatio-temporal representativeness of the observations with respect to the model grid box and time sampling. We report three different types of representation errors:

1. purely spatial representation errors only due to the extent of the grid box at stations, for which timeline measurements of 2015 (mostly hourly) were available, and we collocated our simulation with the observations according to Schutgens et al. (2016b), i.e. all observations from the EBAS database;
2. spatio-temporal representation errors for measurement stations, for which only annual averages were available or the years differed from the evaluation year (2015); and
3. (spatio-)temporal representation errors for the aircraft measurements from ATom, for which we used daily averaged model output, and the measurement years differ from the simulation year. The spatial representation error is reduced by the fact that we have several measurements for each grid box.

The horizontal downscaling of UFP concentrations addresses and reduces the spatial representation error for types 1 and 2, while the temporal representation error of types 2 and 3 cannot be addressed.

The analysis in Sect. 4.2 showed that even after downscaling discrepancies in the evaluation remain, which are at least partly related to the representativeness of the measurement locations for the modelled grid areas (Schutgens et al., 2017, 2016a), also for UFP_R . Model concentrations of UFPs in rural regions in the vicinity of urban centres tend to be overestimated by the model (i.e. UFP_R). Hence, even for UFP_R the horizontal resolution is still a limiting factor. To further reduce the spatial representation error, the simulations would need to be performed at very high resolution requiring currently impracticable computing resources, at least for the global scale, as well as additional measurement data for UFPs and meteorological parameters in urban and industrialized regions including roadsides and background urban environments. Salma et al. (2014) showed that UFP concentrations can typically vary by a factor of 6 from the urban background to street canopies within cities, and Karner et al. (2010) showed that UFP concentrations reach (urban) background concentrations within about a kilometre. A next step could be to apply high-resolution dynamical downscaling of concentrations guided by comprehensive measurements of which data may become available in the future, using either machine learning methods or a combination of operational street pollution models and human exposure modelling as done by Ketzel et al. (2021). Clearly, to make progress, a much larger number of stations that continuously measure aerosol size distributions will need to be implemented.

³Villa et al. (2017) even observe a decrease of up to 80 %; however these measurements were performed directly at a highway and at some distance from other sources, which is not representative of a grid box covering approx. 180×180 km and overestimates the effect.

6 Conclusions

We presented the first numerical simulation of ultrafine particles (UFPs) at the Earth's surface with the global EMAC model, which includes a relatively detailed representation of aerosol formation and growth processes (i.e. the nucleation and Aitken aerosol size modes). Total concentrations were shown, along with interannual variability in the years 2015–2017 and the dominating particle size segments around the globe. Emissions of gaseous and aerosol species were taken from the CEDS and CAMS databases, and the emission radii for aerosol species were taken or derived from the literature referring to the contributing source sectors.

Simulated UFP and particle number concentrations (PNCs) were evaluated using particle size distributions and PNCs from field measurements, the EBAS database, literature, and published datasets. We generally achieve reasonable agreement between observed and simulated UFP concentrations, with good agreement for remote regions (forest, mountain, polar and ocean) that are not directly influenced by urban and other strong source regions (all simulated concentrations within a factor of 2 of the observations). In grid boxes with a high variance in population density, we obtain larger deviations with observations related to the coarse model resolution (approximately 180×180 km at the Equator). UFP concentrations in urban regions with a high population density are underestimated by the simulation, while they are overestimated in less densely populated regions. This representation error is associated with the high correlation between local population density and observed UFP concentrations (logarithmic correlation of $r = 0.80$) and is studied in depth by Schutgens et al. (2016a, b, 2017).

The relationship between the underestimation of the observed UFP concentrations and the local high-resolution anthropogenic emissions relative to the grid box average was used to redistribute UFPs within each grid box, leaving the total number of particles unchanged. This yields a higher resolution, i.e. downscaled data for grid boxes with dominant anthropogenic influence; increases the agreement between observations and simulations (the logarithmic correlation improves from 0.76 to 0.84 for non-remote regions and root mean squared logarithmic error from 0.57 to 0.43); and decreases the spatial representation error by improving the representation of the anthropogenic impact on UFP concentrations.

We provide two global annually averaged datasets of UFP concentrations for the year 2015 at different horizontal resolutions (<https://doi.org/10.17617/3.7945XI>, Kohl and Pozzer, 2023) that can be used for several purposes. The first dataset is given at a resolution of $1.875^\circ \times 1.875^\circ$ (roughly 180×180 km at the Equator) and is directly derived from the simulation. This dataset can serve the purpose of global-scale analyses of UFP concentrations, e.g. the comparison of different source regions, meteorology and atmospheric chemistry. The downscaled dataset has a much finer resolution

of $0.1^\circ \times 0.1^\circ$ (roughly 10×10 km at the Equator and about 10×8 km at mid-latitudes) and includes the within-grid-box UFP redistribution based on anthropogenic emission data. The latter is recommended to be used to characterize the exposure to UFPs in public health studies with a focus on densely populated regions, in particular the urban environment. Additionally, we make the 2015 UFP concentrations available in three size bins (1.7–20, 20–50 and 50–100 nm).

Future applications may also include studies on seasonality, anthropogenic source sectors and the chemical composition of UFPs and their contribution to the health impacts of fine particulate matter.

Code availability. The Modular Earth Submodel System (MESSy) is continuously further developed and applied by a consortium of institutions. The usage of MESSy and access to the source code are licensed to all affiliates of institutions that are members of the MESSy Consortium. Institutions can become a member of the MESSy Consortium by signing the MESSy Memorandum of Understanding. More information can be found on the MESSy Consortium website (<http://www.messy-interface.org>, last access: 17 February 2023). The code presented here is available as git commit #49a7a544 in the MESSy repository, and all changes have been included in the main repository.

Scientific colour maps (<https://doi.org/10.5281/zenodo.5501399>, Crameri, 2021) are used in this study to prevent visual distortion of the data and exclusion of readers with colour vision deficiencies (Crameri et al., 2020).

Data availability. We provide datasets with annual averages of UFP concentrations for the year 2015, both in model resolution ($1.875^\circ \times 1.875^\circ$) directly derived from the model output, and at a resolution of $0.1^\circ \times 0.1^\circ$ with the observation-guided downscaling based on anthropogenic emissions. Additionally, we provide number concentrations in three size bins, i.e. 1.7–20, 20–50 and 50–100 nm. The datasets are publicly available at <https://doi.org/10.17617/3.7945XI> (Kohl and Pozzer, 2023).

Emission datasets from CEDS and EDGAR are available at <https://doi.org/10.5281/zenodo.3754964> (McDuffie et al., 2020b) and https://edgar.jrc.ec.europa.eu/index.php/dataset_ap61 (Crippa et al., 2022), respectively. Emission size distributions are available at <https://previous.iiasa.ac.at/web/home/research/researchPrograms/air/PN.html> (Paasonen, 2016). Gridded Population of the World (v4) is taken from <https://doi.org/10.7927/H4F47M65> (CIESIN, 2018). Measurement data from the ATom aircraft campaign are taken from <https://doi.org/10.3334/ORNLDAAAC/1671> (Brock et al., 2019).

Author contributions. SC, AP and JL planned the research. AP prepared the model set-up and performed the simulation with the help of MK and SC. DS, YC, SNT, MS, GP and HW helped in collecting and/or directly provided the observational datasets. SE developed the MESSy submodels NAN and IONS. MK analysed the model results, evaluated the simulation, developed the downscaling procedure and wrote the paper with the help of AP. JL and AP super-

vised the project. All authors discussed the results and contributed to the review and editing of the paper.

Competing interests. At least one of the (co-)authors is a member of the editorial board of *Atmospheric Chemistry and Physics*. The peer-review process was guided by an independent editor, and the authors also have no other competing interests to declare.

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References

- ACTRIS: ACTRIS Recommendation for mobility particle size spectrometer measurements, ACTRIS, <https://www.actris-ecac.eu/pnsd-10-to-800nm.html> (last access: 17 February 2023), 2021.
- Amann, M., Bertok, I., Borken-Kleefeld, J., Cofala, J., Heyes, C., Höglund-Isaksson, L., Klimont, Z., Nguyen, B., Posch, M., Rafaj, P., Sandler, R., Schöpp, W., Wagner, F., and Winiwarter, W.: Cost-effective control of air quality and greenhouse gases in Europe: Modeling and policy applications, *Environ. Model. Softw.*, 26, 1489–1501, <https://doi.org/10.1016/j.envsoft.2011.07.012>, 2011.
- Andreae, M. O.: Correlation between cloud condensation nuclei concentration and aerosol optical thickness in remote and polluted regions, *Atmos. Chem. Phys.*, 9, 543–556, <https://doi.org/10.5194/acp-9-543-2009>, 2009.
- Andreae, M. O.: Emission of trace gases and aerosols from biomass burning – an updated assessment, *Atmos. Chem. Phys.*, 19, 8523–8546, <https://doi.org/10.5194/acp-19-8523-2019>, 2019.
- Andreae, M. O., Afchine, A., Albrecht, R., Holanda, B. A., Artaxo, P., Barbosa, H. M. J., Borrmann, S., Cecchini, M. A., Costa, A., Dollner, M., Fütterer, D., Järvinen, E., Jurkat, T., Klimach, T., Konemann, T., Knote, C., Krämer, M., Krisna, T., Machado, L. A. T., Mertes, S., Minikin, A., Pöhlker, C., Pöhlker, M. L., Pöschl, U., Rosenfeld, D., Sauer, D., Schlager, H., Schnaiter, M., Schneider, J., Schulz, C., Spanu, A., Sperling, V. B., Voigt, C., Walser, A., Wang, J., Weinzierl, B., Wendisch, M., and Ziereis, H.: Aerosol characteristics and particle production in the upper troposphere over the Amazon Basin, *Atmos. Chem. Phys.*, 18, 921–961, <https://doi.org/10.5194/acp-18-921-2018>, 2018.
- Baldauf, R. W., Devlin, R. B., Gehr, P., Giannelli, R., Hassett-Sipple, B., Jung, H., Martini, G., McDonald, J., Sacks, J. D., and Walker, K.: Ultrafine Particle Metrics and Research Considerations: Review of the 2015 UFP Workshop, *Int. J. Environ. Res. Pu.*, 13, 1054, <https://doi.org/10.3390/ijerph13111054>, 2016.
- Bellouin, N., Quaas, J., Gryspeerdt, E., Kinne, S., Stier, P., Watson-Parris, D., Boucher, O., Carslaw, K. S., Christensen, M., Daniau, A.-L., Dufresne, J.-L., Feingold, G., Fiedler, S., Forster, P., Gettelman, A., Haywood, J. M., Lohmann, U., Malavelle, F., Mauritsen, T., McCoy, D. T., Myhre, G., Mülmenstädt, J., Neubauer, D., Possner, A., Rugenstein, M., Sato, Y., Schulz, M., Schwartz, S. E., Sourdeval, O., Storelvmo, T., Toll, V., Winker, D., and Stevens, B.: Bounding Global Aerosol Radiative Forcing of Climate Change, *Rev. Geophys.*, 58, e2019RG000660, <https://doi.org/10.1029/2019RG000660>, 2020.
- Berrisford, P., Dee, D., Poli, P., Brugge, R., Fielding, M., Fuentes, M., Källberg, P., Kobayashi, S., Uppala, S., and Simmons, A.: The ERA-Interim archive Version 2.0, Tech. Rep. 1, ECMWF, Shinfield Park, Reading, <https://www.ecmwf.int/node/8174> (last access: 17 February 2023), 2011.
- Brock, C., Kupc, A., Williamson, C., Froyd, K., Erdesz, F., Murphy, D., Schill, G., Gesler, D., McLaughlin, R., Richardson, M., Wagner, N., and Wilson, J.: ATom: L2 In Situ Measurements of Aerosol Microphysical Properties (AMP), ORNL Distributed Active Archive Center [data set], <https://doi.org/10.3334/ORNLDAAAC/1671>, 2019.
- Burnett, R. T., Pope, C. A., Ezzati, M., Olives, C., Lim, S. S., Mehta, S., Shin, H. H., Singh, G., Hubbell, B., Brauer, M., Anderson, H. R., Smith, K. R., Balmes, J. R., Bruce, N. G., Kan, H., Laden, F., Prüss-Ustün, A., Turner, M. C., Gapstur, S. M., Diver, W. R., and Cohen, A.: An Integrated Risk Function for Estimating the Global Burden of Disease Attributable to Ambient Fine Particulate Matter Exposure, *Environ. Health Persp.*, 122, 397–403, <https://doi.org/10.1289/ehp.1307049>, 2014.
- CEN/TC 264/WG 32 – Air quality – Determination of the particle number concentration: CEN/TS 16976:2016: Ambient air – Determination of the particle number concentration of atmospheric aerosol, iTeh Stan-

- dards, <https://standards.iteh.ai/catalog/standards/cen/91f1ac67-f6d6-408c-af89-e81763194fd3/cen-ts-16976-2016> (last access: 17 February 2023), 2016.
- CEN/TC 264/WG 32 – Air quality – Determination of the particle number concentration: CEN/TS 17434:2020: Ambient air – Determination of the particle number size distribution of atmospheric aerosol using a Mobility Particle Size Spectrometer (MPSS), iTeh Standards, <https://standards.iteh.ai/catalog/standards/cen/a841bc08-ed34-4fa8-94ca-8c5e07b99db9/cen-ts-17434-2020> (last access: 17 February 2023), 2020.
- Chen, X., Yu, F., Yang, W., Sun, Y., Chen, H., Du, W., Zhao, J., Wei, Y., Wei, L., Du, H., Wang, Z., Wu, Q., Li, J., An, J., and Wang, Z.: Global-regional nested simulation of particle number concentration by combining microphysical processes with an evolving organic aerosol module, *Atmos. Chem. Phys.*, 21, 9343–9366, <https://doi.org/10.5194/acp-21-9343-2021>, 2021.
- Chowdhury, S., Pozzer, A., Haines, A., Klingmüller, K., Münzel, T., Paasonen, P., Sharma, A., Venkataraman, C., and Lelieveld, J.: Global health burden of ambient PM_{2.5} and the contribution of anthropogenic black carbon and organic aerosols, *Environ. Int.*, 159, 107020, <https://doi.org/10.1016/j.envint.2021.107020>, 2022.
- Christensen, M. W., Jones, W. K., and Stier, P.: Aerosols enhance cloud lifetime and brightness along the stratus-to-cumulus transition, *P. Natl. Acad. Sci. USA*, 117, 17591–17598, <https://doi.org/10.1073/pnas.1921231117>, 2020.
- CIESIN (Center for International Earth Science Information Network): Gridded Population of the World, Version 4 (GPWv4): Population Density Adjusted to Match 2015 Revision UN WPP Country Totals, Revision 11, Center for International Earth Science Information Network [data set], <https://doi.org/10.7927/H4F47M65>, 2018.
- Cohen, A. J., Ross Anderson, H., Ostro, B., Pandey, K. D., Krzyzanowski, M., Künzli, N., Gutschmidt, K., Pope, A., Romieu, I., Samet, J. M., and Smith, K.: The global burden of disease due to outdoor air pollution, *J. Toxicol. Env. Heal. A*, 68, 1301–1307, <https://doi.org/10.1080/15287390590936166>, 2005.
- Cramer, F.: Scientific colour maps, Zenodo [code], <https://doi.org/10.5281/zenodo.5501399>, 2021.
- Cramer, F., Shephard, G. E., and Heron, P. J.: The misuse of colour in science communication, *Nat. Commun.*, 11, 1–10, <https://doi.org/10.1038/s41467-020-19160-7>, 2020.
- Crippa, M., Solazzo, E., Huang, G., Guizzardi, D., Koffi, E., Muntean, M., Schieberle, C., Friedrich, R., and Janssens-Maenhout, G.: High resolution temporal profiles in the Emissions Database for Global Atmospheric Research, *Scientific Data*, 7, 1–17, <https://doi.org/10.1038/s41597-020-0462-2>, 2020.
- Crippa, M., Guizzardi, D., Muntean, M., Schaaf, E., Monforti-Ferrario, F., Banja, M., Pagani, F., and Solazzo, E.: EDGAR v6.1 global air pollutant emissions, European Commission [data set], https://edgar.jrc.ec.europa.eu/index.php/dataset_ap61 (last access: 17 February 2023), 2022.
- Curtius, J.: Nucleation of atmospheric aerosol particles, *C. R. Phys.*, 7, 1027–1045, <https://doi.org/10.1016/j.crhy.2006.10.018>, 2006.
- d’Almeida, G. A.: On the variability of desert aerosol radiative characteristics, *J. Geophys. Res.-Atmos.*, 92, 3017–3026, <https://doi.org/10.1029/JD092iD03p03017>, 1987.
- Delfino, R. J., Sioutas, C., and Malik, S.: Potential Role of Ultrafine Particles in Associations between Airborne Particle Mass and Cardiovascular Health, *Environ. Health Persp.*, 113, 934–946, <https://doi.org/10.1289/ehp.7938>, 2005.
- Dentener, F., Kinne, S., Bond, T., Boucher, O., Cofala, J., Geroso, S., Ginoux, P., Gong, S., Hoelzemann, J. J., Ito, A., Marelli, L., Penner, J. E., Putaud, J.-P., Textor, C., Schulz, M., van der Werf, G. R., and Wilson, J.: Emissions of primary aerosol and precursor gases in the years 2000 and 1750 prescribed data-sets for AeroCom, *Atmos. Chem. Phys.*, 6, 4321–4344, <https://doi.org/10.5194/acp-6-4321-2006>, 2006.
- Diesch, J.-M., Drewnick, F., Klimach, T., and Borrmann, S.: Investigation of gaseous and particulate emissions from various marine vessel types measured on the banks of the Elbe in Northern Germany, *Atmos. Chem. Phys.*, 13, 3603–3618, <https://doi.org/10.5194/acp-13-3603-2013>, 2013.
- Downward, G. S., van Nunen, E. J., Kerckhoffs, J., Vineis, P., Brunekreef, B., Boer, J. M., Messier, K. P., Roy, A., Verschuren, W. M. M., van der Schouw, Y. T., Sluijs, I., Gulliver, J., Hoek, G., and Vermeulen, R.: Long-term exposure to ultrafine particles and incidence of cardiovascular and cerebrovascular disease in a prospective study of a Dutch cohort, *Environ. Health Persp.*, 126, 127007, <https://doi.org/10.1289/EHP3047>, 2018.
- Du, W., Zhao, J., Wang, Y., Zhang, Y., Wang, Q., Xu, W., Chen, C., Han, T., Zhang, F., Li, Z., Fu, P., Li, J., Wang, Z., and Sun, Y.: Simultaneous measurements of particle number size distributions at ground level and 260 m on a meteorological tower in urban Beijing, China, *Atmos. Chem. Phys.*, 17, 6797–6811, <https://doi.org/10.5194/acp-17-6797-2017>, 2017.
- Dunne, E. M., Gordon, H., Kürten, A., Almeida, J., Duplissy, J., Williamson, C., Ortega, I. K., Pringle, K. J., Adamov, A., Baltensperger, U., Barmet, P., Benduhn, F., Bianchi, F., Breitenlechner, M., Clarke, A., Curtius, J., Dommen, J., Donahue, N. M., Ehrhart, S., Flagan, R. C., Franchin, A., Guida, R., Hakala, J., Hansel, A., Heinritzi, M., Jokinen, T., Kangasluoma, J., Kirkby, J., Kulmala, M., Kupc, A., Lawler, M. J., Lehtipalo, K., Makhmutov, V., Mann, G., Mathot, S., Merikanto, J., Miettinen, P., Nenes, A., Onnela, A., Rap, A., Reddington, C. L. S., Riccobono, F., Richards, N. A. D., Rissanen, M. P., Rondo, L., Sarnela, N., Schobesberger, S., Sengupta, K., Simon, M., Sipilä, M., Smith, J. N., Stozhkov, Y., Tomé, A., Tröstl, J., Wagner, P. E., Wimmer, D., Winkler, P. M., Worsnop, D. R., and Carslaw, K. S.: Global atmospheric particle formation from CERN CLOUD measurements, *Science*, 354, 1119–1124, <https://doi.org/10.1126/science.aaf2649>, 2016.
- Ehrhart, S., Dunne, E. M., Manninen, H. E., Nieminen, T., Lelieveld, J., and Pozzer, A.: Two new submodels for the Modular Earth Submodel System (MESSy): New Aerosol Nucleation (NAN) and small ions (IONS) version 1.0, *Geosci. Model Dev.*, 11, 4987–5001, <https://doi.org/10.5194/gmd-11-4987-2018>, 2018.
- Fountoukis, C., Riipinen, I., Denier van der Gon, H. A. C., Charalampidis, P. E., Pilinis, C., Wiedensohler, A., O’Dowd, C., Putaud, J. P., Moerman, M., and Pandis, S. N.: Simulating ultrafine particle formation in Europe using a regional CTM: contribution of primary emissions versus secondary formation to aerosol number concentrations, *Atmos. Chem. Phys.*, 12, 8663–8677, <https://doi.org/10.5194/acp-12-8663-2012>, 2012.

- Franco, M. A., Ditas, F., Krempner, L. A., Machado, L. A. T., Andreae, M. O., Araújo, A., Barbosa, H. M. J., de Brito, J. F., Carbone, S., Holanda, B. A., Morais, F. G., Nascimento, J. P., Pöhlker, M. L., Rizzo, L. V., Sá, M., Saturno, J., Walter, D., Wolff, S., Pöschl, U., Artaxo, P., and Pöhlker, C.: Occurrence and growth of sub-50 nm aerosol particles in the Amazonian boundary layer, *Atmos. Chem. Phys.*, 22, 3469–3492, <https://doi.org/10.5194/acp-22-3469-2022>, 2022.
- Frohn, L. M., Ketzel, M., Christensen, J. H., Brandt, J., Im, U., Massling, A., Andersen, C., Plejdrup, M. S., Nielsen, O.-K., van der Gon, H. D., Manders-Groot, A., and Raaschou-Nielsen, O.: Modelling ultrafine particle number concentrations at address resolution in Denmark from 1979–2018 – Part 1: Regional and urban scale modelling and evaluation, *Atmos. Environ.*, 264, 118631, <https://doi.org/10.1016/j.atmosenv.2021.118631>, 2021.
- Gani, S., Bhandari, S., Patel, K., Seraj, S., Soni, P., Arub, Z., Habib, G., Hildebrandt Ruiz, L., and Apte, J. S.: Particle number concentrations and size distribution in a polluted megacity: the Delhi Aerosol Supersite study, *Atmos. Chem. Phys.*, 20, 8533–8549, <https://doi.org/10.5194/acp-20-8533-2020>, 2020.
- Gordon, H., Kirkby, J., Baltensperger, U., Bianchi, F., Breitenlechner, M., Curtius, J., Dias, A., Dommen, J., Donahue, N. M., Dunne, E. M., Duplissy, J., Ehrhart, S., Flagan, R. C., Frege, C., Fuchs, C., Hansel, A., Hoyle, C. R., Kulmala, M., Kürten, A., Lehtipalo, K., Makhmutov, V., Molteni, U., Rissanen, M. P., Stozkhov, Y., Tröstl, J., Tsagkogeorgas, G., Wagner, R., Williamson, C., Wimmer, D., Winkler, P. M., Yan, C., and Carslaw, K. S.: Causes and importance of new particle formation in the present-day and preindustrial atmospheres, *J. Geophys. Res.-Atmos.*, 122, 8739–8760, <https://doi.org/10.1002/2017JD026844>, 2017.
- Granier, C., Darras, S., Denier van der Gon, H., Doubalova, J., Elguindi, N., Galle, B., Gauss, M., Guevara, M., Jalkanen, J.-P., Kuenen, J., Liousse, C., Quack, B., Simpson, D., and Sindelarova, K.: The Copernicus Atmosphere Monitoring Service global and regional emissions (April 2019 version), Copernicus Atmosphere Monitoring Service (CAMS) report, <https://doi.org/10.24380/d0bn-kx16>, 2019.
- Guelle, W., Schulz, M., Balkanski, Y., and Dentener, F.: Influence of the source formulation on modeling the atmospheric global distribution of sea salt aerosol, *J. Geophys. Res.-Atmos.*, 106, 27509–27524, <https://doi.org/10.1029/2001JD900249>, 2001.
- Harrison, R. M., Beddows, D. C. S., Alam, M. S., Singh, A., Brea, J., Xu, R., Kotthaus, S., and Grimmond, S.: Interpretation of particle number size distributions measured across an urban area during the FASTER campaign, *Atmos. Chem. Phys.*, 19, 39–55, <https://doi.org/10.5194/acp-19-39-2019>, 2019.
- Heintzenberg, J., Birmili, W., Otto, R., Andreae, M. O., Mayer, J.-C., Chi, X., and Panov, A.: Aerosol particle number size distributions and particulate light absorption at the ZOTTO tall tower (Siberia), 2006–2009, *Atmos. Chem. Phys.*, 11, 8703–8719, <https://doi.org/10.5194/acp-11-8703-2011>, 2011.
- Hoesly, R. M., Smith, S. J., Feng, L., Klimont, Z., Janssens-Maenhout, G., Pitkanen, T., Seibert, J. J., Vu, L., Andres, R. J., Bolt, R. M., Bond, T. C., Dawidowski, L., Kholod, N., Kurokawa, J.-I., Li, M., Liu, L., Lu, Z., Moura, M. C. P., O'Rourke, P. R., and Zhang, Q.: Historical (1750–2014) anthropogenic emissions of reactive gases and aerosols from the Community Emissions Data System (CEDS), *Geosci. Model Dev.*, 11, 369–408, <https://doi.org/10.5194/gmd-11-369-2018>, 2018.
- Hong, G. and Jee, Y.-K.: Special issue on ultrafine particles: where are they from and how do they affect us?, *Exp. Mol. Med.*, 52, 309–310, <https://doi.org/10.1038/s12276-020-0395-z>, 2020.
- Janhäll, S., Andreae, M. O., and Pöschl, U.: Biomass burning aerosol emissions from vegetation fires: particle number and mass emission factors and size distributions, *Atmos. Chem. Phys.*, 10, 1427–1439, <https://doi.org/10.5194/acp-10-1427-2010>, 2010.
- Jeuken, A. B. M., Siegmund, P. C., Heijboer, L. C., Feichter, J., and Bengtsson, L.: On the potential of assimilating meteorological analyses in a global climate model for the purpose of model validation, *J. Geophys. Res.-Atmos.*, 101, 16939–16950, <https://doi.org/10.1029/96JD01218>, 1996.
- Jöckel, P., Tost, H., Pozzer, A., Brühl, C., Buchholz, J., Ganzeveld, L., Hoor, P., Kerkweg, A., Lawrence, M. G., Sander, R., Steil, B., Stiller, G., Tanarhte, M., Taraborrelli, D., van Aardenne, J., and Lelieveld, J.: The atmospheric chemistry general circulation model ECHAM5/MESSy1: consistent simulation of ozone from the surface to the mesosphere, *Atmos. Chem. Phys.*, 6, 5067–5104, <https://doi.org/10.5194/acp-6-5067-2006>, 2006.
- Jöckel, P., Kerkweg, A., Pozzer, A., Sander, R., Tost, H., Riede, H., Baumgaertner, A., Gromov, S., and Kern, B.: Development cycle 2 of the Modular Earth Submodel System (MESSy2), *Geosci. Model Dev.*, 3, 717–752, <https://doi.org/10.5194/gmd-3-717-2010>, 2010.
- Karner, A. A., Eisinger, D. S., and Niemeier, D. A.: Near-Roadway Air Quality: Synthesizing the Findings from Real-World Data, *Environ. Sci. Technol.*, 44, 5334–5344, <https://doi.org/10.1021/es100008x>, 2010.
- Kasper, A., Aufdenblatten, S., Forss, A., Mohr, M., and Bartscher, H.: Particulate Emissions from a Low-Speed Marine Diesel Engine, *Aerosol Sci. Tech.*, 41, 24–32, <https://doi.org/10.1080/02786820601055392>, 2007.
- Kerkweg, A., Sander, R., Tost, H., and Jöckel, P.: Technical note: Implementation of prescribed (OFFLEM), calculated (ONLEM), and pseudo-emissions (TNUDGE) of chemical species in the Modular Earth Submodel System (MESSy), *Atmos. Chem. Phys.*, 6, 3603–3609, <https://doi.org/10.5194/acp-6-3603-2006>, 2006.
- Ketzel, M., Frohn, L. M., Christensen, J. H., Brandt, J., Massling, A., Andersen, C., Im, U., Jensen, S. S., Khan, J., Nielsen, O.-K., Plejdrup, M. S., Manders, A., van der Gon, H. D., Kumar, P., and Raaschou-Nielsen, O.: Modelling ultrafine particle number concentrations at address resolution in Denmark from 1979 to 2018 – Part 2: Local and street scale modelling and evaluation, *Atmos. Environ.*, 264, 118633, <https://doi.org/10.1016/j.atmosenv.2021.118633>, 2021.
- Kohl, M. and Pozzer, A.: Simulated global ultrafine particle (UFP) concentrations, Edmond, V1 [data set], <https://doi.org/10.17617/3.7945XI>, 2023.
- Kirkby, J., Curtius, J., Almeida, J., Dunne, E., Duplissy, J., Ehrhart, S., Franchin, A., Gagné, S., Ickes, L., Kürten, A., Kupc, A., Metzger, A., Riccobono, F., Rondo, L., Schobesberger, G. T., Tsagkogeorgas, G., Wimmer, D., Amorim, A., Bianchi, F., Breitenlechner, M., David, A., Dommen, J., Downard, A., Ehn, M., Flagan, R. C., Haider, S., Hansel, A., Hauser, D., Jud, W., Junninen, H., Kreissl, F., Kvashin, A., Laaksonen, A., Lehtipalo, K.,

- Lima, J., Lovejoy, E. R., Makhmutov, V., Mathot, S., Mikkilä, J., Minginette, P., Mogo, S., Nieminen, T., Onnela, A., Pereira, P., Petaja, T., Schnitzhofer, R., Seinfeld, J., Sipila, M., Stozhkov, Y., Stratmann, F., Tome, A., Vanhanen, J., Viisanen, Y., Virtala, A., Wagner, P., Walther, H., Weingartner, E., Wex, H., Winkler, P., Carslaw, K., Worsnop, D., Baltensperger, U., and Kulmala, M.: Role of sulphuric acid, ammonia and galactic cosmic rays in atmospheric aerosol nucleation, *Nature*, 476, 429–433, <https://doi.org/10.1038/nature10343>, 2011.
- Kirkby, J., Duplissy, J., Sengupta, K., Frege, C., Gordon, H., Williamson, C., Heinritzi, M., Simon, M., Yan, C., Almeida, J., Tröstl, J., Nieminen, T., Ortega, I. K., Wagner, R., Adamov, A., Amorim, A., Bernhammer, A.-K., Bianchi, F., Breitenlechner, M., Brilke, S., Chen, X., Craven, J., Dias, A., Ehrhart, S., Flagan, R. C., Franchin, A., Fuchs, C., Guida, R., Hakala, J., Hoyle, C. R., Jokinen, T., Junninen, H., Kangasluoma, J., Kim, J., Krapf, M., Kürten, A., Laaksonen, A., Lehtipalo, K., Makhmutov, V., Mathot, S., Molteni, U., Onnela, A., Peräkylä, O., Piel, F., Petäjä, T., Praplan, A. P., Pringle, K., Rap, A., Richards, N. A. D., Riipinen, I., Rissanen, M. P., Rondo, L., Sarnela, N., Schobesberger, S., Scott, C. E., Seinfeld, J. H., Sipilä, M., Steiner, G., Stozhkov, Y., Stratmann, F., Tomé, A., Virtanen, A., Vogel, A. L., Wagner, A. C., Wagner, P. E., Weingartner, E., Wimmer, D., Winkler, P. M., Ye, P., Zhang, X., Hansel, A., Dommen, J., Donahue, N. M., Worsnop, D. R., Baltensperger, U., Kulmala, M., Carslaw, K. S., and Curtius, J.: Ion-induced nucleation of pure biogenic particles, *Nature*, 533, 521–526, <https://doi.org/10.1038/nature17953>, 2016.
- Klingmüller, K., Metzger, S., Abdelkader, M., Karydis, V. A., Stenichkov, G. L., Pozzer, A., and Lelieveld, J.: Revised mineral dust emissions in the atmospheric chemistry–climate model EMAC (MESSy 2.52 DU_Astithal KKDU2017 patch), *Geosci. Model Dev.*, 11, 989–1008, <https://doi.org/10.5194/gmd-11-989-2018>, 2018.
- Kukkonen, J., Karl, M., Keuken, M. P., Denier van der Gon, H. A. C., Denby, B. R., Singh, V., Douras, J., Manders, A., Samaras, Z., Moussiopoulos, N., Jonkers, S., Aarnio, M., Karpinen, A., Kangas, L., Lützenkirchen, S., Petäjä, T., Vouitsis, I., and Sokhi, R. S.: Modelling the dispersion of particle numbers in five European cities, *Geosci. Model Dev.*, 9, 451–478, <https://doi.org/10.5194/gmd-9-451-2016>, 2016.
- Kulmala, M., Vehkamäki, H., Petäjä, T., Dal Maso, M., Lauri, A., Kerminen, V.-M., Birmili, W., and McMurry, P.: Formation and growth rates of ultrafine atmospheric particles: a review of observations, *J. Aerosol Sci.*, 35, 143–176, <https://doi.org/10.1016/j.jaerosci.2003.10.003>, 2004.
- Kumar, P., Morawska, L., Birmili, W., Paasonen, P., Hu, M., Kulmala, M., Harrison, R. M., Norford, L., and Britter, R.: Ultrafine particles in cities, *Environ. Int.*, 66, 1–10, <https://doi.org/10.1016/j.envint.2014.01.013>, 2014.
- Kwon, H.-S., Ryu, M. H., and Carlsten, C.: Ultrafine particles: unique physicochemical properties relevant to health and disease, *Exp. Mol. Med.*, 52, 318–328, <https://doi.org/10.1038/s12276-020-0405-1>, 2020.
- Lelieveld, J., Evans, J. S., Fnais, M., Giannadaki, D., and Pozzer, A.: The contribution of outdoor air pollution sources to premature mortality on a global scale, *Nature*, 525, 367–371, <https://doi.org/10.1038/nature15371>, 2015.
- Lelieveld, J., Klingmüller, K., Pozzer, A., Burnett, R. T., Haines, A., and Ramanathan, V.: Effects of fossil fuel and total anthropogenic emission removal on public health and climate, *P. Natl. Acad. Sci. USA*, 116, 7192–7197, <https://doi.org/10.1073/pnas.1819989116>, 2019.
- Lelieveld, J., Pozzer, A., Pöschl, U., Fnais, M., Haines, A., and Münzel, T.: Loss of life expectancy from air pollution compared to other risk factors: a worldwide perspective, *Cardiovasc. Res.*, 116, 1910–1917, <https://doi.org/10.1093/cvr/cvaa025>, 2020.
- Li, G., Su, H., Ma, N., Tao, J., Kuang, Y., Wang, Q., Hong, J., Zhang, Y., Kuhn, U., Zhang, S., Pan, X., Lu, N., Tang, M., Zheng, G., Wang, Z., Gao, Y., Cheng, P., Xu, W., Zhou, G., Zhao, C., Yuan, B., Shao, M., Ding, A., Zhang, Q., Fu, P., Sun, Y., Pöschl, U., and Cheng, Y.: Multiphase chemistry experiment in Fogs and Aerosols in the North China Plain (McFAN): integrated analysis and intensive winter campaign 2018, *Faraday Discuss.*, 226, 207–222, <https://doi.org/10.1039/D0FD00099J>, 2021.
- Liu, M. and Matsui, H.: Secondary Organic Aerosol Formation Regulates Cloud Condensation Nuclei in the Global Remote Troposphere, *Geophys. Res. Lett.*, 49, e2022GL100543, <https://doi.org/10.1029/2022GL100543>, 2022.
- Liu, Y., Yan, C., Feng, Z., Zheng, F., Fan, X., Zhang, Y., Li, C., Zhou, Y., Lin, Z., Guo, Y., Zhang, Y., Ma, L., Zhou, W., Liu, Z., Dada, L., Dällenbach, K., Kontkanen, J., Cai, R., Chan, T., Chu, B., Du, W., Yao, L., Wang, Y., Cai, J., Kangasluoma, J., Kokkonen, T., Kujansuu, J., Rusanen, A., Deng, C., Fu, Y., Yin, R., Li, X., Lu, Y., Liu, Y., Lian, C., Yang, D., Wang, W., Ge, M., Wang, Y., Worsnop, D. R., Junninen, H., He, H., Kerminen, V.-M., Zheng, J., Wang, L., Jiang, J., Petäjä, T., Bianchi, F., and Kulmala, M.: Continuous and comprehensive atmospheric observations in Beijing: a station to understand the complex urban atmospheric environment, *Big Earth Data*, 4, 295–321, <https://doi.org/10.1080/20964471.2020.1798707>, 2020.
- Lohmann, U. and Feichter, J.: Global indirect aerosol effects: a review, *Atmos. Chem. Phys.*, 5, 715–737, <https://doi.org/10.5194/acp-5-715-2005>, 2005.
- Lu, X., Zhang, S., Xing, J., Wang, Y., Chen, W., Ding, D., Wu, Y., Wang, S., Duan, L., and Hao, J.: Progress of Air Pollution Control in China and Its Challenges and Opportunities in the Ecological Civilization Era, *Engineering*, 6, 1423–1431, <https://doi.org/10.1016/j.eng.2020.03.014>, 2020.
- McDuffie, E. E., Smith, S. J., O'Rourke, P., Tibrewal, K., Venkataraman, C., Marais, E. A., Zheng, B., Crippa, M., Brauer, M., and Martin, R. V.: A global anthropogenic emission inventory of atmospheric pollutants from sector- and fuel-specific sources (1970–2017): an application of the Community Emissions Data System (CEDS), *Earth Syst. Sci. Data*, 12, 3413–3442, <https://doi.org/10.5194/essd-12-3413-2020>, 2020a.
- McDuffie, E., Smith, S., O'Rourke, P., Tibrewal, K., Venkataraman, C., Marais, E., Zheng, B., Crippa, M., Brauer, M., and Martin, R.: CEDS_GBD-MAPS: Global Anthropogenic Emission Inventory of NO_x, SO₂, CO, NH₃, NMVOCs, BC, and OC from 1970–2017 (2020_v1.0), Zenodo [data set], <https://doi.org/10.5281/zenodo.3754964>, 2020b.
- Monahan, E. C.: The Ocean as a Source for Atmospheric Particles, Springer Netherlands, Dordrecht, 129–163, https://doi.org/10.1007/978-94-009-4738-2_6, 1986.
- Murray, C. J., Aravkin, A. Y., Zheng, P., et al.: Global burden of 87 risk factors in 204 countries and territories, 1990–2019:

- a systematic analysis for the Global Burden of Disease Study 2019, *Lancet*, 396, 1223–1249, [https://doi.org/10.1016/S0140-6736\(20\)30752-2](https://doi.org/10.1016/S0140-6736(20)30752-2), 2020.
- Paasonen, P.: Global particle number emissions, International Institute for Applied System Analysis [data set], <https://previous.iiasa.ac.at/web/home/research/researchPrograms/air/PN.html> (last access: 17 February 2023), 2016.
- Paasonen, P., Kupiainen, K., Klimont, Z., Visschedijk, A., Denier van der Gon, H. A. C., and Amann, M.: Continental anthropogenic primary particle number emissions, *Atmos. Chem. Phys.*, 16, 6823–6840, <https://doi.org/10.5194/acp-16-6823-2016>, 2016.
- Pan, X., Ichoku, C., Chin, M., Bian, H., Darmenov, A., Colarco, P., Ellison, L., Kucsera, T., da Silva, A., Wang, J., Oda, T., and Cui, G.: Six global biomass burning emission datasets: inter-comparison and application in one global aerosol model, *Atmos. Chem. Phys.*, 20, 969–994, <https://doi.org/10.5194/acp-20-969-2020>, 2020.
- Petzold, A. and Schröder, F. P.: Jet Engine Exhaust Aerosol Characterization, *Aerosol Sci. Tech.*, 28, 62–76, <https://doi.org/10.1080/02786829808965512>, 1998.
- Petzold, A., Stein, C., Nyeki, S., Gysel, M., Weingartner, E., Baltensperger, U., Giebl, H., Hitznerberger, R., Döpelheuer, A., Vrchotický, S., Puxbaum, H., Johnson, M., Hurley, C. D., Marsh, R., and Wilson, C. W.: Properties of jet engine combustion particles during the PartEmiss experiment: Microphysics and Chemistry, *Geophys. Res. Lett.*, 30, 1719, <https://doi.org/10.1029/2003GL017283>, 2003.
- Petzold, A., Hasselbach, J., Lauer, P., Baumann, R., Franke, K., Gurk, C., Schlager, H., and Weingartner, E.: Experimental studies on particle emissions from cruising ship, their characteristic properties, transformation and atmospheric lifetime in the marine boundary layer, *Atmos. Chem. Phys.*, 8, 2387–2403, <https://doi.org/10.5194/acp-8-2387-2008>, 2008.
- Pope, C. A. and Dockery, D. W.: Health Effects of Fine Particulate Air Pollution: Lines that Connect, *J. Air Waste Manage. Assoc.*, 56, 709–742, <https://doi.org/10.1080/10473289.2006.10464485>, 2006.
- Pöschl, U., von Kuhlmann, R., Poisson, N., and Crutzen, P. J.: Development and intercomparison of condensed isoprene oxidation mechanisms for global atmospheric modeling, *J. Atmos. Chem.*, 37, 29–52, <https://doi.org/10.1023/A:1006391009798>, 2000.
- Pozzer, A., Jöckel, P., and Van Aardenne, J.: The influence of the vertical distribution of emissions on tropospheric chemistry, *Atmos. Chem. Phys.*, 9, 9417–9432, <https://doi.org/10.5194/acp-9-9417-2009>, 2009.
- Pozzer, A., de Meij, A., Pringle, K. J., Tost, H., Doering, U. M., van Aardenne, J., and Lelieveld, J.: Distributions and regional budgets of aerosols and their precursors simulated with the EMAC chemistry-climate model, *Atmos. Chem. Phys.*, 12, 961–987, <https://doi.org/10.5194/acp-12-961-2012>, 2012.
- Pozzer, A., de Meij, A., Yoon, J., Tost, H., Georgoulas, A. K., and Astitha, M.: AOD trends during 2001–2010 from observations and model simulations, *Atmos. Chem. Phys.*, 15, 5521–5535, <https://doi.org/10.5194/acp-15-5521-2015>, 2015.
- Pozzer, A., Reifenberg, S. F., Kumar, V., Franco, B., Kohl, M., Taraborrelli, D., Gromov, S., Ehrhart, S., Jöckel, P., Sander, R., Fall, V., Rosanka, S., Karydis, V., Akritidis, D., Emmerichs, T., Crippa, M., Guizzardi, D., Kaiser, J. W., Clarisse, L., Kiendler-Scharr, A., Tost, H., and Tsimpidi, A.: Simulation of organics in the atmosphere: evaluation of EMACv2.54 with the Mainz Organic Mechanism (MOM) coupled to the OR-ACLE (v1.0) submodel, *Geosci. Model Dev.*, 15, 2673–2710, <https://doi.org/10.5194/gmd-15-2673-2022>, 2022.
- Pringle, K. J., Tost, H., Message, S., Steil, B., Giannadaki, D., Nenes, A., Fountoukis, C., Stier, P., Vignati, E., and Lelieveld, J.: Description and evaluation of GMX: a new aerosol submodel for global simulations (v1), *Geosci. Model Dev.*, 3, 391–412, <https://doi.org/10.5194/gmd-3-391-2010>, 2010.
- Reid, J. S., Koppmann, R., Eck, T. F., and Eleuterio, D. P.: A review of biomass burning emissions part II: intensive physical properties of biomass burning particles, *Atmos. Chem. Phys.*, 5, 799–825, <https://doi.org/10.5194/acp-5-799-2005>, 2005.
- Riccobono, F., Schobesberger, S., Scott, C. E., Dommen, J., Ortega, I. K., Rondo, L., Almeida, J., Amorim, A., Bianchi, F., Breitenlechner, M., David, A., Downard, A., Dunne, E. M., Duplissy, J., Ehrhart, S., Flanagan, R. C., Franchin, A., Hansel, A., Junninen, H., Kajos, M., Keskinen, H., Kupc, A., Kürten, A., Kvashin, A. N., Laaksonen, A., Lehtipalo, K., Makhmutov, V., Mathot, S., Nieminen, T., Onnela, A., Petäjä, T., Praplan, A. P., Santos, F. D., Schallhart, S., Seinfeld, J. H., Sipilä, M., Spracklen, D. V., Stozhkov, Y., Stratmann, F., Tomé, A., Tsagkogeorgas, G., Vaattovaara, P., Viisanen, Y., Vrtala, A., Wagner, P. E., Weingartner, E., Wex, H., Wimmer, D., Carslaw, K. S., Curtius, J., Donahue, N. M., Kirkby, J., Kulmala, M., Worsnop, D. R., and Baltensperger, U.: Oxidation Products of Biogenic Emissions Contribute to Nucleation of Atmospheric Particles, *Science*, 344, 717–721, <https://doi.org/10.1126/science.1243527>, 2014.
- Roeckner, E., Bäuml, G., Bonaventura, L., Brokopf, R., Esch, M., Giorgetta, M., Hagemann, S., Kirchner, I., Kornblueh, L., Manzini, E., Hagemann, S., Kirchner, I., Kornblueh, L., Manzini, E., Schlese, U., and Schulzweida, U.: The atmospheric general circulation model ECHAM 5. PART I: Model description, Tech. rep., Max-Planck-Institut für Meteorologie, <https://doi.org/10.17617/2.995269>, 2003.
- Rose, C., Collaud Coen, M., Andrews, E., Lin, Y., Bossert, I., Lund Myhre, C., Tuch, T., Wiedensohler, A., Fiebig, M., Aalto, P., Alastuey, A., Alonso-Blanco, E., Andrade, M., Artíñano, B., Arsov, T., Baltensperger, U., Bastian, S., Bath, O., Beukes, J. P., Brem, B. T., Bukowiecki, N., Casquero-Vera, J. A., Conil, S., Eleftheriadis, K., Favez, O., Flentje, H., Gini, M. I., Gómez-Moreno, F. J., Gysel-Beer, M., Hallar, A. G., Kalapov, I., Kalivitis, N., Kasper-Giebl, A., Keywood, M., Kim, J. E., Kim, S.-W., Kristensson, A., Kulmala, M., Lihavainen, H., Lin, N.-H., Lyamani, H., Marinoni, A., Martins Dos Santos, S., Mayol-Bracero, O. L., Meinhardt, F., Merkel, M., Metzger, J.-M., Mihalopoulos, N., Ondracek, J., Pandolfi, M., Pérez, N., Petäjä, T., Petit, J.-E., Picard, D., Pichon, J.-M., Pont, V., Putaud, J.-P., Reisen, F., Sellegri, K., Sharma, S., Schauer, G., Sheridan, P., Sherman, J. P., Schwerin, A., Sohmer, R., Sorribas, M., Sun, J., Tulet, P., Vakkari, V., van Zyl, P. G., Velarde, F., Villani, P., Vratolis, S., Wagner, Z., Wang, S.-H., Weinhold, K., Weller, R., Yela, M., Zdimal, V., and Laj, P.: Seasonality of the particle number concentration and size distribution: a global analysis retrieved from the network of Global Atmosphere Watch (GAW) near-surface observatories, *Atmos. Chem. Phys.*, 21, 17185–17223, <https://doi.org/10.5194/acp-21-17185-2021>, 2021.

- Saha, P. K., Hankey, S., Marshall, J. D., Robinson, A. L., and Presto, A. A.: High-Spatial-Resolution Estimates of Ultrafine Particle Concentrations across the Continental United States, *Environ. Sci. Technol.*, 55, 10320–10331, <https://doi.org/10.1021/acs.est.1c03237>, 2021.
- Salma, I., Borsós, T., Németh, Z., Weidinger, T., Aalto, P., and Kulmala, M.: Comparative study of ultrafine atmospheric aerosol within a city, *Atmos. Environ.*, 92, 154–161, <https://doi.org/10.1016/j.atmosenv.2014.04.020>, 2014.
- Sander, R., Baumgaertner, A., Cabrera-Perez, D., Frank, F., Grovov, S., Grooß, J.-U., Harder, H., Huijnen, V., Jöckel, P., Karydis, V. A., Niemeyer, K. E., Pozzer, A., Riede, H., Schultz, M. G., Taraborrelli, D., and Tauer, S.: The community atmospheric chemistry box model CAABA/MECCA-4.0, *Geosci. Model Dev.*, 12, 1365–1385, <https://doi.org/10.5194/gmd-12-1365-2019>, 2019.
- Schraufnagel, D. E.: The health effects of ultrafine particles, *Exp. Mol. Med.*, 52, 311–317, <https://doi.org/10.1038/s12276-020-0403-3>, 2020.
- Schraufnagel, D. E., Balmes, J. R., Cowl, C. T., De Matteis, S., Jung, S.-H., Mortimer, K., Perez-Padilla, R., Rice, M. B., Riojas-Rodriguez, H., Sood, A., Thurston, G. D., To, T., Vanker, A., and Wuebbles, D. J.: Air Pollution and Noncommunicable Diseases: A Review by the Forum of International Respiratory Societies' Environmental Committee, Part 2: Air Pollution and Organ Systems, *Chest*, 155, 417–426, <https://doi.org/10.1016/j.chest.2018.10.041>, 2019.
- Schutgens, N. A. J., Gryspeerdt, E., Weigum, N., Tsyro, S., Goto, D., Schulz, M., and Stier, P.: Will a perfect model agree with perfect observations? The impact of spatial sampling, *Atmos. Chem. Phys.*, 16, 6335–6353, <https://doi.org/10.5194/acp-16-6335-2016>, 2016a.
- Schutgens, N. A. J., Partridge, D. G., and Stier, P.: The importance of temporal collocation for the evaluation of aerosol models with observations, *Atmos. Chem. Phys.*, 16, 1065–1079, <https://doi.org/10.5194/acp-16-1065-2016>, 2016b.
- Schutgens, N., Tsyro, S., Gryspeerdt, E., Goto, D., Weigum, N., Schulz, M., and Stier, P.: On the spatio-temporal representativeness of observations, *Atmos. Chem. Phys.*, 17, 9761–9780, <https://doi.org/10.5194/acp-17-9761-2017>, 2017.
- Sebastian, M., Kompalli, S. K., Kumar, V. A., Jose, S., Babu, S. S., Pandithurai, G., Singh, S., Hooda, R. K., Soni, V. K., Pierce, J. R., Vakkari, V., Asmi, E., Westervelt, D. M., Hyvärinen, A.-P., and Kanawade, V. P.: Observations of particle number size distributions and new particle formation in six Indian locations, *Atmos. Chem. Phys.*, 22, 4491–4508, <https://doi.org/10.5194/acp-22-4491-2022>, 2022.
- Seinfeld, J. H. and Pandis, S. N.: *Atmospheric chemistry and physics: from air pollution to climate change*, John Wiley & Sons, ISBN 978-1-118-94740-1, 2016.
- Shen, X., Sun, J., Ma, Q., Zhang, Y., Zhong, J., Yue, Y., Xia, C., Hu, X., Zhang, S., and Zhang, X.: Long-term trend of new particle formation events in the Yangtze River Delta, China and its influencing factors: 7-year dataset analysis, *Sci. Total Environ.*, 807, 150783, <https://doi.org/10.1016/j.scitotenv.2021.150783>, 2022.
- Stone, V., Miller, M. R., Clift, M. J., Elder, A., Mills, N. L., Möller, P., Schins, R. P., Vogel, U., Kreyling, W. G., Jensen, K. A., Kuhlbusch, T. A., Schwarze, P. E., Hoet, P., Pietroiusti, A., Vizcaya-Ruiz, A. D., Baeza-Squiban, A., Teixeira, J. P., Tran, C. L., and Cassee, F. R.: Nanomaterials Versus Ambient Ultrafine Particles: An Opportunity to Exchange Toxicology Knowledge, *Environ. Health Persp.*, 125, 106002, <https://doi.org/10.1289/EHP424>, 2017.
- Thamban, N. M., Lalchandani, V., Kumar, V., Mishra, S., Bhattu, D., Slowik, J. G., Prevot, A. S., Satish, R., Rastogi, N., and Tripathi, S. N.: Evolution of size and composition of fine particulate matter in the Delhi megacity during later winter, *Atmos. Environ.*, 267, 118752, <https://doi.org/10.1016/j.atmosenv.2021.118752>, 2021.
- Tost, H., Jöckel, P., Kerkweg, A., Sander, R., and Lelieveld, J.: Technical note: A new comprehensive SCAVenging submodel for global atmospheric chemistry modelling, *Atmos. Chem. Phys.*, 6, 565–574, <https://doi.org/10.5194/acp-6-565-2006>, 2006.
- Trecher, P., Garcia-Marles, M., Liu, X., Reche, C., Pérez, N., Savadkoobi, M., Beddows, D., Salma, I., Vörösmarty, M., Casans, A., Casquero-Vera, J. A., Hueglin, C., Marchand, N., Chazeau, B., Gille, G., Kalkavouras, P., Mihalopoulos, N., Ondracek, J., Zikova, N., Niemi, J. V., Manninen, H. E., Green, D. C., Tremper, A. H., Norman, M., Vratolis, S., Eleftheriadis, K., Gómez-Moreno, F. J., Alonso-Blanco, E., Gerwig, H., Wiedensohler, A., Weinhold, K., Merkel, M., Bastian, S., Petit, J.-E., Favez, O., Crumeyrolle, S., Ferlay, N., Martins Dos Santos, S., Putaud, J.-P., Timonen, H., Lampilahti, J., Asbach, C., Wolf, C., Kaminski, H., Altug, H., Hoffmann, B., Rich, D. Q., Pandolfi, M., Harrison, R. M., Hopke, P. K., Petäjä, T., Alastuey, A., and Querol, X.: Phenomenology of ultrafine particle concentrations and size distribution across urban Europe, *Environ. Int.*, 172, 107744, <https://doi.org/10.1016/j.envint.2023.107744>, 2023.
- Tsimpidi, A. P., Karydis, V. A., Pozzer, A., Pandis, S. N., and Lelieveld, J.: ORACLE 2-D (v2.0): an efficient module to compute the volatility and oxygen content of organic aerosol with a global chemistry–climate model, *Geosci. Model Dev.*, 11, 3369–3389, <https://doi.org/10.5194/gmd-11-3369-2018>, 2018.
- Twomey, S.: The nuclei of natural cloud formation part II: The supersaturation in natural clouds and the variation of cloud droplet concentration, *Geofisica Pura e Applicata*, 43, 243–249, <https://doi.org/10.1007/BF01993560>, 1959.
- Villa, T., Jayaratne, E., Gonzalez, L., and Morawska, L.: Determination of the vertical profile of particle number concentration adjacent to a motorway using an unmanned aerial vehicle, *Environ. Pollut.*, 230, 134–142, <https://doi.org/10.1016/j.envpol.2017.06.033>, 2017.
- Wang, J., Krejci, R., Giangrande, S., Kuang, C., Barbosa, H. M. J., Brito, J., Carbone, S., Chi, X., Comstock, J., Ditas, F., Lavric, J., Manninen, H. E., Mei, F., Moran-Zuloaga, D., Pöhlker, C., Pöhlker, M. L., Saturno, J., Schmid, B., Souza, R. A. F., Springston, S. R., Tomlinson, J. M., Toto, T., Walter, D., Wimmer, D., Smith, J. N., Kulmala, M., Machado, L. A. T., Artaxo, P., Andreae, M. O., Petäjä, T., and Martin, S. T.: Amazon boundary layer aerosol concentration sustained by vertical transport during rainfall, *Nature*, 539, 416–419, <https://doi.org/10.1038/nature19819>, 2016.
- Weigel, R., Mähne, C., Baumgartner, M., Dragoneas, A., Vogel, B., Ploeger, F., Viciani, S., D'Amato, F., Bucci, S., Legras, B., Luo, B., and Borrmann, S.: In situ observation of new particle formation (NPF) in the tropical tropopause layer of the 2017 Asian monsoon anticyclone – Part 1: Summary of

- StratoClim results, *Atmos. Chem. Phys.*, 21, 11689–11722, <https://doi.org/10.5194/acp-21-11689-2021>, 2021.
- WHO (World Health Organization): WHO Air quality guidelines for particulate matter, ozone, nitrogen dioxide and sulfur dioxide: global update 2005: summary of risk assessment, World Health Organization, Occupational and Environmental Health Team, <https://www.who.int/publications/i/item/WHO-SDE-PHE-OEH-06-02> (last access: 17 February 2023), 2006.
- Williamson, C. J., Kupc, A., Axisa, D., Bilsback, K. R., Bui, T., Campuzano-Jost, P., Dollner, M., Froyd, K. D., Hodshire, A. L., Jimenez, J. L., Kodros, J. K., Luo, G., Murphy, D. M., Nault, B. A., Ray, E. A., Weinzierl, B., Wilson, J. C., Yu, F., Yu, P., Pierce, J. R., and Brock, C. A.: A large source of cloud condensation nuclei from new particle formation in the tropics, *Nature*, 574, 399–403, <https://doi.org/10.1038/s41586-019-1638-9>, 2019.
- Wu, T. and Boor, B. E.: Urban aerosol size distributions: a global perspective, *Atmos. Chem. Phys.*, 21, 8883–8914, <https://doi.org/10.5194/acp-21-8883-2021>, 2021.
- Wu, Z., Hu, M., Lin, P., Liu, S., Wehner, B., and Wiedensohler, A.: Particle number size distribution in the urban atmosphere of Beijing, China, *Atmos. Environ.*, 42, 7967–7980, <https://doi.org/10.1016/j.atmosenv.2008.06.022>, 2008.
- Zeng, Y., Cao, Y., Qiao, X., Seyler, B. C., and Tang, Y.: Air pollution reduction in China: Recent success but great challenge for the future, *Sci. Total Environ.*, 663, 329–337, <https://doi.org/10.1016/j.scitotenv.2019.01.262>, 2019.
- Zhao, B., Shrivastava, M., Donahue, N. M., Gordon, H., Schervish, M., Shilling, J. E., Zaveri, R. A., Wang, J., Andreae, M. O., Zhao, C., Gaudet, B., Liu, Y., Fan, J., and Fast, J. D.: High concentration of ultrafine particles in the Amazon free troposphere produced by organic new particle formation, *P. Natl. Acad. Sci. USA*, 117, 25344–25351, <https://doi.org/10.1073/pnas.2006716117>, 2020.

4 CHAPTER

Simulating volcanic emissions – Submodel for Explosive Volcanic ERuptions (EVER)

This chapter presents a revised manuscript accepted for publication in *Geoscientific Model Development* on April 14th, 2025. I am the first author of and the main contributor to this study. I developed the new submodel with the help of Andrea Pozzer and Patrick Jöckel, and established the default namelist setup with the help of Christoph Brühl. I evaluated the simulations, analysed the results, and wrote the manuscript. More detailed information on the Author contributions are provided at the end of the manuscript.

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New submodel for emissions from Explosive Volcanic ERUptions (EVER v1.1) within the Modular Earth Submodel System (MESSy, version 2.55.1)

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Abstract. This work documents the operation of a new submodel for tracer emissions from Explosive Volcanic ERUptions (EVER v1.1), developed within the Modular Earth Submodel System (MESSy, version 2.55.1). EVER calculates additional tendencies of gaseous and aerosol tracers based on emission source parameters, aligned to specific sequences of volcanic eruptions or other atmospheric emission sources, and allowing various vertical emission profiles. We show that volcanic SO₂ plumes can be reasonably reproduced through EVER emissions in numerical simulations with the ECHAM/MESSy Atmospheric Chemistry Model (EMAC), using satellite observations of SO₂ columns and mixing ratios following the explosive eruption of the Nabro volcano (Eritrea) in 2011 and a degassing event of the Kilauea volcano (2018) in Hawaii. Previous volcanic studies showed large variability in stratospheric SO₂ burdens depending on the chosen volcanic emission databases and parameters. Sensitivity studies on SO₂ emissions from the Nabro volcano explore perturbations of the emission source parameters, revealing that emission altitude and the emitted mass above the tropopause are most important for the mid- to long-term evolution of stratospheric SO₂ plumes and the resulting stratospheric aerosol, while the correct timing and geographical location of the stratospheric entrance is crucial for the short-term plume evolution. We integrate information from a volcanic SO₂ emission inventory, additional satellite observations, and our findings from the sensitivity studies to establish a historical standard setup for volcanic eruptions impacting stratospheric SO₂ from 1990 to 2023. This setup was successfully evaluated with satellite observations of stratospheric SO₂ burden and aerosol optical properties. We advocate for this to be a standardized setup in all simulations within the MESSy framework concentrating on the upper troposphere and stratosphere in this period. Further potential applications of EVER involve studies on volcanic ash, wildfires, solar radiation modification, and atmospheric transport processes.

1 Introduction

20 Volcanic eruptions strongly impact atmospheric chemistry, climate dynamics, and air pollution. The most explosive eruptions reach the Upper Troposphere and Lower Stratosphere (UTLS), carrying primarily emitted particles (mostly volcanic ash) and volcanic gases, that lead to the formation and growth of aerosol particles. The resulting additional stratospheric aerosol loading may exert a negative radiative forcing (Schallock et al., 2023; Schmidt et al., 2018), and serves as surfaces for heterogeneous reactions, thus, impacting stratospheric composition in general and ozone in particular (e. g. Klobas et al., 2017; Tie and
25 Brasseur, 1995). Moreover, in the troposphere, gaseous and particulate emissions from degassing volcanoes can affect the environment and public health via inhalation or acid rain (Durand and Grattan, 2001; Stewart et al., 2022).

Although volcanically emitted gases are typically dominated by water vapour and carbon dioxide, the respective total amount is mostly negligible¹ in comparison to global emissions and concentrations (Textor et al., 2004). However, in general, the third most abundant species in volcanic plumes, sulfur dioxide (SO₂), exerts the most significant impact on atmospheric chemistry
30 and climate, substantially enriching atmospheric SO₂ levels (Textor et al., 2004). Subsequently, SO₂ undergoes oxidation to form sulfuric acid (H₂SO₄), which rapidly converts to the liquid aerosol phase under most atmospheric conditions (e. g. Kremser et al., 2016), but can also form solid salts depending on atmospheric conditions. We refer to "sulfur aerosol" as the sum of liquid and solid sulfur aerosol in the following.

The (global) impact of volcanic eruptions strongly depends on the strength of the eruption and its geographical location.
35 SO₂ emissions from degassing volcanoes and smaller eruptions, failing to reach the stratosphere, primarily influence the local environment, as emitted species and their products are usually removed in the troposphere by rainout processes within weeks². When volcanic SO₂ emissions reach the stratosphere, the subsequently formed sulfur aerosol enhances stratospheric aerosol and is distributed widely across the globe. The lifetime of stratospheric aerosol can exceed 2 years when injected in the tropics, leading to sustained impacts on atmospheric radiative balance and climate dynamics. The strongest eruption in
40 recent times, Mount Pinatubo in 1991, significantly increased the stratospheric Aerosol Optical Depth (sAOD) and resulted in a radiative forcing of -3 to -5 Wm⁻² (Schallock et al., 2023; Schmidt et al., 2018), comparable to the magnitude of positive anthropogenic radiative forcing attributed to greenhouse gas emissions (IPCC, 2021).

In addition to gaseous emissions, volcanic eruptions release varying amounts of primary aerosol, mainly consisting of volcanic ash, directly into the atmosphere. While the lifetime of ash particles in the atmosphere is relatively short, resulting in
45 mostly negligible climate effects, recent studies have indicated that ash can persist in the atmosphere for longer durations than previously anticipated (Vernier et al., 2016). Furthermore, ash particles can interact with other atmospheric constituents, such as SO₂ and H₂SO₄, thereby influencing atmospheric chemistry (Zhu et al., 2020), and pose severe hazards to aviation and

¹Notable exceptions arise during eruptions of submarine volcanoes, such as the Hunga Tonga-Hunga Ha'apai volcano in January 2022 (e. g. Vömel et al., 2022; Sellitto et al., 2022; Schoeberl et al., 2022; Xu et al., 2022), that can inject substantial amounts of water vapour into the stratosphere, thereby impacting climate, atmospheric dynamics, and radiative forcing.

²However, they can exert a strong risk for public health and even influence the short-term climate by altering cloud properties (e. g. the 2014-2015 fissure eruption in the Holuhraun vent of Bardarbunga, Iceland, leading to a global-mean radiative forcing of -0.2 Wm⁻²; Malavelle et al., 2017)

affect public health and the environment, as they deposit on the Earth’s surface (Durand and Grattan, 2001; Stewart et al., 2022).

50 The understanding of the aforementioned impacts of volcanic eruptions on climate and atmospheric chemistry heavily relies on atmospheric numerical modelling. Numerical simulations can study the impact of volcanoes on the radiative budget, atmospheric chemistry, and dynamics of the atmosphere. Furthermore, the incorporation of volcanic eruptions is indispensable for model-based studies on global atmospheric aerosol, particularly in comparison to observations from satellites and aircraft campaigns. Without accurate accounting for volcanic eruptions, models may underestimate upper tropospheric and stratospheric
55 aerosol concentrations, thereby compromising the accuracy of simulated atmospheric aerosol distributions (e. g., Reifenberg et al., 2022). Thus, atmospheric models need the capability to account for gaseous and particulate volcanic emissions in general, and SO₂ in particular.

Several volcanic SO₂ emission databases have been developed, each providing a basis for implementing best-estimate emission parameters for individual eruptions into global atmospheric chemistry models. Timmreck et al. (2018) recommend four
60 different emission inventories within the Interactive Stratospheric Aerosol Model Intercomparison Project (ISA-MIP). Neely III and Schmidt (2016) and Mills et al. (2016) provide an inventory for tropospheric and stratospheric volcanoes, covering daily emissions and providing plume top and minimal height. The emission database of Carn et al. (2017) provides a comprehensive list of individual explosive volcanic SO₂ injections, covering both tropospheric and stratospheric eruptions, however not distinguishing between the tropospheric and stratospheric part of the plume. Brühl et al. (2018) developed a volcanic SO₂
65 emission database from 1990 to 2021 (updated by Schallcock et al., 2023), focusing only on the stratospheric part of the plume, also including smaller eruptions. Finally, the emission database from Diehl et al. (2012) only covers volcanoes up to 2010.

The treatment of SO₂ from volcanic eruptions based on the available emission inventories in global atmospheric models varies widely (e. g. Quaglia et al., 2023; Timmreck et al., 2018). Quaglia et al. (2023) performed a model intercomparison study focusing on the Pinatubo eruption, finding that inter- and intra-model differences in the response of SO₂ and sulfur
70 aerosol on the Pinatubo eruption are large for a range of sensitivity experiments. The differences were mostly attributed to differing stratospheric transport, emission databases, aerosol microphysics and stratospheric chemistry. Vattioni et al. (2024) investigated the impact of microphysical settings within the SOCOL-AERv2 aerosol chemistry-climate model and found that the microphysical timestep, as well as the order of the microphysical processes can lead to vastly differing resulting stratospheric aerosol.

75 Brodowsky et al. (2021) analysed how small- and medium-sized explosive eruptions (from 2008 to 2012) are represented within interactive models, investigating the sensitivity of the resulting aerosol burdens to the different emission databases, internal model variability, dynamic nudging and the vertical resolution. The largest uncertainties resulted from the emission databases and their application. Most interactive aerosol volcanic model studies inject SO₂ in columns at the geographical location of the volcano (e. g. Quaglia et al., 2023; Mills et al., 2016; Schmidt et al., 2018; Brodowsky et al., 2021), however
80 the vertical extent of the column is mostly unknown and depends on a number of assumptions. For instance, Brodowsky et al. (2021) emitted the recommended SO₂ amounts (for the emission inventories of Brühl et al., 2018; Carn et al., 2017; Diehl et al., 2012) from the prescribed plume top height down to two thirds of that altitude. However, this approach may not align

with how the emission inventory was derived, i. e. if only the stratospheric plume is considered for the derived SO₂ mass (as in the database from Brühl et al. (2018)) or the tropospheric part of the plume as well (as in the databases from Carn et al. (2017) and Diehl et al. (2012)).

In this study, we expand the capabilities of the Modular Earth Submodel System, MESSy (version 2.55.1, Jöckel et al., 2010), to incorporate the seamless simulation of volcanic eruptions with a new submodel for Explosive Volcanic ERuptions (EVER). In previous simulations within the MESSy framework 3D mixing ratio perturbations of SO₂ were added manually to existing mixing ratios at fixed points in time (e. g. Schallock et al., 2023; Brühl et al., 2018). This required a high manual effort, and thus stratospheric volcanic eruptions were not part of standard simulations. The new submodel along with a newly developed namelist setup for stratospheric volcanic eruptions from 1990 to 2023 (based on the emission inventory from Schallock et al. (2023) and Brühl et al. (2018)), that automates this functionality, is presented in Sect. 2.

Using the new EVER submodel, we perform three numerical experiments:

1. First, we investigate the initial SO₂ injection parameters, such as emission altitude, geographical location, emitted mass and vertical extent, with sensitivity simulations varying the aforementioned parameters. The resulting short- to mid-term SO₂ plume development is compared to satellite observations of SO₂ columns and stratospheric mixing ratios for the Nabro eruption in 2011 to tackle uncertainties in the initial stratospheric SO₂ burden introduced by the different emission inventories. We provide recommendations regarding the implementation of stratospheric SO₂ injections from volcanic eruptions within the MESSy framework (applied in the historic default namelist setup for EVER), as well as in atmospheric models more broadly.
2. Second, we evaluate a historic default namelist setup for the EVER submodel for the period from 2008-2011. This setup is developed based on the emission database from Schallock et al. (2023), and we further refine it here using observations from the IASI satellite (Clarisse et al., 2012, 2014) and the findings from our sensitivity studies. Simulated stratospheric SO₂ burden and the optical properties of the resulting stratospheric aerosol are compared to satellite observations.
3. Third, we evaluate the EVER submodel with regard to the additional use case of tropospheric degassing volcanoes with SO₂ column observations of the Kilauea degassing event from June 2018. We use SO₂ emission rates from a previous study, and provide a method to tune the emission rates, such that observed SO₂ column amounts can be reproduced.

The methods and the experimental setup is outlined in Sect. 3, and the results of the numerical experiments are presented in Sect. 4.

2 New submodel for emissions from Explosive Volcanic ERuptions (EVER)

The new submodel EVER is developed as an extension to the second version of MESSy (version 2.55.1, Jöckel et al., 2010), which can be coupled with various basemodels, i. e. General Circulation Models (GCM). MESSy employs strict coding structures across its submodels to ensure portability and high flexibility in chemistry-climate simulations.

2.1 Submodel description

The core of EVER is based on the MESSy submodel TREXP (Jöckel et al., 2010), primarily employed for artificial tracer studies and capable of emitting point sources and uniformly distributed columns of trace gases. To enable the simulation of volcanic eruptions, novel functionalities were introduced as part of the new EVER submodel, including the incorporation of different types of vertical distributions, primary emission of aerosol species, and seamless coupling with aerosol submodels.

Volcanic eruptions or other local atmospheric emission sources can be simulated and controlled via namelist. Each volcanic eruption or emission is initiated using the following parameters:

- Geographical location (latitude and longitude)
- Type of vertical distribution (see Sect. 2.1.2)
- Altitude range (minimum to maximum altitude) [km above surface level (asl)]
- Midpoint [km asl] and sigma [km] of the eruption plume (only needed for Gaussian vertical distributions)
- Period of the (volcanic) emission (date and time of start and end)
- List of emitted tracers (corresponding to tracer names)
- List of tracer masses (in Tg)
- List of aerosol parameter sets (see Sect. 2.1.1)

The EVER module code converts the individual emitted tracer masses into tracer increments, that are added to the total mixing ratios during runtime, uniformly distributed over the specified time range, within the grid-column over the defined location, and following a user-specified vertical distribution. Aerosol parameter sets only need to be provided for aerosol species, and are defined as described in Sect. 2.1.1. An example Fortran95 namelist setup for the EVER submodel is provided in the supplement (ever_example.nml), in which both SO₂ and volcanic ash are emitted using the same vertical distribution and time frame. If the temporal or spatial extents differ, new emission points must be defined accordingly.

2.1.1 Primary aerosol emissions

The MESSy framework includes several alternative submodels for interactively simulating atmospheric aerosols. The new EVER submodel is currently designed to flexibly interface with modal aerosol submodels to emit primary aerosol, and was tested for the three MESSy aerosol submodels GMXe (Pringle et al., 2010), MADE3 (Kaiser et al., 2014, 2019) and PTRAC (Jöckel et al., 2008), while future coupling to sectional aerosol models is possible with adjustments to the interface. However, currently MESSy does not contain any sectional aerosol submodel. While PTRAC only requires an increase of the mixing ratio of the tracer for primary aerosol emissions (as diameter and density of each aerosol tracer remain fixed), the submodels GMXe and MADE3 require an additional increase in number concentration of the corresponding aerosol size mode. For these two submodels, the respective channel name ("aermod_channel" in namelist, see supplement) has to be provided.

The emitted number concentration is calculated based on aerosol parameter sets, which can be defined via Fortran95 namelist
 145 using the following variables:

- Aerosol parameter set name (referenced by the volcanic eruption points)
- Density of the emitted aerosols ρ
- Median emission particle diameter of the emitted species d_{md} [m]
- Size mode as defined in the aerosol submodel

150 The median emission particle diameter should reside within the diameter boundaries of the corresponding aerosol mode to ensure proper treatment of the emission. The sigma of the log-normal distribution (σ_{ln}) is obtained from the defined aerosol submodel for the given size mode. In EVER v1.0, which is included in the latest MESSy releases 2.55.1 and 2.55.2, primary emissions were handled differently. Specifically, EVER could only be directly integrated with GMXe, requiring the GMXe mode as a string, while sigma had to be explicitly provided for other submodels. The improved coupling with aerosol submodels
 155 was introduced in EVER v1.1 and will be available in all subsequent releases.

On the basis of the aerosol parameter sets, number concentrations are calculated using

$$N_{aer} = \frac{6 \cdot M_{trac}}{\pi \cdot \rho \cdot \exp(3 \ln d_{md} + 4.5 \ln^2 \sigma_{ln})} \quad (1)$$

where M_{trac} denotes the mass of the emitted species and ρ , d_{md} and σ_{ln} as described above.

2.1.2 Vertical distributions

160 As volcanic plumes typically span a range of altitudes rather than being centred at a specific altitude, it is reasonable to specify vertical distributions as well for the corresponding emissions. Presently, two types of vertical distributions are supported, with the potential for expansion in the future:

1. **Uniform distribution:** In this distribution, the mass is uniformly distributed between the minimum and maximum altitudes, proportionally to the height of the grid cell. The lowermost and uppermost grid cells within the altitude range are
 165 filled based on the fraction of the grid cell covered by the altitude range.
2. **Gaussian distribution:** In this distribution, the mass follows a Gaussian-shaped vertical profile with mean altitude and sigma specified in the EVER emission namelist. It can be confined to a vertical extent as for the **Uniform distribution**, truncating the tail and scaling accordingly. The emission amount in each grid cell is calculated by considering the fraction of the error function integrated from the bottom to the top of the grid cell (for the lowermost and uppermost grid cells,
 170 the minimum and maximum altitudes are used, respectively) relative to the integral of the error function across the entire confined vertical extent.

Although each injection is realized within a single column of grid boxes horizontally, multiple emission points can be defined to accurately reproduce the observed horizontal extent of a single eruption. This is especially important for large volcanic

plumes entering the stratosphere, exceeding the area of one grid box (Schallock et al., 2023). Tilmes et al. (2023) showed, that the emission of the Pinatubo SO₂ plume over a horizontal area, rather than in one single column, leads to a significantly improved agreement with observations. However, in this study we only used one emission point for each eruption.

2.2 Historic volcanic eruption namelist setup

A primary aim of this study is the development of a methodology, that automatically integrates volcanic eruptions significantly impacting the stratosphere in standard simulations using the MESSy framework, and thus reproduces stratospheric SO₂ mixing ratios. Therefore, we established a historic volcanic eruption namelist configuration for the EVER submodel based on the SO₂ emission inventory developed by Schallock et al. (2023), which we have now extended to the period from 1990 to 2023 (encompassing 774 significant explosive volcanic eruptions), and refined with observations from the IASI satellite.

The emission inventory from Schallock et al. (2023) provides information regarding the mass of emitted SO₂ reaching the stratosphere, the plume altitude as observed from satellites, the initial satellite observation date, and the geographic coordinates of the volcano. This inventory was translated into an EVER namelist. The development of the emission inventory involved satellite observations and information from the Global Volcanism Program, Smithsonian Institute (<https://volcano.si.edu/>, last access: 25 March 2024). It is important to note that this inventory may not include all relevant volcanic events, and we encourage the community to contribute additional significant volcanic events to the extendable namelist.

The emission inventory is refined using IASI observations as follows: For each volcano, we conduct a scan of both, temporal and spatial parameters, extending ± 10 days from the emission inventory date and $\pm 10^\circ$ latitude and $\pm 15^\circ$ longitude from the volcano's geographical coordinates. From this analysis, we extract the horizontal space-time point exhibiting the maximum stratospheric SO₂ mixing ratios observed by IASI as the optimal estimate for both, timing and geographical location for injecting the plume into the stratosphere.

The SO₂ mass is then distributed vertically in a Gaussian profile centred 1 km below the maximum altitude (sigma of 2 km, confined to the vertical extent of the maximum plume altitude down 2 km, truncating the Gaussian distribution at $\sigma/2$) provided in the emission inventory from Schallock et al. (2023), at the horizontal geographical location derived from IASI observations, over 6 hours around the identified date and time of peak mixing ratio. In reality, eruption duration and plume vertical extent may vary, and can be adjusted for the study of specific eruptions. It is important to note that IASI became operational only in 2007 and primarily observes larger volcanoes. SO₂ injections from volcanic eruptions occurring before 2007 or those not observed by IASI are introduced with the same vertical distribution, however, at the geographical location of the volcano and from 9:00 to 15:00 UTC on the date provided by the emission inventory from Schallock et al. (2023). Consequently, these emissions are subject to uncertainties as discussed later.

We provide the namelist setup as supplement (ever_historic_stratVolcanoes.nml) for direct application in numerical simulations with the EVER submodel. The 54 strong injections with optimized horizontal geographical location and timing of the stratospheric entry point (e. g. Raikoke 2019, Ulawun 2019, Taal 2020) are marked accordingly.

Table 1. Summary of satellite instruments and the respective observed quantities used for the evaluation of model simulations with the EVER submodel. More technical details in the observations and the applied retrievals can be found in the provided references.

Instrument & Satellite	Observation method	Retrieved quant.	Operation	Reference
Infrared Atmospheric Sounding Interferometer (IASI) – MetOp	Thermal infrared emission from Earth (nadir)	SO₂ – column + altitude retrieval	2006–now	Clarisse et al. (2012, 2014)
Ozone Monitoring Instrument (OMI) – Aura	UV & vis. solar backscatter radiation from Earth’s surface (nadir)	SO₂ – total column	2004–now	Li et al. (2020)
Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) – ENVISAT	Mid-infrared emission spectrometer (limb)	SO₂ – 3D (stratosphere)	2002–2012	Höpfner et al. (2013, 2015)
TROPOspheric Monitoring Instrument (TROPOMI) – Sentinel-5P	As OMI, extended to UV, visible, near-IR & shortwave	SO₂ – column (troposphere)	2017–now	Theys et al. (2017)
Optical, Spectroscopic and Infrared Remote Imaging System (OSIRIS) – ODIN	Spectrally dispersed, scattered sunlight (limb)	Aerosol extinction at 750 nm – 3D (strat.)	2001–now	Rieger et al. (2019)

3 Methods

3.1 Observations

SO₂ and optical properties of aerosols in the atmosphere are continuously monitored by satellites. We use satellite observations of volcanic plumes for the evaluation of the new submodel, shortly summarized in Table 1. More details can be found in the provided references. Section 3.2 outlines which observations were used for which study. The versions of the used datasets are provided in the data availability section.

Satellites typically do not directly observe the retrieved variables but rather functions of these variables, which rely on certain a priori information (Rodgers and Connor, 2003; Rodgers, 1990; Raspollini et al., 2006). Additionally, observed values are influenced by the horizontal and vertical resolutions used for retrieval, leading to contributions from neighbouring layers. To account for this effect, averaging kernel matrices (AKM) can be applied to model data for comparison purposes. In this analysis, AKMs are used for comparisons with MIPAS observations. It is important to note that it is not possible to invert the AKM to correct the retrieved product.

3.2 EMAC model and experimental set-up

We perform three separate numerical model experiments for the evaluation of the new submodel EVER and the established historic namelist setup. All simulations are performed with the ECHAM5/MESSy Atmospheric Chemistry (EMAC) model (Jöckel et al., 2006) that couples MESSy2 (Jöckel et al., 2010) to the general circulation model ECHAM5 (version 5.3.02, Roeckner et al., 2003). Temperature, the logarithm of the surface pressure, divergence and vorticity are “nudged” (more details

by Jöckel et al., 2006; Jeuken et al., 1996) towards meteorological reanalysis data (ERA5, Hersbach et al., 2020) from the European Centre for Medium-Range weather forecasts (ECMWF) below 100 hPa. In addition, we employ the QBO submodel to weakly nudge the simulations to QBO zonal wind observations between 10 and 90 hPa (Giorgetta et al., 2002) to avoid a phase drift. Simulations are performed at T63 horizontal resolution for the simulation of stratospheric volcanic emissions, and at T255 for the degassing case.

Gas-phase chemistry is addressed by the MECCA submodel (Sander et al., 2019). Details on the selected chemical mechanism in the different experiments are given below. Emissions from anthropogenic and biogenic sources, and from biomass burning are introduced as described by Kohl et al. (2023). Sedimentation, dry and wet deposition are simulated using the submodels SEDI, DDEP (both Kerkweg et al., 2006), and SCAV (Tost et al., 2006), respectively. Namelist setup, chemical mechanism and runscript for the stratospheric (Sect. 4.1 & 4.2) and the Kilauea (Sect. 4.3) simulation can be found in the supplement.

3.2.1 SO₂ in explosive volcanic eruptions — Nabro (2011)

First, we evaluate the EVER submodel and investigate the sensitivity of the resulting SO₂ plume to the injection parameters with the eruption of the Nabro volcano, starting on 13 June 2011 (Global Volcanism Program, 2011). The volcanic cloud predominantly comprised water and SO₂, reaching up to 20 km in altitude. As it was observed by a number of satellite instruments in contrast to the stronger eruption of Mount Pinatubo, it offers a perfect case study to investigate the spatio-temporal evolution of volcanic SO₂ in the stratosphere.

Figure 1 illustrates the evolution of the plume during the first days after the initial eruption, as observed from IASI and OMI. The top panel provides SO₂ height retrievals from IASI (Clarisse et al., 2014). The second and third row show retrieved SO₂ columns from IASI and OMI, respectively. SO₂ columns derived from IASI are presented only for coordinates where the retrieved SO₂ height exceeds 14 km, whereas OMI observes the total column. It is noteworthy that the precise timing of the satellite overpasses does not align for IASI and OMI observations.

The initial stratospheric plume followed a northwestward trajectory during its ascent, entering the stratosphere at approximately 18°N, 30° E, and an altitude of up to 18 km (see Fig. 1 a-c0). Within the UTLS, the SO₂ plume is influenced by the Asian Monsoon Anticyclone, subsequently evolving in a northeastward direction (Fig. 1 a-c1). In the night of June 15 to June 16, an additional plume entered the stratosphere, as evident in the IASI observations from June 16 (Fig. 1 a2). Concurrently, a wind shear within the AMA around the tropical tropopause induced a separation between tropospheric and stratospheric SO₂, with the stratospheric component further north (Fig. 1 a2,3). Subsequently, the SO₂ originating from the stratospheric entries mixed and was transported in the anticyclone. Approximately 10 days after the initial eruption, stratospheric SO₂ was distributed widely across the displayed region (not shown).

The difference between the stratospheric IASI SO₂ observations and OMI total columns confirms the altitude-specific findings depicted in the top panel of Fig. 1, enabling a distinction between tropospheric and stratospheric contributions to the total column. Furthermore, it highlights that while the Nabro volcano continuously emitted, plumes entered the stratosphere exclusively on June 13 and June 16. It is debated, if these stratospheric entries resulted from a direct injection caused by the

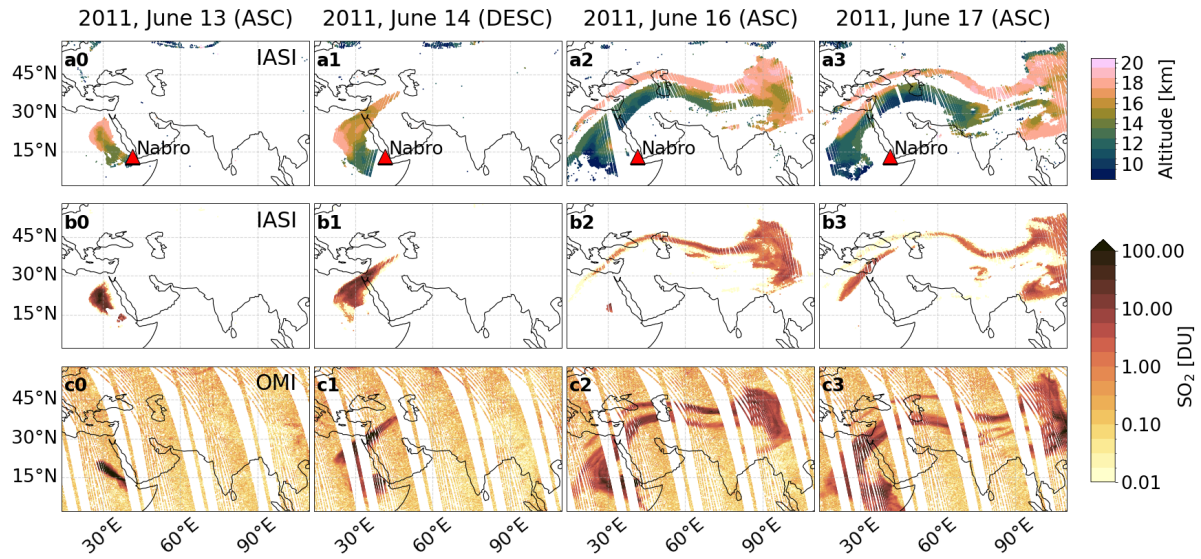


Figure 1. Observations of the Nabro plume on selected dates in the first week after the Nabro eruption. a) SO₂ plume height retrievals from IASI satellite observations (Clarisse et al., 2014) during ascending (ASC) and descending (DESC) orbits. b) Derived SO₂ columns from IASI (Clarisse et al., 2014) observations. c) Derived SO₂ columns from OMI (Li et al., 2020) observations. IASI SO₂ columns are calculated assuming that all SO₂ is centred at the retrieved plume height (top row), and we only display pixels where the retrieved SO₂ plume is detected in the stratosphere (above 14 km), whereas OMI displays the total column. Note that the timing of the observations does not coincide in general.

eruption, or if the initial plumes failed to reach the stratosphere, subsequently being uplifted within the South Asian monsoon system (Bourassa et al., 2012b; Fromm et al., 2013; Vernier et al., 2013; Bourassa et al., 2013; Clarisse et al., 2014). Especially the second stratospheric plume on June 16 could comprise remnants of the tropospheric plume from June 13, that are uplifted. This study does not engage in this ongoing discussion, but exclusively concentrates on the stratospheric entrance points of the plume.

For the Nabro eruption, we carried out a series of EMAC simulations to assess the sensitivity of stratospheric SO₂ burdens to varying emission source parameters. The simulations are performed at T63 horizontal resolution (approx. 190 × 190 km at the equator), with 90 vertical levels up to 0.01 hPa and a model timestep of 8 minutes. The SO₂ injection parameters of the reference simulation are based on the emission inventory developed by Schallock et al. (2023), refined with IASI observations (see below). We employed the Mainz Isoprene Mechanism version 1 (MIM1; Pöschl et al., 2000; Jöckel et al., 2016) within MECCA (see supplement for detailed mechanism), considering oxidation of SO₂ with OH to form SO₃, subsequently reacting with H₂O to form H₂SO₄. Carbonyl sulfide (OCS), a precursor for stratospheric aerosols, is constrained using monthly averaged surface concentrations as outlined by Montzka et al. (2007). Dimethyl sulfide (DMS) emissions from the ocean are

270 computed using the MESSy submodel AIRSEA (Pozzer et al., 2006) and global ocean surface DMS concentrations derived by Lana et al. (2011) in the stratospheric setup.

The following simulations were performed:

- **reference**: Column emission centred at altitude from the emission inventory minus 1 km (17 km); Gaussian vertical distribution with a sigma of 2 km, confined to the altitude range from 16 to 18 km (see Sect. 2.1.2); horizontal position and timing derived from IASI (one column encompassing 22.9°N, 29.7°E on June 13, 2011, 16:00 - 22:00 UTC); SO₂ amount (406 kt) from the emission inventory from Schallrock et al. (2023).
- **optimized**: Same as **reference**, but with the total amount distributed on two stratospheric entry points (67% on June 13 and 33% on June 16 based on the qualitative findings from the IASI observations).
- **reduced**: Same as **reference**, but with reduced SO₂ emissions (280 kt).
- 280 – **volc_pos**: Same as **reference**, but emissions injected at the geographical location of the volcano (13°N, 41°E).
- **point**: Same as **reference**, but using only one single grid-box at the emission inventory altitude minus 1 km (17 km).
- **min_2days**: Same as **reference**, but emissions shifted by two days (June 11, 2011, 16:00 - 22:00 UTC).
- **plus_1km**: Same as **reference**, but emissions shifted by 1 km in altitude (18 km).
- 285 – **mills_et_al**: Emissions as described by Mills et al. (2016). The emissions are injected over several days and in columns covering the free troposphere and the UTLS region, ranging from 2.5 to 17 km. The only partly stratospheric emission takes place on June 13, emitting 1500 kt, uniformly distributed over the altitude range from 9.7 to 17 km.

IASI observations of SO₂ columns and plume altitude are used for the qualitative evaluation of the short-term plume evolution (first week after the eruption; Sect. 4.1.1), by sampling simulated SO₂ columns at the time of the satellite’s overpass using the SORBIT submodel (Jöckel et al., 2010). For the mid-term plume evolution and quantitative assessment of simulated SO₂ mixing ratios, we use three-dimensional observations from the MIPAS instrument³ (Sect. 4.1.2).

295 For the comparison with MIPAS observations, we calculate the stratospheric SO₂ burden, defined here as the sum over the SO₂ mass from lower integration limits depending on latitude (16 km for 0-30°, 14 km for 30-60° and 12 km for 60-90°)⁴ up to 30 km altitude. We investigate the sensitivity of the model-observation agreement to the lower integration limit in Appendix A, showing changes in absolute values at similar agreement when varying the lower integration limit, thus justifying the usage of fixed lower integration limits for the evaluation.

³MIPAS observations are not well-suited for the short-term evaluation due to potential rejection of plume air masses and saturation of spectral lines (Höpfner et al., 2015).

⁴as the model tropopause does not necessarily coincide with the observed tropopause

3.2.2 Multi-year simulation with historic eruption namelist

To evaluate the new historic eruption namelist (EVER v1.1, see Sect. 2.2), we performed an EMAC simulation from January 2008 to December 2011, configured as detailed in Sect. 3.2.1. This timeframe is characterized by high volcanic activity, including three strong eruptions — Kasatochi (August 2008), Sarychev (June 2009) and Nabro (June 2011) — alongside several smaller eruptions. Over the simulated four years, a total of 107 stratospheric injections from volcanic events are documented by Schallock et al. (2023) and considered in the simulation.

We use 3-dimensional observations from the MIPAS instrument to evaluate the total stratospheric SO₂ burden (Sect. 4.2.1) with the same lower integration limit as described above. Whilst this study primarily focuses on the evaluation of simulated SO₂, this being the emitted species handled by EVER, we present here also the EMAC simulated extinction and sAOD at 750 nm, evaluated with observations from the OSIRIS instrument (Bourassa et al., 2007, 2012a; Rieger et al., 2019) in the tropics (0-25°N, lower integration limit 16 km for the calculation of sAOD) and at higher northern latitudes (45-80°N, lower integration limit 12 km), as the tropopause altitude is fairly constant in these regions (Sect. 4.2.2). The additional evaluation of the simulated aerosol optical properties serves as a self-consistency check for the model, for context within the climate impacts of the resulting stratospheric aerosol, and as a comparison to the simulation done by Schallock et al. (2023), who used a similar setup.

Aerosol microphysics are treated with the submodel GMXe (Pringle et al., 2010). In our simulations, we apply four hydrophilic modes (Nucleation, Aitken, Accumulation, and Coarse) and three hydrophobic modes (Aitken, Accumulation, and Coarse). We use the same GMXe setup as detailed in Schallock et al. (2023). Inorganic aerosol thermodynamics are treated with ISORROPIA-II (Fountoukis and Nenes, 2007), and sulfuric acid-water nucleation follows the parameterization by Vehkamäki et al. (2002). We calculate simulated AERosol OPTical properties of the GMXe aerosol populations with the AEROPT sub-model (Dietmüller et al., 2016), providing extinction coefficients and aerosol optical thickness of each model layer at various wavelengths.

3.2.3 SO₂ from a tropospheric degassing case — Kilauea (2018)

In addition to emissions from explosive volcanic eruptions, we evaluate the new submodel’s capability to simulate emissions of degassing volcanoes by analysing a series of eruptive fissures at the Kilauea volcano in Hawaii, USA, in summer 2018 (Kern et al., 2020). SO₂ emissions from Kilauea were studied with the EMAC model before for a different period (Beirle et al., 2014).

Simulations for the degassing case are performed at a horizontal resolution of T255 (approximately 50 × 50 km at the equator) and 31 model levels (up to an altitude of about 30 km) to capture the tropospheric transport. For these simulations, we use a simpler chemical mechanism (compared to the T63 simulations) in the global model covering the basic tropospheric chemistry (as we do not focus on the stratosphere here), including O₃, OH, NO_x, NO_y and basic sulfur chemistry (see supplement for details), to reduce the computational cost at this increased horizontal resolution and decreased timestep of 2.5 minutes. Oxidation of SO₂ to H₂SO₄ is directly realized via reaction with OH in this simplified chemistry, without producing any

intermediates. We do not consider DMS and OCS here (see supplement), leading to a potential underestimation of background maritime SO₂ concentrations.

The Kilauea SO₂ emissions are clearly observed from TROPOMI (ESA, 2018), serving as a basis for comparison with simulated columns. Jost (2021) derived daily SO₂ emission rates for Kilauea based on TROPOMI observations (SO₂ repro v1.1) and wind fields from ECMWF, using the divergence method developed by Beirle et al. (2019). Within this study, we use the original emission rates scaled up by a factor of 4.3 following recent updates, resulting in higher emission estimates. These adjustments include:

- (a) A factor of 3.2 due to non-linear effects caused by strong SO₂ absorption in the Kilauea volcanic plume (compare Theys et al., 2017). While Jost (2021) initially deemed this effect negligible, we included it after re-examination.
- (b) A factor of 1.12 due to the change of the assumed plume height from 2000 m to 1000 m (compare Kern et al., 2020).
- (c) A factor of 1.19 from the consideration of topographic effects on the divergence calculation (Sun, 2022; Beirle et al., 2023).

To explore the potential for further emission optimization, we perform simulations with tuned SO₂ emission rates (see Fig. 9), based on the results of the reference simulation with the emission rates from Jost (2021). For that purpose, we establish a linear relationship between SO₂ emissions and simulated SO₂ by fitting the implemented emission rates of the three preceding days to the resulting simulated SO₂ columns at each day in June 2018 (the month of most intense SO₂ emissions), and subsequently use this linear relationship to derive optimized emission rates resulting in the observed columns from TROPOMI, applying a stochastic gradient descent algorithm (e. g. Ruder, 2016). More details on the optimization can be found in Appendix B.

4 Evaluation & Results

4.1 SO₂ in explosive volcanic eruptions — Nabro (2011)

4.1.1 Short-term SO₂ plume evolution

Figures 2 and 3 compare the simulated altitude of maximum SO₂ mixing ratios and SO₂ columns from the reference and sensitivity simulations to the corresponding IASI observations for the morning overpass on June 14 (one day after the initial Nabro eruption) and the afternoon overpass on June 17. SO₂ emissions in the simulation are confined to the stratosphere (except the approach by Mills et al. (2016)), and thus only stratospheric amounts (altitude ≥ 14 km) are shown. Indeed, this approach neglects the significant amount of tropospheric SO₂ injected during the eruption. However, tropospheric SO₂ typically has a much shorter lifetime compared to stratospheric SO₂, which we focus on in this study. The **reduced** simulation is not shown in these figures, as it is equivalent to the **reference** in altitude and only exhibits reduced columns.

Overall, the simulated columns appear to be underestimated when compared to observations (see Fig. 3b*). However, this might be attributed to the retrieval procedure of the column from the IASI observations. The column estimation assumes that

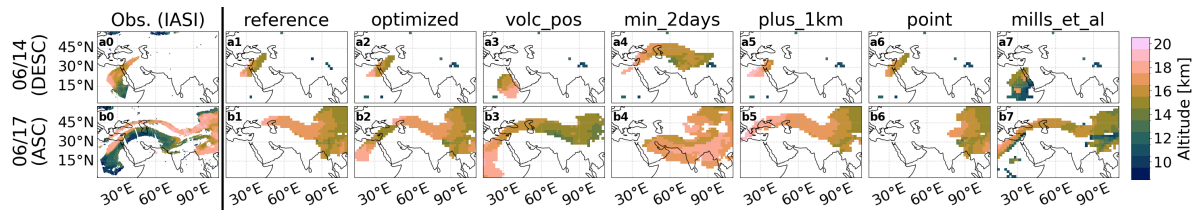


Figure 2. Derived plume altitude from the IASI satellite observations (left; compare Fig. 1) is compared with the altitude of maximum SO₂ mixing ratios for each vertical column from the sensitivity simulations, one day (top) and four days (bottom) after the initial Nabro eruption. In the simulations SO₂ was only injected into the stratosphere, except for **mills_et_al**.

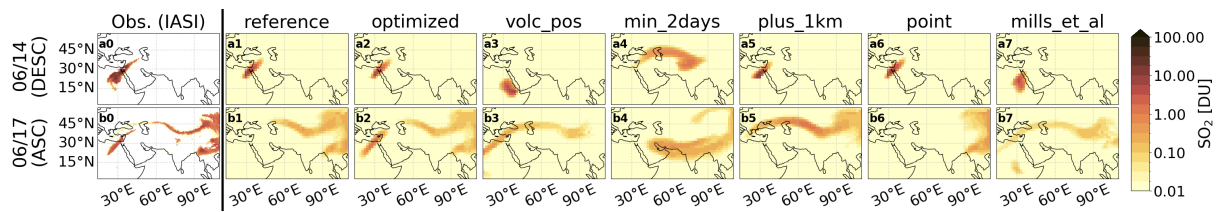


Figure 3. Column SO₂ derived from IASI satellite observations (left; compare Fig. 1) is compared with the respective column from the sensitivity simulations, one day (top) and four days (bottom) after the initial Nabro eruption. In the simulations SO₂ was only injected into the stratosphere, except for **mills_et_al**.

all SO₂ of the plume is centred at the respective altitude depicted in Fig. 2. However, it takes into account the complete column, also including the tropospheric part of the plume. Hence, we only conduct a qualitative comparison between the simulated and observed column.

The **reference** simulation appears to reasonably capture the stratospheric evolution when considering only pixels where the altitude exceeds 14 km, although some discrepancies are evident. While the simulated altitude distribution horizontally broadens over time (Fig. 2 b1), the total column analysis (Fig. 3, June 17 **reference**) shows that the columns at the edges of the plume are very low, falling below the detection limit of IASI. Notably, as the **reference** simulation assumes only one stratospheric entrance, it fails to reproduce the observed second plume as expected (compare Fig. 3 b0 and b1).

In the **optimized** simulation the emissions are distributed across two space-time points, based on a detailed analysis of the IASI observations (refer to Fig. 1). As a result, the second plume is successfully reproduced, leading to better agreement with the observations (Fig. 3 b2). By varying the geographical location of the stratospheric entrance (**volc_pos**), the plume encounters different meteorological patterns, resulting in a distinct evolution pattern. Similarly, adjusting the timing parameter (**min_2days**) leads to a more advanced evolution within the anticyclone (Fig. 3 b4). These sensitivity analyses highlight the importance of accurately representing the timing and location of stratospheric injections for capturing the short-term evolution of volcanic plumes in atmospheric models.

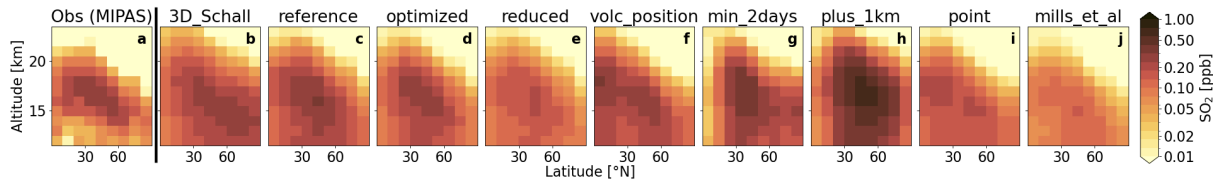


Figure 4. Zonally and 5-day averaged (July 14 - July 18, 2011) profile of northern hemispheric stratospheric SO₂ mixing ratios derived from MIPAS observations (leftmost panel) is compared with the respective SO₂ mixing ratios simulated in the reference and sensitivity simulations one month after the eruption of the Nabro volcano.

As previously mentioned, a vertical wind shear leads to a displacement of the stratospheric part of the plume towards higher latitudes. Moreover, there appears to be a vertical gradient in wind speed above 16 km. When the emission altitude is increased by 1 km (**plus_1km**), the evolution within the anticyclone is attenuated (Fig. 2 b5), suggesting lower wind speeds at higher altitudes. Additionally, the **point** simulation, where all emissions are centred at 17 km, only reproduces the rapid branch of the observed plume evolution. Consequently, it does not encounter the lower wind speeds experienced at higher altitudes (Fig. 2 b6). In contrast, Mills et al. (2016) implemented emissions over several days and uniformly over altitudes ranging from 2.5 to 17 km, accounting for the continuous emissions that did not reach the stratosphere. However, in the **mills_et_al** simulation, stratospheric emissions are underestimated, leading to a substantial fraction of emissions being lost due to scavenging in the free and upper troposphere (Fig. 2 b6).

4.1.2 Mid-term SO₂ mixing ratios

The zonal and 5-day averaged profiles of simulated and observed (MIPAS) stratospheric SO₂ mixing ratios on July 16, 2011, approximately one month after the eruption, are shown in Figure 4 for the Northern Hemisphere. In addition to the sensitivity simulations discussed earlier, a simulation using 3D emission fields of SO₂ mixing ratios (**3D_Schall** in the following) is included for comparison (Schallock et al., 2023). These emission fields were derived from various satellite observations and applied several days after the initial eruption, specifically on June 21 for the Nabro eruption. Details on the methodology are described by Schallock et al. (2023). To simulate the effect of limited vertical resolution associated with MIPAS observations, the AKM (see Sect. 3.1) was applied to all simulated SO₂ mixing ratios.

The comparison between simulated and observed SO₂ distributions reveals some discrepancies, particularly in the vertical extent of the elevated SO₂ mixing ratios. The SO₂ distributions exhibit a slightly larger vertical extent compared to the observations. This discrepancy suggests a potential overestimation of the AKM or limitations in the vertical resolution of the simulation at the respective altitudes (approximately 500 meters). The **3D_Schall** simulation, which uses 3D emissions derived directly from satellite observations, shows the widest distribution (b). This discrepancy can be attributed to the fact that the 3D emissions already incorporate the smoothing effects introduced during the retrieval process (i. e. the AKM). Consequently, the simulated SO₂ mixing ratios in the **3D_Schall** simulation represent observed mixing ratios rather than actual mixing ratios. Therefore, after applying the AKM to these simulated mixing ratios, the resulting altitude resolution appears too wide.

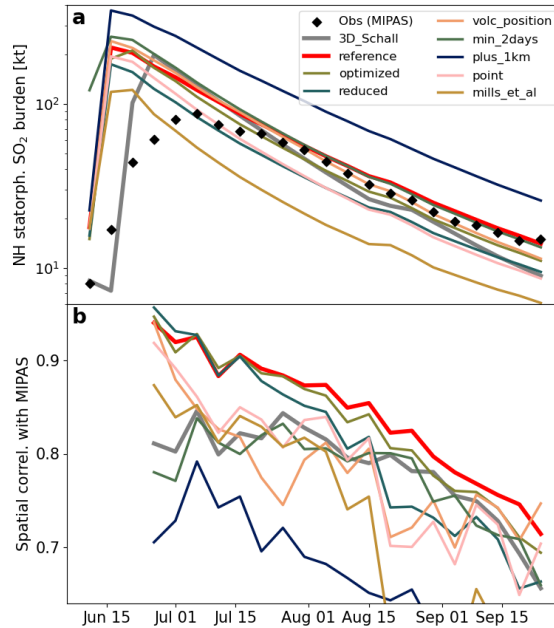


Figure 5. The top panel displays the total northern hemispheric (NH) stratospheric SO₂ burden as observed from the MIPAS satellite (black dots) and from the sensitivity simulations (5-day averages). In the bottom panel, we depict the spatial correlation in the latitude-altitude plane between the simulations and MIPAS observations, i. e. the spatial correlation between the first and all other panels in Fig. 4.

The sensitivity studies exhibit consistent patterns, with the highest mixing ratios of SO₂ typically observed between 15 and 20 km in altitude and 20° and 60° N in latitude, and decreasing altitudes of highest SO₂ mixing ratios with increasing latitude. The distribution follows the typical stratospheric circulation pattern and resembles the observed distribution. Lower mixing ratios compared to observations are found in the **reduced**, **point**, and **mills_et_al** simulations. In the **reduced** simulation, the reduced emissions directly lead to lower mixing ratios (e). However, in the **mills_et_al** simulation, a significant portion of SO₂ is removed in the upper troposphere, as discussed earlier (j). In the **point** simulation, restricting emissions to a single grid box omits the altitude range between 17 and 18 km, leading to a reduced stratospheric lifetime (i). Conversely, the **plus_1km** simulation shows higher mixing ratios due to the increased stratospheric lifetime associated with higher injection altitudes. The **volc_pos** (f) and **min_2days** (g) simulations exhibit slightly different spatial distributions, with the former indicating higher and the latter lower SO₂ mixing ratios at low latitudes. Hence, the varied meteorological conditions experienced in the initial days post-eruption consistently lead to diverse mid-term evolutions of stratospheric SO₂.

In addition to examining the spatial distributions at specific time points, we explored the mid-term changes of northern hemispheric (NH) stratospheric SO₂ burden (Fig. 5a) and the mid-term spatial agreement between observed and simulated SO₂ mixing ratios (Fig. 5b) after the Nabro eruption. MIPAS observations of NH stratospheric SO₂ burden exhibit a gradual

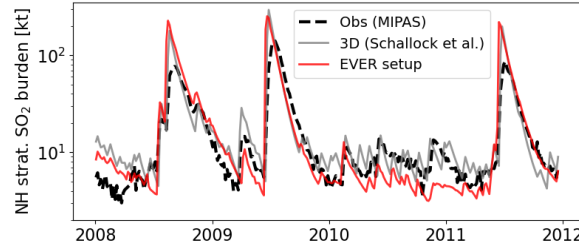


Figure 6. Timeline of total northern hemispheric (NH) stratospheric SO₂ mass as observed from the MIPAS instrument (black dashed line) compared with simulations using the EMAC model with the new EVER historic volcanic setup (red) and using 3D emission fields (Schallock et al., 2023).

increase following the volcanic eruption, unlike the simulations, as discussed earlier. The **3D_Schall** simulation's onset is delayed by a week, as the simulation is based on observations after the first plume evolution in the stratosphere. The bottom panel of Fig. 5 illustrates the spatial correlation between the zonal profile of MIPAS observations and the sensitivity studies within the altitude-latitude window depicted in Fig. 4.

The stratospheric SO₂ burden of the **reference**, **optimized**, **volc_pos** and **min_2days** simulations follow the same logarithmic decay, mostly coinciding with the observations after mid-July. These simulations also exhibit very similar spatial correlations with some fluctuations, showing correlations around 0.9 approximately 3 weeks after the eruptions, which gradually decrease to values between 0.75 and 0.8 in late September. The **min_2days** simulation shows a weaker correlation in the initial phase but approaches the others in the mid-term, most likely due to the different initial meteorological conditions.

The **reduced**, **point** and **mills_et_al** simulations exhibit consistently lower stratospheric SO₂ mass as observed earlier, while the decay is parallel to the aforementioned. The spatial correlation of the **reduced** and **point** simulations is similar to the **reference** simulation, while the **mills_et_al** simulation shows a slightly smaller but comparable correlation. Emissions at higher altitudes (**plus_1km**) lead to higher SO₂ burden, longer lifetime, and lower spatial correlation with the observations. Initially, the simulation with three-dimensional emissions (**3D_Schall**) shows a comparable total stratospheric SO₂ burden to the reference simulation. However, it displays a slightly shorter lifetime as the burden decays more rapidly. Additionally, a slightly lower spatial correlation is observed, potentially attributed to the wider distribution of emissions.

The overall slightly faster decline of the stratospheric SO₂ burden in the simulation compared to the observations appears consistent across all simulations. It can either be attributed to an overestimation of the chemical removal, i. e. the oxidation with OH, or too efficient transport from the stratosphere to the troposphere, that also depends on the injection altitude and to a lesser extent the injection location.

4.2 Evaluation of the historic eruption namelist setup in a multi-year simulation from 2008 to 2011

4.2.1 Stratospheric SO₂ burden

Figure 6 illustrates total NH stratospheric SO₂ burdens, analogously to the upper panel of Fig. 5 for the simulated timeframe from January 2008 to December 2011. We compare our simulation using the new EVER setup to the simulation from Schallock et al. (2023) using 3D emissions and observations from MIPAS.

The three primary peaks are largely reproduced similarly in both simulations, as anticipated, given that the injection mass from the emission inventory is derived from the 3D emission fields in the work of Schallock et al. (2023). However, in general, the observed peaks are lower than the simulated ones. This discrepancy can be attributed to the limited reliability of MIPAS observations shortly after strong eruptions (see Sect. 3.2.1 and 4.1.2 for details), underestimating actual SO₂ mixing ratios.

The EVER simulation exhibits an underestimation of background SO₂ mixing ratios, particularly evident during the relatively quiet period from the end of 2010 until the eruption of Nabro in June 2011, whereas the 3D simulation mostly reproduces the background mixing ratios. The discrepancy in the EVER setup is likely attributable to limitations in the general model configuration or the overestimation of the vertical extent of the emissions for smaller eruptions, only reaching the tropopause. Future efforts will concentrate on enhancing the representation of background SO₂ and aerosol in both, the free and upper troposphere, as well as in the stratosphere. However, it is worth noting that the simulation does reproduce smaller volcanic events, albeit at reduced magnitudes. As previously discussed, these smaller injections are not optimized in terms of horizontal position and timing, as they fall below the detection limit of IASI.

4.2.2 Optical properties of the resulting stratospheric aerosol

The main focus of the observational evaluation of EVER is on the spatio-temporal evolution of volcanic SO₂ as the emitted species. However, once in the atmosphere, the SO₂ can convert to sulfur aerosol particles, with e-folding times varying between 2 and 40 days (Carn et al., 2016; Höpfner et al., 2015). For an additional validation and comparison to the simulation performed by Schallock et al. (2023), we compare the optical properties of the resulting stratospheric aerosol to observations from the OSIRIS instrument.

Figure 7 shows the sAOD for the four simulated years, categorized into two regions: the NH tropics (0° to 25°N) and the mid- to higher latitudes in the northern hemisphere (45°N to 80°N). In both regions, the three major peaks corresponding to the volcanic eruptions are evident. Discrepancies between the EVER simulation and observations are expected in regions where the stratospheric injection occurred, attributable to cloud overlap and saturation effects in the observations. This discrepancy is observed for Kasatochi (2008) and Sarychev (2009) at higher latitudes, and for Nabro at lower latitudes. Conversely, the 3D emissions, directly derived from satellite observations and applied with a delay, reproduce these measurement biases.

In the aftermath of the Nabro eruption in June 2011, the sAOD in the EVER simulation exhibits a sharp increase followed by notable fluctuations. The fluctuations can be attributed in part to the movement of the plume centre across the 25° latitude band, resulting in variable alignment with the observed latitude window. The fluctuations could also be a consequence of

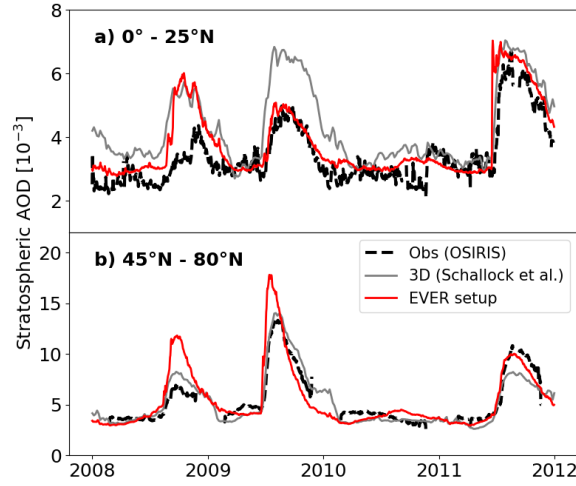


Figure 7. Timeline of sAOD at 750 nm (for the vertical range of the integration see Sect.3.2.2) as observed from the OSIRIS instrument (black dashed line) compared with simulations using the EMAC model with the new EVER historic volcanic setup (red) and with 3D emission fields (Schallock et al., 2023). The sAOD is evaluated in the northern hemispheric tropical latitudinal band from 0° to 25°N (top) and at higher northern latitudes from 45° to 80°N (bottom).

microphysical processes, that are outside of the scope of the manuscript. Notably, the observations show similar fluctuations in the following.

Another interesting feature is the pronounced overestimation of sAOD in the tropical latitudes following the Kasatochi eruption. This anomaly could be attributed to an overestimation of transport from higher latitudes to the tropical stratosphere, or a general overestimation of the emissions. Notably, this feature is not observed after the Sarychev eruption in the EVER simulation, although it is present in the 3D simulation. The discrepancies between the two simulations in the mid-term transport after the Sarychev (to the tropics) and Nabro (to high latitudes) eruptions may be attributed to the different timing of the emissions. While emission times in the EVER simulation were optimized to the first detection of stratospheric SO₂ from the IASI satellite, 3D emissions were applied on the dates specified in Schallock et al. (2023). The interaction with the South Asian monsoon anticyclone potentially causes differing transport to lower or higher latitudes, respectively, if altitude, timing and geographical location of the emission do not coincide.

Figure 8 displays the aerosol extinction at 750 nm, as observed from the OSIRIS instrument and simulated. The effect of the three major volcanic eruptions is mostly evident between 16 and 20 km in the tropics, and between 12 and 18 km at higher latitudes. Similar to Fig. 7, the magnitude of extinction is differing between the observations and the two simulations. In addition, discrepancies are noticeable in the maximum altitude of the plume and below the tropopause. While the 3D simulation largely reproduces the observed maximum altitude, using satellite observations with, in case of MIPAS, the incorporated AKM as input, the EVER simulation may present more realistic maximum altitudes. The differences between the observations and

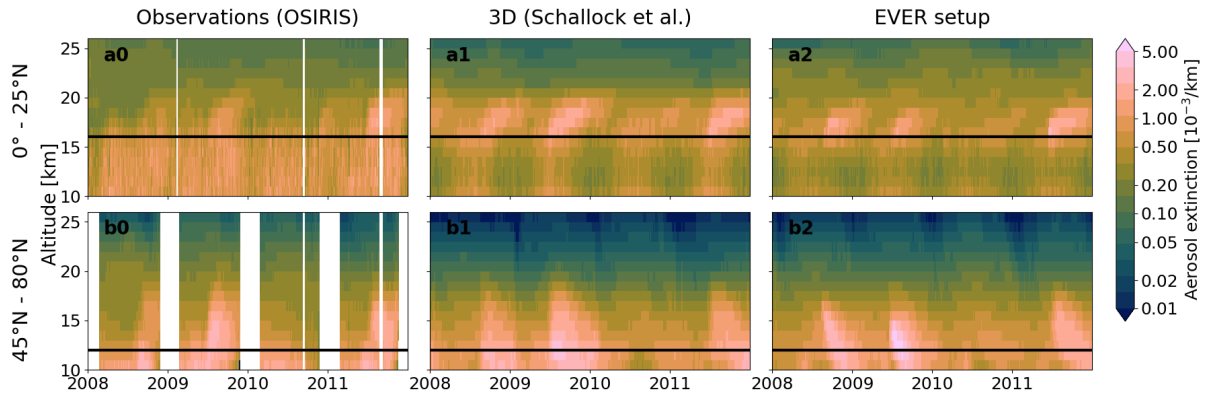


Figure 8. Timeline of aerosol extinction at 750 nm as observed from the OSIRIS instrument (left) and simulated by the EMAC model using 3D emissions (middle; Schallock et al., 2023) and using the new EVER historic volcanic setup (right), evaluated in the northern hemispheric tropical latitudinal band from 0° to 25°N (top), and at higher northern latitudes from 45° to 80°N (bottom).

the simulation below the tropopause are strongly driven by the coincidence with clouds which hinder the retrieval of aerosol extinction. The slight differences between the two simulations from 10 to 15 km in the tropics can most likely be attributed to the differences between the simulation setups. In addition, the QBO signal with differing aerosol concentrations above 20 km, depending on the QBO phase (e. g. Hommel et al., 2015), is more pronounced in the simulation, potentially subject to future studies.

In the sensitivity simulations from Brodowsky et al. (2021), the emission database from Brühl et al. (2018) consistently showed the lowest stratospheric aerosol amounts compared to the other emission databases in bad agreement with observations. However, we show much better agreement with observations in our simulations and see even higher SO₂ mixing ratios, when applying the emission inventory of Schallock et al. (2023, extension of Brühl et al. (2018)) compared to the emission database from Mills et al. (2016). This contradiction can be attributed to the vertical extent of the emission. In Brodowsky et al. (2021), "the emitted SO₂ plume is assumed to be evenly distributed [from] the given plume top downwards one third of the way to the Earth's surface", which would mean a vertical extent of 12 to 18 km for the Nabro volcano. While this approach seems reasonable for the databases of Diehl et al. (2012) and Carn et al. (2017), as they consider the tropospheric and stratospheric part of the plume, it leads to a vast underestimation of the stratospheric SO₂ when applied on the stratospheric emissions by Brühl et al. (2018). In this study, SO₂ is emitted between 16 and 18 km for the Nabro volcano in our simulations.

4.3 SO₂ from degassing volcanoes — Kilauea (2018)

In the following, we investigate the degassing event at Kilauea. Figure 9 depicts a comparison between observed columns from TROPOMI in the top row (a*) and simulated columns from the simulations with derived emission rates from Jost (2021) (Jost2021, b*) and with optimized emission rates (optimized, c*). Generally, SO₂ columns are highest in proximity to the

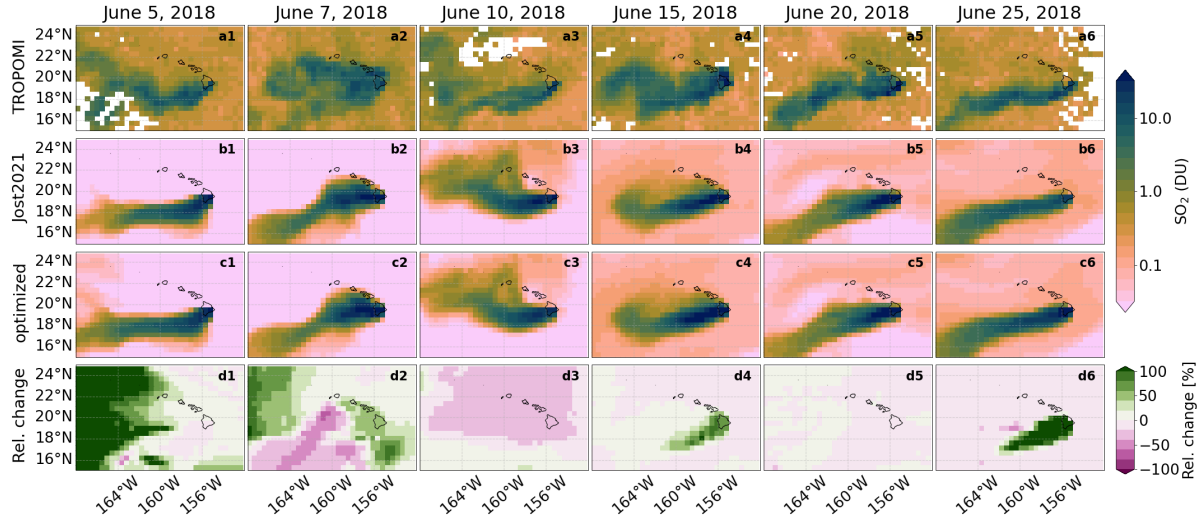


Figure 9. Observed (top) and simulated SO_2 columns resulting from the degassing of the Kilauea volcano at selected days in June 2018 at a model resolution of T255. a) Observations from TROPOMI, regridded to T255 resolution. b) Simulation with emission rates derived by Jost (2021, scaled by a factor of 4.3 – see text for details). c) Simulation with optimized emission rates, based on a comparison between model and observations (refer to the text for more details). d) Relative change from the simulation with derived emission rates by Jost (2021) to the optimized simulation.

Kilauea volcano and disperse according to meteorological conditions. Qualitatively, we observe reasonable agreement in the horizontal dispersion of the plume on most days (Fig. 9), with some exceptions, such as June 7 and June 10. However, there is stronger variability in the observations within the transported plume compared to the simulations, where we predominantly observe gradually decreasing SO_2 columns with distance from the volcano. This effect is attributed to the diurnal variability of the SO_2 emissions, which are not represented in the emission dataset.

Figure 10 provides a quantitative assessment of the horizontal extent depicted in Figure 9 (spanning from -168° to -152° W, 15° to 25° N), showing spatially averaged SO_2 column within this horizontal window, $\overline{\text{SO}_{2(\text{col},d)}}$ as observed and simulated at each day in June 2018 d (bottom), and spatial correlation between simulated and observed logarithmic SO_2 columns (top) within this horizontal window. The **Jost2021** numerical results exhibits some noticeable fluctuations in $\overline{\text{SO}_{2(\text{col},d)}}$, with periods of underestimation in the initial days and on June 25, as well as instances of significant overestimation, such as on June 11 and June 21. The presence of a low-bias can be primarily attributed to missing or very low emission data from Jost (2021) due to missing orbits or cloud cover hindering reasonable SO_2 retrieval. As expected, the simulated $\overline{\text{SO}_{2(\text{col},d)}}$ from the **optimized** simulation follows the observed pattern much more closely as a result of the tuning.

On most days, a strong spatial correlation between the simulated and observed horizontal dispersion of the SO_2 plume is evident, ranging from 0.6 to 0.9. However, the pronounced exceptions on June 7 and June 10 (also depicted in Fig. 9) may

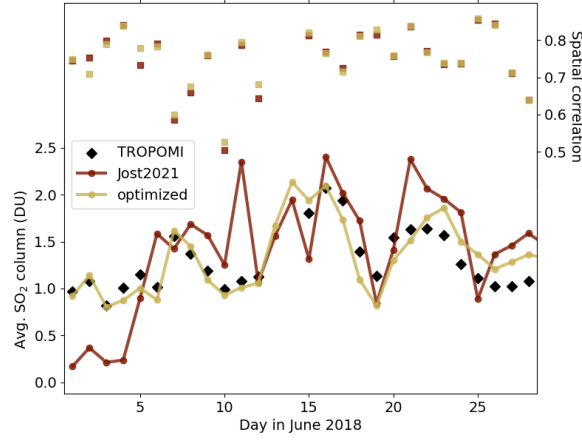


Figure 10. Derived spatially averaged SO₂ column from TROPOMI observations (-168° to -152° W, 15° to 25° N; horizontal window displayed in Fig. 9) in June 2018 compared with simulations, using emission rates from Jost (2021, scaled by a factor of 4.3 – see text for details) and optimized emission rates, based on a comparison between model and observations. On the top, we present spatial correlations in the same horizontal window between the observations and the respective simulations, using the same colour as for the SO₂ column.

be attributed to misrepresentations of the meteorological conditions or generally lower wind speeds, leading to more turbulent flow. Barely any improvement in spatial correlation can be observed from the **Jost2021** simulation to the **optimized** one. This lack of difference is primarily due to the intra-day fluctuations of SO₂ emissions, which contribute to the observed variations and are also not accounted for in the optimized simulation, and potential differences in injection height, that are not considered in the simulation. To address the former, emission rates would need to be implemented at higher temporal resolution (e. g. hourly) and simulations performed at higher horizontal resolution.

5 Discussion

We showed that SO₂ emissions from explosive volcanic eruptions and the subsequent plume evolution can be reasonably reproduced in EMAC within the MESSy model system, using either 3D emissions (Schallock et al., 2023) derived from satellite observations, or column emissions with differing vertical profiles. The different approaches exhibit strengths and weaknesses, and reveal information on the general capabilities of the model.

The usage of 3D emissions, as investigated by Schallock et al. (2023), offers significant advantages for assessing the mid- and long-term impact of volcanic eruptions. By directly deriving emissions from 3D satellite observations several days after the initial eruption, this approach ensures an accurate representation of the plume’s horizontal and vertical evolution, particularly during the crucial initial phase post-eruption, which is heavily influenced by local meteorological conditions. However, a notable drawback of this method is the inherent limitation in the vertical sensitivity of the satellite observations, leading to an overestimation of the vertical extent of the plume, and thus not reproducing the real distribution. Consequently, this discrepancy

impacts the plume’s subsequent evolution and results in differing stratospheric lifetimes for the volcanic plume. Additionally, due to the reliance on 3D satellite observations and the delayed injection, short-term volcanic effects cannot be adequately examined for the simulation with 3D emissions. Another limitation of the approach outlined by Schallock et al. (2023) lies in its technical implementation, which necessitates manual addition of retrieved tracer perturbations and subsequent model restarts after each volcanic event or the use of large import files, posing practical challenges for operational use.

Column emissions rely on several assumptions regarding critical parameters such as plume height and location, emitted mass, and emission profile. With sufficient observational data, these parameters can be effectively constrained, enabling accurate predictions of SO₂ mixing ratios. However, the creation and updating of such inventories also needs manual work, retrieving the relevant parameters. As demonstrated with the Nabro volcano, we used data from the volcanic SO₂ emission inventory compiled by Schallock et al. (2023) in combination with observations from IASI to accurately constrain these parameters. Consequently, our model simulations exhibited strong agreement with both, short- and mid-term observations of SO₂ mixing ratios and aerosol optical properties obtained from the IASI, MIPAS, and OSIRIS satellite instruments, specifically evaluated for strong eruptions.

The sensitivity studies revealed that the importance of accurately constraining the emission parameters for adequately simulating the volcanic SO₂ plume differs for each parameter. Primarily, emitting an appropriate quantity of SO₂ at the correct altitude appears to be the most critical factor. Variations in the SO₂ amount directly influence stratospheric SO₂ mixing ratios, with the mixing ratios approximately proportional to the mass emitted. Conversely, discrepancies in the injection altitude substantially impact the stratospheric lifetime of the resulting SO₂ and sulfur aerosol, as well as the meteorological conditions encountered by the plume.

For that reason, we opted for the emission database developed by Brühl et al. (2018) and Schallock et al. (2023) for the historic default namelist setup, as they specifically target SO₂ injections above the tropopause, while the databases from Diehl et al. (2012) and Carn et al. (2017) do not distinguish between the tropospheric and stratospheric part of the plume, providing no information about the SO₂ amount above the tropopause. Mills et al. (2016) do provide a minimum and maximum plume altitude, however potentially underestimate the stratospheric part when uniformly emitting over this altitude range as can be seen in the sensitivity study on the Nabro volcano (see Sect. 4.1.2). The unclear stratospheric contribution of the initial SO₂ emission is also the main reason for the vastly differing stratospheric sulfur burden estimated by Brodowsky et al. (2021) from the different emission databases, again highlighting the importance of the correct retrieval and application of the stratospheric part of the plume. The emission database from Schallock et al. (2023) additionally explicitly includes smaller eruptions reaching the stratosphere.

However, column or point emissions come with inherent limitations as well. First, emissions are constrained to a single grid box or columns of grid boxes in this study. In detailed studies of strong eruptions, we additionally recommend exploring the effects of emissions over multiple columns and an extended time period to avoid non-linearities due to very high local concentrations (see also Sect. 2.1.2). Second, volcanic activity typically extends beyond a single day, with SO₂ emissions occurring over prolonged periods, occasionally reaching the stratosphere (see Sect. 4.1 for the Nabro eruption). While related discrepancies dissipated in the mid-term for the Nabro volcano, this may not be necessarily the case for other volcanic eruptions.

Third, the exact timing and geographical location of the stratospheric entry point cannot always be accurately estimated. Our sensitivity analyses uncovered short- and mid-term disparities when such information is lacking.

Based on the insights from the sensitivity studies, we developed a historical namelist configuration for the new EVER submodel spanning the past three decades. Despite the aforementioned limitations, our evaluation simulation demonstrates a satisfactory alignment with observations of both, SO_2 mixing ratios and aerosol optical properties. Notably, the historic namelist setup accurately reproduces significant eruptions, thereby representing the primary contribution of volcanic events to the stratospheric SO_2 and aerosol loading. This aligns with our conclusion that altitude and mass are the most crucial emission parameters, which were directly determined from the emission inventory and do not depend on the availability of IASI observations. However, we showed that the timing and geographical location of the stratospheric entry point can lead to additional uncertainties, that remain when no IASI observations are available.

Consequently, we recommend the adoption of the submodel EVER with the proposed historical namelist setup (see Sect. 2.2 and supplement) in all numerical simulations using the MESSy framework at global or regional scale, particularly those encompassing the stratospheric and upper tropospheric domains. This is the first study to systematically incorporate volcanic eruptions into atmospheric simulations within MESSy using the EMAC model, presenting a more flexible and easy-to-implement alternative to the 3D emission approach. Furthermore, in cases where disparities with observations arise, owing to the aforementioned uncertainties, or when focusing on specific volcanic events, the namelist setup can be adjusted accordingly, especially regarding the horizontal and vertical extent of the plume. Moreover, comparisons with simulations using 3D emission fields may offer additional insights into the evolution of individual volcanic plumes.

In section 4.3, we demonstrated the additional capability of EVER to simulate SO_2 from degassing volcanoes. However, it is essential to apply a model with a sufficiently fine resolved horizontal grid (for a global model) to accurately capture the observed phenomena and the small-scale wind fluctuations. Simulations with equal emissions performed at more standard horizontal resolutions, such as T63 and T106, failed to reproduce the observations adequately (not shown), whereas these resolutions are mostly sufficient for stratospheric simulations. Furthermore, we applied a model-driven tuning for the corresponding emission rates, reproducing observed SO_2 column amounts.

This optimization has the potential to be extended to historic degassing volcanoes, facilitating the development of a default setup akin to the stratospheric default setup for explosive volcanoes. This process would include integrating TROPOMI observations with an initial simulation using rough estimates of degassing emissions, which could subsequently be refined through stochastic gradient descent (SGD) optimization as described in Appendix B, potentially extending or replacing the outdated climatology by Diehl et al. (2012) used in many EMAC simulations. However, implementing this approach may encounter challenges related to the required high horizontal resolution, the estimation of the injection altitude, and the identification and initial approximate estimation of all significant degassing volcano emissions.

Up to this point, our focus has been primarily on volcanic SO_2 . However, it is worth mentioning the versatility of the submodel for a wide range of use cases where gaseous or aerosol tracers are injected into the atmosphere in vertical distributions, with limited horizontal extent. This includes the following use cases:

- **Volcanic ash:** Apart from emitting trace gases, volcanic eruptions also release primary aerosols, such as volcanic ash. The EVER submodel is explicitly designed to simulate the evolution of aerosol species, including volcanic ash, after volcanic eruptions.
- 605 – **Water vapour:** Eruptions of submarine volcanoes, such as the notable event at Hunga Tonga-Hunga Ha’apai in January 2022 (e. g., Vömel et al., 2022; Sellitto et al., 2022; Schoeberl et al., 2022; Xu et al., 2022), release substantial quantities of water vapor into the atmosphere. The EVER module can be used to investigate the effects of enhanced water vapor concentrations in the stratosphere due to volcanic activity. For the Hunga Tonga eruption we recommend the simultaneous injection of water vapor in multiple columns to avoid quick removal by ice formation and to be consistent with
610 observations.
- **Wildfires:** Strong wildfires can inject significant amounts of carbonaceous aerosols and various trace gases directly into the stratosphere via pyro-cumulonimbi. EVER can be used to model these emissions from wildfires.
- **Solar radiation modification:** Studies on solar radiation modification, particularly artificial injections of SO₂ or other trace gases into the stratosphere to form aerosols that reflect sunlight back into space, can benefit from the capabilities
615 of EVER. These scenarios involve large uncertainties, which can be addressed with studies using EVER.
- **Transport processes:** Transport processes play a crucial role throughout the atmosphere. EVER allows for the emission of active and passive aerosols and trace gases throughout the atmosphere, enabling the study of processes such as the exchange between the troposphere and stratosphere.
- **Sensitivity studies:** Atmospheric properties can be highly sensitive to perturbations in trace gas or aerosol mixing ratios.
620 By injecting the respective atmospheric constituents with EVER, it is possible to estimate the sensitivity of climate, atmospheric dynamics, and the ozone column to these perturbations.

6 Conclusions

We presented the new submodel for tracer emissions from Explosive Volcanic ERuptions (EVER v1.1), developed within the Modular Earth Submodel System (MESSy, version 2.55.1), and performed numerical experiments with the ECHAM5/MESSy
625 Atmospheric Model (EMAC) and the new submodel.

EVER is designed for the addition of gaseous and aerosol tracer tendencies following volcanic eruptions in columns with user-specified vertical profiles at point or area sources. We evaluated the EVER submodel with the simulation of volcanic SO₂ emissions in the EMAC model for the explosive eruption of Nabro in June 2011 and a degassing event from Kilauea in July 2018, employing satellite observations from IASI, MIPAS, OMI, TROPOMI, and OSIRIS. The evaluation showed that
630 volcanic emission plumes can be reasonably simulated with EVER. The new submodel is available from MESSy version 2.55.1 and will be continuously developed further.

We investigated the sensitivity of the volcanic SO₂ plume evolution after the Nabro eruption to variations in the emission parameters. The results emphasized the importance of the emission of a reasonable amount of SO₂ above the tropopause with an appropriate altitude distribution. In previous studies with various emission databases, the SO₂ mass emitted in the stratosphere was unclear, resulting in large differences. We showed that the application of a dedicated stratospheric SO₂ emission inventory can reproduce observed SO₂ burdens and aerosol optical properties. Horizontal position and emission timing were found to have a minor impact on the mid-term SO₂ burden in the stratosphere. Nevertheless, these parameters play a crucial role in detailed process studies during the initial weeks after an eruption.

Furthermore, we conclude that simulations of volcanic eruptions can be effectively performed with the help of 3D- and column emissions, if the emitted stratospheric SO₂ mass is well constrained. However, both approaches have shortcomings. The optimal approach depends on the specific use case, with column emissions excelling in the short-term, and similar performance in the mid- to long-term.

Finally, we developed a historic submodel setup for EVER, incorporating stratospheric significant volcanic eruptions spanning from 1990 to 2023. It is based on the volcanic SO₂ emission inventory by Schallrock et al. (2023). We additionally optimized the timing and geographical location of the volcanic plume entering the stratosphere, using the findings of the sensitivity study and SO₂ observations from the IASI satellite. However, this information was only available from 2007 on, and for strong volcanic eruptions only. The historic namelist setup was successfully evaluated with regard to resulting SO₂ mixing ratios and aerosol optical properties. It is provided as a supplement, and we advocate its inclusion in simulations using the MESSy framework focusing on the upper troposphere and the stratosphere. For very strong eruptions, it may be beneficial to distribute the emissions over multiple columns horizontally and an extended time period, or adjust the vertical plume extent, if discrepancies with observations occur.

In addition to the extensively discussed application to explosive volcanic eruptions, the EVER submodel's versatility opens up a range of additional research opportunities. These may include investigations into the interplay between SO₂ and volcanic ash post-eruption, exploration of solar radiation modification scenarios, modelling of wildfires, and analyses of atmospheric transport processes. Future work could involve the development of a climatology of SO₂ emissions from degassing volcanoes employing the new submodel.

Appendix A: Sensitivity of the evaluation results to the lower integration limit for the calculation of stratospheric properties

As the tropopause does not necessarily coincide in the simulation and the observations, we applied latitude-dependent fixed altitude lower integration limits for the calculation of stratospheric SO₂ burden and sAOD. Here, we study the sensitivity of the evaluation results to the chosen lower integration limit.

The stratospheric SO₂ burden is defined in this work as the SO₂ mass above 16 km from 0-30°, above 14 km from 30-60°, and above 12 km from 60-90°. We compare these stratospheric SO₂ burdens to burdens with perturbed lower integration limits (+1 km: 17,15,13 km; -1 km: 15,13,11 km). The results are depicted in Fig. A1. While the total burden varies in the three

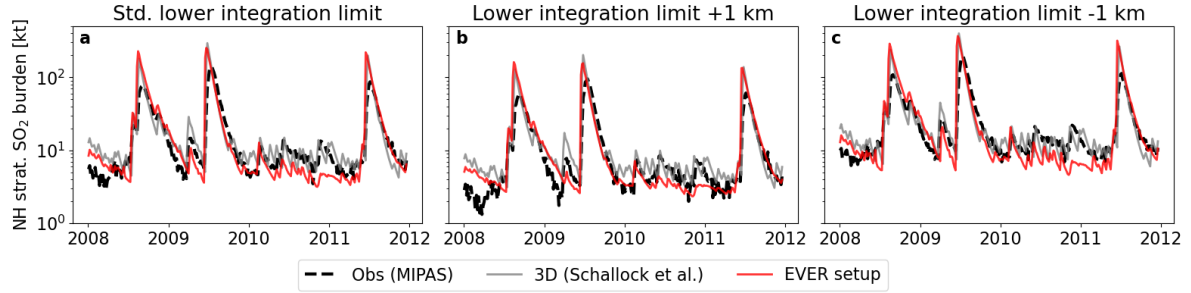


Figure A1. Sensitivity of the evaluation results to the lower integration limit (applied to observations and simulations) for the derivation of the stratospheric SO₂ burden. a) Lower integration limit as applied in the manuscript (16 km from 0-30°N, 14 km from 30-60°N and 12 km from 60-90°N). Variations of plus 1 km (b) and minus 1 km (c) show similar agreement at different absolute values. Slight differences can be seen at background levels with increased agreement in 2010 for the increased lower integration limit (b).

scenarios as expected, the agreement between the simulations and the observations is very similar, especially considering the three major volcanic peaks. However, in the quiet period in 2010 and 2011, the agreement improves, when the lower integration limit increases (b). This can be either attributed to an incorrect altitude distribution of the background SO₂ or to the emission of the smaller eruptions.

The sAOD is defined here as the integral over the aerosol extinction above 16 km from 0-25°, and above 12 km from 45-80°. As before, we vary this lower integration limit by ± 1 km (see Fig. A2). Again, we see similar agreement at differing absolute values. The strongest difference can be seen in the Sarychev peak at higher latitudes (e), that seems to be underestimated when increasing the lower integration limit. This might either be connected to the missing AKM, or the plume top is underestimated.

Overall, we conclude, that our evaluation result are not very sensitive to the chosen lower integration limit for the calculation of the stratospheric SO₂ burden and the sAOD. Thus, the evaluation results are mostly independent on the chosen "tropopause definition".

Appendix B: Optimization of the emission rates from the Kilauea degassing event

In Sect. 4.3, we optimized the emission rates of the Kilauea volcano, such that the model reproduces the observed average SO₂ column $\overline{\text{SO}_{2(\text{col},d,\text{obs})}}$ for each day d over the chosen area. Here, the optimization is described in more detail.

In the first step, we investigated the relation between the implemented emission rates and resulting simulated $\overline{\text{SO}_{2(\text{col},d,\text{sim})}}$. Therefore, we considered the emission rates from the three preceding days of each sampled day in June 2018, denoted as $\text{emis}_{\text{SO}_2;d-i}$ ($i = 0, 1, 2$), and constructed a linear predictor of $\overline{\text{SO}_{2(\text{col},d)}}$:

$$\overline{\text{SO}_{2(\text{col},d,\text{sim})}} = \text{SO}_{2(\text{col},\text{BG})} + \sum_{i=0}^3 a_{d-i} * \text{emis}_{\text{SO}_2;d-i} \quad (\text{B1})$$

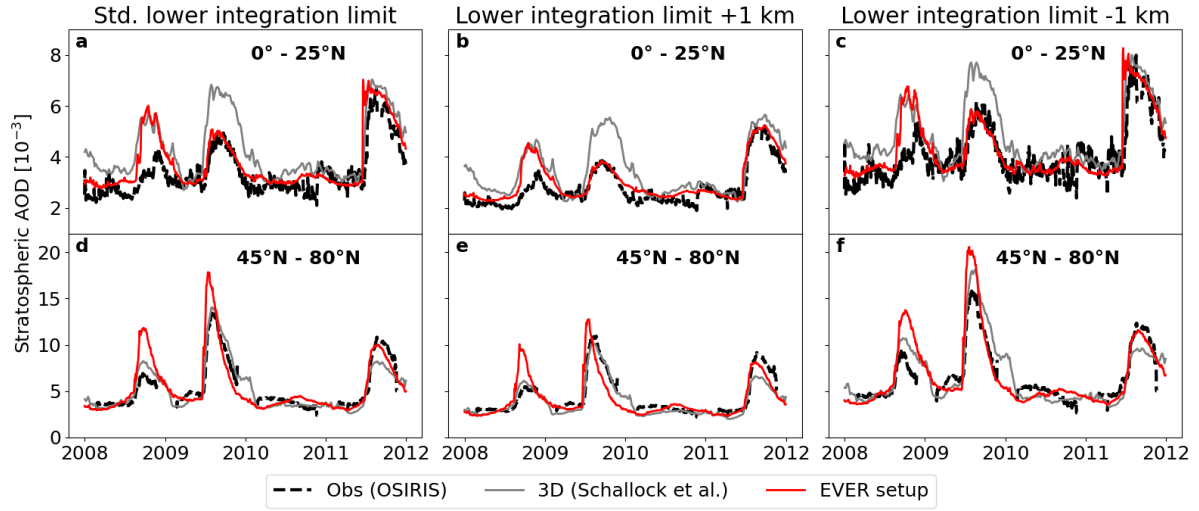


Figure A2. Sensitivity of the evaluation result to the lower integration limit (applied to observations and simulations) for the derivation of sAOD. a,d) Lower integration limit as applied in the manuscript (16 km from 0-25°N and 12 km from 45-80°N). Variations of plus 1 km (b,e) and minus 1 km (c,f) show similar agreement at different absolute values. The most significant differences are observed for the Sarychev volcano, that shows strongly decreased simulated sAOD at increased lower integration limit at higher latitudes (e).

The coefficients a_{d-i} , representing the importance of the emissions of each of the three preceding days, respectively, and the background SO_2 column amount, $\text{SO}_{2(\text{col}, \text{BG})}$, are the free parameters in the linear predictor and were determined through a least squares fit. Subsequently, the linear predictor with these coefficients was utilized to compute optimized emission rates for each day using a stochastic gradient descent (SGD) algorithm, iteratively optimizing the emission rates, by reducing the loss function (e. g. Ruder, 2016). The optimization scheme is given in pseudocode in Algorithm B1.

Algorithm B1 SGD optimizer for daily SO₂ emission rates

Require: niter, lrate

```

1: while j<niter do
2:   for d=0, d<30, k++ do
3:     for k=0, k<3, k++ do
4:       gradient=2*ad-k*(SO2(col,dobs)
        - ∑i=02 ad-i*emisSO2;d-i-SO2(col,BG))
5:       emisSO2;d-k+=lrate*gradient
6:     end for
7:   end for
8:   j++
9: end while

```

Code availability. The Modular Earth Submodel System (MESSy, <https://zenodo.org/doi/10.5281/zenodo.8360186>) is continuously further developed and applied by a consortium of institutions. The usage of MESSy and access to the source code is licenced to all affiliates of institutions which are members of the MESSy Consortium. Institutions can become a member of the MESSy Consortium by signing the MESSy Memorandum of Understanding. More information can be found on the MESSy Consortium Website (<http://www.messy-interface.org>). The code presented here is available in MESSy version 2.55.1 (<https://zenodo.org/records/8367075>). The respective namelists, chemical mechanisms and run scripts used are made available via supplement (see Sect. 3.2).

Scientific colour maps (<https://doi.org/10.5281/zenodo.5501399>, Crameri, 2021) are used in this study to prevent visual distortion of the data and exclusion of readers with colour vision deficiencies (Crameri et al., 2020).

The historic default namelist setup for the new submodel EVER is available as supplement (*ever_historic_stratVolcanoes.nml*).

Data availability. Level 2 and Level 3 (5-day averages) retrieved (V5R_SO2_220 & V5R_SO2_221) SO₂ data from MIPAS observations used in this study is available after registration at <http://www.imk-asf.kit.edu/english/308.php> (Höpfner et al., 2013, 2015). Level 2 OMI SO₂ observations (v003) are taken from https://disc.gsfc.nasa.gov/datasets/OMSO2_003/summary (Li et al., 2020). IASI SO₂ products (linear product version 2) are available at <https://doi.org/10.25326/41> (Clarisse, 2023). We obtained the OSIRIS aerosol product version 7.2 from <https://research-groupstest.usask.ca/osiris/data-products.php> (Rieger et al., 2019). The TROPOMI Level 2 SO₂ product (repro, v1.1) is publicly available at <https://doi.org/10.5270/S5P-yr8kdpp> (ESA, 2018). Model output and setups are archived at the DKRZ in Hamburg, and are available on request. The historic SO₂ emission inventory of explosive volcanic eruptions is available at https://doi.org/10.26050/WDCC/SSIRC_3 (Brühl et al., 2021; Schallock et al., 2023).

705 *Author contributions.* MK and AP planned the research. MK developed the EVER submodel with the help of AP and PJ. JS and CB provided the emission inventory, performed the simulation with 3D emissions, and assisted in the provision of satellite data and information on volcanoes. AJ and SB provided the daily SO₂ emission rates for the Kilauea volcano. MH provided the MIPAS observations with the respective averaging kernel matrix. MK evaluated the simulations, analysed the model results, developed the historic namelist setup, and wrote the manuscript. AP and HT supervised the project. All authors discussed the results and contributed to the review and editing of the
710 manuscript.

Competing interests. Two co-authors are member of the editorial board of Geoscientific Model Developments.

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References

- Beirle, S., Hörmann, C., Penning de Vries, M., Dörner, S., Kern, C., and Wagner, T.: Estimating the volcanic emission rate and atmospheric lifetime of SO₂ from space: a case study for Kīlauea volcano, Hawai‘i, *Atmospheric Chemistry and Physics*, 14, 8309–8322, <https://doi.org/10.5194/acp-14-8309-2014>, 2014.
- Beirle, S., Borger, C., Dörner, S., Li, A., Hu, Z., Liu, F., Wang, Y., and Wagner, T.: Pinpointing nitrogen oxide emissions from space, *Science Advances*, 5, <https://doi.org/10.1126/sciadv.aax9800>, 2019.
- Beirle, S., Borger, C., Jost, A., and Wagner, T.: Improved catalog of NO_x point source emissions (version 2), *Earth System Science Data*, 15, 3051–3073, <https://doi.org/10.5194/essd-15-3051-2023>, 2023.
- Bourassa, A. E., Degenstein, D. A., Gattinger, R. L., and Llewellyn, E. J.: Stratospheric aerosol retrieval with optical spectrograph and infrared imaging system limb scatter measurements, *Journal of Geophysical Research: Atmospheres*, 112, <https://doi.org/https://doi.org/10.1029/2006JD008079>, 2007.
- Bourassa, A. E., Rieger, L. A., Lloyd, N. D., and Degenstein, D. A.: Odin-OSIRIS stratospheric aerosol data product and SAGE III inter-comparison, *Atmospheric Chemistry and Physics*, 12, 605–614, <https://doi.org/10.5194/acp-12-605-2012>, 2012a.
- Bourassa, A. E., Robock, A., Randel, W. J., Deshler, T., Rieger, L. A., Lloyd, N. D., Llewellyn, E. J. T., and Degenstein, D. A.: Large Volcanic Aerosol Load in the Stratosphere Linked to Asian Monsoon Transport, *Science*, 337, 78–81, <https://doi.org/10.1126/science.1219371>, 2012b.
- Bourassa, A. E., Robock, A., Randel, W. J., Deshler, T., Rieger, L. A., Lloyd, N. D., Llewellyn, E. J., and Degenstein, D. A.: Response to Comments on "Large Volcanic Aerosol Load in the Stratosphere Linked to Asian Monsoon Transport", *Science*, 339, 647–647, <https://doi.org/10.1126/science.1227961>, 2013.
- Brodowsky, C., Sukhodolov, T., Feinberg, A., Höpfner, M., Peter, T., Stenke, A., and Rozanov, E.: Modeling the Sulfate Aerosol Evolution After Recent Moderate Volcanic Activity, 2008–2012, *Journal of Geophysical Research: Atmospheres*, 126, e2021JD035472, <https://doi.org/https://doi.org/10.1029/2021JD035472>, 2021.
- Brühl, C., Schallock, J., Klingmüller, K., Robert, C., Bingen, C., Clarisse, L., Heckel, A., North, P., and Rieger, L.: Stratospheric aerosol radiative forcing simulated by the chemistry climate model EMAC using Aerosol CCI satellite data, *Atmospheric Chemistry and Physics*, 18, 12 845–12 857, <https://doi.org/10.5194/acp-18-12845-2018>, 2018.
- Brühl, C., Schallock, J., and Diehl, T.: Volcanic SO₂ data derived from limb viewing satellites for the lower stratosphere from 1990 to 2019, and from nadir viewing satellites for the troposphere, including the corresponding radiative forcing computed by the CCM EMAC, World Data Center for Climate (WDCC) at DKRZ [data set], https://doi.org/10.26050/WDCC/SSIRC_3, 2021.
- Carn, S., Clarisse, L., and Prata, A.: Multi-decadal satellite measurements of global volcanic degassing, *Journal of Volcanology and Geothermal Research*, 311, 99–134, <https://doi.org/10.1016/j.jvolgeores.2016.01.002>, 2016.
- Carn, S., Fioletov, V., McLinden, C., Li, C., and Krotkov, N.: A decade of global volcanic SO₂ emissions measured from space, *Scientific reports*, 7, 44 095, <https://doi.org/10.1038/srep44095>, 2017.
- Clarisse, L.: Daily IASI/Metop-A ULB-LATMOS sulphur dioxide (SO₂) L2 product (columns and altitude, AERIS) [data set], <https://doi.org/10.25326/41>, 2023.
- Clarisse, L., Hurtmans, D., Clerbaux, C., Hadji-Lazaro, J., Ngadi, Y., and Coheur, P.-F.: Retrieval of sulphur dioxide from the infrared atmospheric sounding interferometer (IASI), *Atmospheric Measurement Techniques*, 5, 581–594, <https://doi.org/10.5194/amt-5-581-2012>, 2012.

- Clarisse, L., Coheur, P.-F., Theys, N., Hurtmans, D., and Clerbaux, C.: The 2011 Nabro eruption, a SO₂ plume height analysis using IASI measurements, *Atmospheric Chemistry and Physics*, 14, 3095–3111, <https://doi.org/10.5194/acp-14-3095-2014>, 2014.
- Crameri, F.: Scientific colour maps, Zenodo [code], <https://doi.org/10.5281/zenodo.5501399>, 2021.
- Crameri, F., Shephard, G. E., and Heron, P. J.: The misuse of colour in science communication, *Nature communications*, 11, 1–10, <https://doi.org/10.1038/s41467-020-19160-7>, 2020.
- Diehl, T., Heil, A., Chin, M., Pan, X., Streets, D., Schultz, M., and Kinne, S.: Anthropogenic, biomass burning, and volcanic emissions of black carbon, organic carbon, and SO₂ from 1980 to 2010 for hindcast model experiments, *Atmospheric Chemistry and Physics Discussions*, 12, 24 895–24 954, <https://doi.org/10.5194/acpd-12-24895-2012>, 2012.
- Dietmüller, S., Jöckel, P., Tost, H., Kunze, M., Gellhorn, C., Brinkop, S., Frömming, C., Ponater, M., Steil, B., Lauer, A., and Hendricks, J.: A new radiation infrastructure for the Modular Earth Submodel System (MESSy, based on version 2.51), *Geoscientific Model Development*, 9, 2209–2222, <https://doi.org/10.5194/gmd-9-2209-2016>, 2016.
- Durand, M. and Grattan, J.: Effects of volcanic air pollution on health, *The Lancet*, 357, 164, [https://doi.org/10.1016/S0140-6736\(00\)03586-8](https://doi.org/10.1016/S0140-6736(00)03586-8), 2001.
- European Space Agency (ESA): Copernicus Sentinel-5P (processed by ESA), TROPOMI Level 2 Sulphur Dioxide Total Column. Version 01., <https://doi.org/10.5270/S5P-yr8kdpp>, 2018.
- Fountoukis, C. and Nenes, A.: ISORROPIA II: a computationally efficient thermodynamic equilibrium model for K⁺–Ca²⁺–Mg²⁺–NH₄⁺–Na⁺–SO₄²⁻–NO₃⁻–Cl⁻–H₂O aerosols, *Atmospheric Chemistry and Physics*, 7, 4639–4659, <https://doi.org/10.5194/acp-7-4639-2007>, 2007.
- Fromm, M., Nedoluha, G., and Charvát, Z.: Comment on "Large Volcanic Aerosol Load in the Stratosphere Linked to Asian Monsoon Transport", *Science*, 339, 647–647, <https://doi.org/10.1126/science.1228605>, 2013.
- Giorgetta, M. A., Manzini, E., and Roeckner, E.: Forcing of the quasi-biennial oscillation from a broad spectrum of atmospheric waves, *Geophysical Research Letters*, 29, 86–1–86–4, <https://doi.org/10.1029/2002GL014756>, 2002.
- Global Volcanism Program: Report on Nabro (Eritrea) (Wunderman, R., ed.), *Bulletin of the Global Volcanism Network*, 36:9, <https://doi.org/10.5479/si.GVP.BGVN201109-221101>, 2011.
- Hersbach, H., Bell, B., Berrisford, P., Hirahara, S., Horányi, A., Muñoz-Sabater, J., Nicolas, J., Peubey, C., Radu, R., Schepers, D., Simmons, A., Soci, C., Abdalla, S., Abellan, X., Balsamo, G., Bechtold, P., Biavati, G., Bidlot, J., Bonavita, M., De Chiara, G., Dahlgren, P., Dee, D., Diamantakis, M., Dragani, R., Flemming, J., Forbes, R., Fuentes, M., Geer, A., Haimberger, L., Healy, S., Hogan, R. J., Hólm, E., Janisková, M., Keeley, S., Laloyaux, P., Lopez, P., Lupu, C., Radnoti, G., de Rosnay, P., Rozum, I., Vamborg, F., Villaume, S., and Thépaut, J.-N.: The ERA5 global reanalysis, *Quarterly Journal of the Royal Meteorological Society*, 146, 1999–2049, <https://doi.org/https://doi.org/10.1002/qj.3803>, 2020.
- Hommel, R., Timmreck, C., Giorgetta, M. A., and Graf, H. F.: Quasi-biennial oscillation of the tropical stratospheric aerosol layer, *Atmospheric Chemistry and Physics*, 15, 5557–5584, <https://doi.org/10.5194/acp-15-5557-2015>, 2015.
- Höpfner, M., Glatthor, N., Grabowski, U., Kellmann, S., Kiefer, M., Linden, A., Orphal, J., Stiller, G., von Clarmann, T., Funke, B., and Boone, C. D.: Sulfur dioxide (SO₂) as observed by MIPAS/Envisat: temporal development and spatial distribution at 15–45 km altitude, *Atmospheric Chemistry and Physics*, 13, 10 405–10 423, <https://doi.org/10.5194/acp-13-10405-2013>, 2013.
- Höpfner, M., Boone, C. D., Funke, B., Glatthor, N., Grabowski, U., Günther, A., Kellmann, S., Kiefer, M., Linden, A., Lossow, S., Pumphrey, H. C., Read, W. G., Roiger, A., Stiller, G., Schlager, H., von Clarmann, T., and Wissmüller, K.: Sulfur dioxide (SO₂) from MIPAS in the

upper troposphere and lower stratosphere 2002–2012, *Atmospheric Chemistry and Physics*, 15, 7017–7037, <https://doi.org/10.5194/acp-15-7017-2015>, 2015.

Intergovernmental Panel on Climate Change (IPCC): Climate Change 2021 – The Physical Science Basis: Working Group I Contribution to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change, Cambridge University Press, <https://doi.org/10.1017/9781009157896>, 2023.

Jeuken, A. B. M., Siegmund, P. C., Heijboer, L. C., Feichter, J., and Bengtsson, L.: On the potential of assimilating meteorological analyses in a global climate model for the purpose of model validation, *Journal of Geophysical Research: Atmospheres*, 101, 16 939–16 950, <https://doi.org/10.1029/96JD01218>, 1996.

Jöckel, P., Tost, H., Pozzer, A., Brühl, C., Buchholz, J., Ganzeveld, L., Hoor, P., Kerkweg, A., Lawrence, M. G., Sander, R., Steil, B., Stiller, G., Tanarhte, M., Taraborrelli, D., van Aardenne, J., and Lelieveld, J.: The atmospheric chemistry general circulation model ECHAM5/MESy1: consistent simulation of ozone from the surface to the mesosphere, *Atmospheric Chemistry and Physics*, 6, 5067–5104, <https://doi.org/10.5194/acp-6-5067-2006>, 2006.

Jöckel, P., Kerkweg, A., Buchholz-Dietsch, J., Tost, H., Sander, R., and Pozzer, A.: Technical Note: Coupling of chemical processes with the Modular Earth Submodel System (MESSy) submodel TRACER, *Atmospheric Chemistry and Physics*, 8, 1677–1687, <https://doi.org/10.5194/acp-8-1677-2008>, 2008.

Jöckel, P., Kerkweg, A., Pozzer, A., Sander, R., Tost, H., Riede, H., Baumgaertner, A., Gromov, S., and Kern, B.: Development cycle 2 of the Modular Earth Submodel System (MESSy2), *Geoscientific Model Development*, 3, 717–752, <https://doi.org/10.5194/gmd-3-717-2010>, 2010.

Jöckel, P., Tost, H., Pozzer, A., Kunze, M., Kirner, O., Brenninkmeijer, C. A. M., Brinkop, S., Cai, D. S., Dyroff, C., Eckstein, J., Frank, F., Garny, H., Gottschaldt, K.-D., Graf, P., Grewe, V., Kerkweg, A., Kern, B., Matthes, S., Mertens, M., Meul, S., Neumaier, M., Nützel, M., Oberländer-Hayn, S., Ruhnke, R., Runde, T., Sander, R., Scharffe, D., and Zahn, A.: Earth System Chemistry integrated Modelling (ESCiMo) with the Modular Earth Submodel System (MESSy) version 2.51, *Geoscientific Model Development*, 9, 1153–1200, <https://doi.org/10.5194/gmd-9-1153-2016>, 2016.

Jost, A.: Determining the SO₂ emission rates of the Kilauea volcano in Hawaii using S5P-TROPOMI satellite measurements, Bachelor’s thesis, Johannes Gutenberg Universität Mainz, 2021.

Kaiser, J. C., Hendricks, J., Righi, M., Riemer, N., Zaveri, R. A., Metzger, S., and Aquila, V.: The MESSy aerosol submodel MADE3 (v2.0b): description and a box model test, *Geoscientific Model Development*, 7, 1137–1157, <https://doi.org/10.5194/gmd-7-1137-2014>, 2014.

Kaiser, J. C., Hendricks, J., Righi, M., Jöckel, P., Tost, H., Kandler, K., Weinzierl, B., Sauer, D., Heimerl, K., Schwarz, J. P., Perring, A. E., and Popp, T.: Global aerosol modeling with MADE3 (v3.0) in EMAC (based on v2.53): model description and evaluation, *Geoscientific Model Development*, 12, 541–579, <https://doi.org/10.5194/gmd-12-541-2019>, 2019.

Kerkweg, A., Sander, R., Tost, H., and Jöckel, P.: Technical note: Implementation of prescribed (OFFLEM), calculated (ONLEM), and pseudo-emissions (TNUDGE) of chemical species in the Modular Earth Submodel System (MESSy), *Atmospheric Chemistry and Physics*, 6, 3603–3609, <https://doi.org/10.5194/acp-6-3603-2006>, 2006.

Kern, C., Lerner, A. H., Elias, T., Nadeau, P. A., Holland, L., Kelly, P. J., Werner, C. A., Clor, L. E., and Cappos, M.: Quantifying gas emissions associated with the 2018 rift eruption of Kilauea Volcano using ground-based DOAS measurements, *Bulletin of Volcanology*, 82, 55, <https://doi.org/10.1007/s00445-020-01390-8>, 2020.

Klobas, J. E., Wilmouth, D. M., Weisenstein, D. K., Anderson, J. G., and Salawitch, R. J.: Ozone depletion following future volcanic eruptions, *Geophysical Research Letters*, 44, 7490–7499, <https://doi.org/10.1002/2017GL073972>, 2017.

- Kohl, M., Lelieveld, J., Chowdhury, S., Ehrhart, S., Sharma, D., Cheng, Y., Tripathi, S. N., Sebastian, M., Pandithurai, G., Wang, H., and
830 Pozzer, A.: Numerical simulation and evaluation of global ultrafine particle concentrations at the Earth's surface, *Atmospheric Chemistry
and Physics*, 23, 13 191–13 215, <https://doi.org/10.5194/acp-23-13191-2023>, 2023.
- Kremser, S., Thomason, L. W., von Hobe, M., Hermann, M., Deshler, T., Timmreck, C., Toohey, M., Stenke, A., Schwarz, J. P., Weigel, R.,
Fueglistaler, S., Prata, F. J., Vernier, J.-P., Schlager, H., Barnes, J. E., Antuña-Marrero, J.-C., Fairlie, D., Palm, M., Mahieu, E., Notholt,
J., Rex, M., Bingen, C., Vanhellemont, F., Bourassa, A., Plane, J. M. C., Klocke, D., Carn, S. A., Clarisse, L., Trickl, T., Neely, R., James,
835 A. D., Rieger, L., Wilson, J. C., and Meland, B.: Stratospheric aerosol—Observations, processes, and impact on climate, *Reviews of
Geophysics*, 54, 278–335, <https://doi.org/10.1002/2015RG000511>, 2016.
- Lana, A., Bell, T. G., Simó, R., Vallina, S. M., Ballabrera-Poy, J., Kettle, A. J., Dachs, J., Bopp, L., Saltzman, E. S., Stefels, J., Johnson,
J. E., and Liss, P. S.: An updated climatology of surface dimethylsulfide concentrations and emission fluxes in the global ocean, *Global
Biogeochemical Cycles*, 25, <https://doi.org/https://doi.org/10.1029/2010GB003850>, 2011.
- 840 Li, C., Krotkov, N. A., Leonard, P., and Joiner, J.: OMI/Aura Sulphur Dioxide (SO₂) Total Column 1-orbit L2 Swath 13x24 km V003,
Goddard Earth Sciences Data and Information Services Center (GES DISC) [data set], <https://doi.org/10.5067/Aura/OMI/DATA2022>,
2020.
- Malavelle, F. F., Haywood, J. M., Jones, A., Gettelman, A., Clarisse, L., Bauduin, S., Allan, R. P., Karset, I. H. H., Kristjánsson, J. E.,
Oreopoulos, L., Cho, N., Lee, D., Bellouin, N., Boucher, O., Grosvenor, D. P., Carslaw, K. S., Dhomse, S., Mann, G. W., Schmidt, A.,
845 Coe, H., Hartley, M. E., Dalvi, M., Hill, A. A., Johnson, B. T., Johnson, C. E., Knight, J. R., O'Connor, F. M., Partridge, D. G., Stier,
P., Myhre, G., Platnick, S., Stephens, G. L., Takahashi, H., and Thordarson, T.: Strong constraints on aerosol–cloud interactions from
volcanic eruptions, *Nature*, 546, 485–491, <https://doi.org/10.1038/nature22974>, 2017.
- Mills, M. J., Schmidt, A., Easter, R., Solomon, S., Kinnison, D. E., Ghan, S. J., Neely III, R. R., Marsh, D. R., Conley, A., Bardeen, C. G., and
Gettelman, A.: Global volcanic aerosol properties derived from emissions, 1990–2014, using CESM1(WACCM), *Journal of Geophysical
850 Research: Atmospheres*, 121, 2332–2348, <https://doi.org/10.1002/2015JD024290>, 2016.
- Montzka, S. A., Calvert, P., Hall, B. D., Elkins, J. W., Conway, T. J., Tans, P. P., and Sweeney, C.: On the global distribution, seasonality,
and budget of atmospheric carbonyl sulfide (COS) and some similarities to CO₂, *Journal of Geophysical Research: Atmospheres*, 112,
<https://doi.org/10.1029/2006JD007665>, 2007.
- Neely III, R. and Schmidt, A.: VolcanEESM: Global volcanic sulphur dioxide (SO₂) emissions database from 1850 to present - Version 1.0
855 [Data set], <https://doi.org/10.5285/76ebdc0b-0eed-4f70-b89e-55e606bcd568>, 2016.
- Pöschl, U., von Kuhlmann, R., Poisson, N., and Crutzen, P. J.: Development and intercomparison of condensed isoprene oxidation mecha-
nisms for global atmospheric modeling, *Journal of Atmospheric Chemistry*, 37, 29–52, <https://doi.org/10.1023/A:1006391009798>, 2000.
- Pozzer, A., Jöckel, P., Sander, R., Williams, J., Ganzeveld, L., and Lelieveld, J.: Technical Note: The MESSy-submodel AIRSEA calculating
the air-sea exchange of chemical species, *Atmospheric Chemistry and Physics*, 6, 5435–5444, <https://doi.org/10.5194/acp-6-5435-2006>,
860 2006.
- Pringle, K. J., Tost, H., Message, S., Steil, B., Giannadaki, D., Nenes, A., Fountoukis, C., Stier, P., Vignati, E., and Lelieveld, J.: De-
scription and evaluation of GMXe: a new aerosol submodel for global simulations (v1), *Geoscientific Model Development*, 3, 391–412,
<https://doi.org/10.5194/gmd-3-391-2010>, 2010.
- Quaglia, I., Timmreck, C., Niemeier, U., Visoni, D., Pitari, G., Brodowsky, C., Brühl, C., Dhomse, S. S., Franke, H., Laakso, A., Mann,
865 G. W., Rozanov, E., and Sukhodolov, T.: Interactive stratospheric aerosol models' response to different amounts and altitudes of SO₂

- injection during the 1991 Pinatubo eruption, *Atmospheric Chemistry and Physics*, 23, 921–948, <https://doi.org/10.5194/acp-23-921-2023>, 2023.
- Raspollini, P., Belotti, C., Burgess, A., Carli, B., Carlotti, M., Ceccherini, S., Dinelli, B. M., Dudhia, A., Flaud, J.-M., Funke, B., Höpfner, M., López-Puertas, M., Payne, V., Piccolo, C., Remedios, J. J., Ridolfi, M., and Spang, R.: MIPAS level 2 operational analysis, *Atmospheric Chemistry and Physics*, 6, 5605–5630, <https://doi.org/10.5194/acp-6-5605-2006>, 2006.
- 870 Reifenberg, S. F., Martin, A., Kohl, M., Bacer, S., Hamryszczak, Z., Tadic, I., Röder, L., Crowley, D. J., Fischer, H., Kaiser, K., Schneider, J., Dörich, R., Crowley, J. N., Tomsche, L., Marsing, A., Voigt, C., Zahn, A., Pöhlker, C., Holanda, B. A., Krüger, O., Pöschl, U., Pöhlker, M., Jöckel, P., Dorf, M., Schumann, U., Williams, J., Bohn, B., Curtius, J., Harder, H., Schlager, H., Lelieveld, J., and Pozzer, A.: Numerical simulation of the impact of COVID-19 lockdown on tropospheric composition and aerosol radiative forcing in Europe, *Atmospheric Chemistry and Physics*, 22, 10901–10917, <https://doi.org/10.5194/acp-22-10901-2022>, 2022.
- 875 Rieger, L. A., Zawada, D. J., Bourassa, A. E., and Degenstein, D. A.: A Multiwavelength Retrieval Approach for Improved OSIRIS Aerosol Extinction Retrievals, *Journal of Geophysical Research: Atmospheres*, 124, 7286–7307, <https://doi.org/10.1029/2018JD029897>, 2019.
- Rodgers, C. D.: Characterization and error analysis of profiles retrieved from remote sounding measurements, *Journal of Geophysical Research: Atmospheres*, 95, 5587–5595, <https://doi.org/10.1029/JD095iD05p05587>, 1990.
- 880 Rodgers, C. D. and Connor, B. J.: Intercomparison of remote sounding instruments, *Journal of Geophysical Research: Atmospheres*, 108, <https://doi.org/10.1029/2002JD002299>, 2003.
- Roeckner, E., Bäuml, G., Bonaventura, L., Brokopf, R., Esch, M., Giorgetta, M., Hagemann, S., Kirchner, I., Kornblüeh, L., Manzini, E., Schlese, U., and Schulzweida, U.: The atmospheric general circulation model ECHAM 5. PART I: Model description, Tech. rep., Max-Planck-Institut für Meteorologie, <https://doi.org/10.17617/2.995269>, 2003.
- 885 Ruder, S.: An overview of gradient descent optimization algorithms, arXiv preprint arXiv:1609.04747, 2016.
- Sander, R., Baumgaertner, A., Cabrera-Perez, D., Frank, F., Gromov, S., Groß, J.-U., Harder, H., Huijnen, V., Jöckel, P., Karydis, V. A., Niemeyer, K. E., Pozzer, A., Riede, H., Schultz, M. G., Taraborrelli, D., and Tauer, S.: The community atmospheric chemistry box model CAABA/MECCA-4.0, *Geoscientific Model Development*, 12, 1365–1385, <https://doi.org/10.5194/gmd-12-1365-2019>, 2019.
- Schallock, J., Brühl, C., Bingen, C., Höpfner, M., Rieger, L., and Lelieveld, J.: Reconstructing volcanic radiative forcing since 1990, using a comprehensive emission inventory and spatially resolved sulfur injections from satellite data in a chemistry-climate model, *Atmospheric Chemistry and Physics*, 23, 1169–1207, <https://doi.org/10.5194/acp-23-1169-2023>, 2023.
- 890 Schmidt, A., Mills, M. J., Ghan, S., Gregory, J. M., Allan, R. P., Andrews, T., Bardeen, C. G., Conley, A., Forster, P. M., Gettelman, A., Portmann, R. W., Solomon, S., and Toon, O. B.: Volcanic Radiative Forcing From 1979 to 2015, *Journal of Geophysical Research: Atmospheres*, 123, 12 491–12 508, <https://doi.org/10.1029/2018JD028776>, 2018.
- 895 Schoeberl, M. R., Wang, Y., Ueyama, R., Taha, G., Jensen, E., and Yu, W.: Analysis and Impact of the Hunga Tonga-Hunga Ha’apai Stratospheric Water Vapor Plume, *Geophysical Research Letters*, 49, e2022GL100 248, <https://doi.org/10.1029/2022GL100248>, 2022.
- Sellitto, P., Podglajen, A., Belhadji, R., Boichu, M., Carboni, E., Cuesta, J., Duchamp, C., Kloss, C., Siddans, R., Begue, N., Blare, L., Jegou, F., Khaykin, S., Renard, J.-B., and Legras, B.: The unexpected radiative impact of the Hunga Tonga eruption of 15th January 2022, *Communications Earth & Environment*, 3, 288, <https://doi.org/10.1038/s43247-022-00618-z>, 2022.
- 900 Stewart, C., Damby, D. E., Horwell, C. J., Elias, T., Ilyinskaya, E., Tomašek, I., Longo, B. M., Schmidt, A., Carlsen, H. K., Mason, E., Baxter, P. J., Cronin, S., and Witham, C.: Volcanic air pollution and human health: recent advances and future directions, *Bulletin of Volcanology*, 84, 11, <https://doi.org/10.1007/s00445-021-01513-9>, 2022.

- Sun, K.: Derivation of Emissions From Satellite-Observed Column Amounts and Its Application to TROPOMI NO₂ and CO Observations, *Geophysical Research Letters*, 49, e2022GL101102, <https://doi.org/10.1029/2022GL101102>, 2022.
- 905 Textor, C., Graf, H.-F., Timmreck, C., and Robock, A.: Emissions from volcanoes, in: *Emissions of Atmospheric Trace Compounds*, edited by Granier, C., Artaxo, P., and Reeves, C. E., pp. 269–303, Springer Netherlands, Dordrecht, https://doi.org/10.1007/978-1-4020-2167-1_7, 2004.
- Theys, N., De Smedt, I., Yu, H., Danckaert, T., van Gent, J., Hörmann, C., Wagner, T., Hedelt, P., Bauer, H., Romahn, F., Pedernana, M., Loyola, D., and Van Roozendael, M.: Sulfur dioxide retrievals from TROPOMI onboard Sentinel-5 Precursor: algorithm theoretical basis, *Atmospheric Measurement Techniques*, 10, 119–153, <https://doi.org/10.5194/amt-10-119-2017>, 2017.
- 910 Tie, X. and Brasseur, G.: The response of stratospheric ozone to volcanic eruptions: Sensitivity to atmospheric chlorine loading, *Geophysical Research Letters*, 22, 3035–3038, <https://doi.org/10.1029/95GL03057>, 1995.
- Tilmes, S., Mills, M. J., Zhu, Y., Bardeen, C. G., Vitt, F., Yu, P., Fillmore, D., Liu, X., Toon, B., and Deshler, T.: Description and performance of a sectional aerosol microphysical model in the Community Earth System Model (CESM2), *Geoscientific Model Development*, 16, 6087–6125, <https://doi.org/10.5194/gmd-16-6087-2023>, 2023.
- 915 Timmreck, C., Mann, G. W., Aquila, V., Hommel, R., Lee, L. A., Schmidt, A., Brühl, C., Carn, S., Chin, M., Dhomse, S. S., Diehl, T., English, J. M., Mills, M. J., Neely, R., Sheng, J., Toohey, M., and Weisenstein, D.: The Interactive Stratospheric Aerosol Model Intercomparison Project (ISA-MIP): motivation and experimental design, *Geoscientific Model Development*, 11, 2581–2608, <https://doi.org/10.5194/gmd-11-2581-2018>, 2018.
- 920 Tost, H., Jöckel, P., Kerkweg, A., Sander, R., and Lelieveld, J.: Technical note: A new comprehensive SCAVenging submodel for global atmospheric chemistry modelling, *Atmospheric Chemistry and Physics*, 6, 565–574, <https://doi.org/10.5194/acp-6-565-2006>, 2006.
- Vattioni, S., Stenke, A., Luo, B., Chiodo, G., Sukhodolov, T., Wunderlin, E., and Peter, T.: Importance of microphysical settings for climate forcing by stratospheric SO₂ injections as modeled by SOCOL-AERv2, *Geoscientific Model Development*, 17, 4181–4197, <https://doi.org/10.5194/gmd-17-4181-2024>, 2024.
- 925 Vehkamäki, H., Kulmala, M., Napari, I., Lehtinen, K. E. J., Timmreck, C., Noppel, M., and Laaksonen, A.: An improved parameterization for sulfuric acid–water nucleation rates for tropospheric and stratospheric conditions, *Journal of Geophysical Research: Atmospheres*, 107, AAC 3–1–AAC 3–10, <https://doi.org/10.1029/2002JD002184>, 2002.
- Vernier, J.-P., Thomason, L. W., Fairlie, T. D., Minnis, P., Palikonda, R., and Bedka, K. M.: Comment on "Large Volcanic Aerosol Load in the Stratosphere Linked to Asian Monsoon Transport", *Science*, 339, 647–647, <https://doi.org/10.1126/science.1227817>, 2013.
- 930 Vernier, J.-P., Fairlie, T. D., Deshler, T., Natarajan, M., Knepp, T., Foster, K., Wienhold, F. G., Bedka, K. M., Thomason, L., and Trepte, C.: In situ and space-based observations of the Kelud volcanic plume: The persistence of ash in the lower stratosphere, *Journal of Geophysical Research: Atmospheres*, 121, 11,104–11,118, <https://doi.org/10.1002/2016JD025344>, 2016.
- Vömel, H., Evan, S., and Tully, M.: Water vapor injection into the stratosphere by Hunga Tonga-Hunga Ha’apai, *Science*, 377, 1444–1447, <https://doi.org/10.1126/science.abq2299>, 2022.
- 935 Xu, J., Li, D., Bai, Z., Tao, M., and Bian, J.: Large Amounts of Water Vapor Were Injected into the Stratosphere by the Hunga Tonga-Hunga Ha’apai Volcano Eruption, *Atmosphere*, 13, <https://doi.org/10.3390/atmos13060912>, 2022.
- Zhu, Y., Toon, O. B., Jensen, E. J., Bardeen, C. G., Mills, M. J., Tolbert, M. A., Yu, P., and Woods, S.: Persisting volcanic ash particles impact stratospheric SO₂ lifetime and aerosol optical properties, *Nature communications*, 11, 1–11, <https://doi.org/10.1038/s41467-020-18352-5>, 2020.

5 CHAPTER

Simulating atmospheric aerosols from the surface to the stratosphere

This chapter presents a manuscript draft, to be submitted to *Atmospheric Chemistry & Physics*, pending minor changes. I am the first author of and the main contributor to this manuscript. I developed the new model setup, performed and evaluated the simulations, analysed the results, and wrote the manuscript. More detailed information on the Author contributions are provided at the end of the manuscript.

Global atmospheric aerosol distributions and composition from the surface to the stratosphere

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Abstract. Atmospheric aerosols play a crucial role in Earth's climate system, yet their global spatio-temporal distribution remains poorly constrained, particularly in the free (FT) and upper troposphere (UT), due to limited observational coverage. This lack of understanding of vertical aerosol profiles is a major contributor to the large uncertainties associated with both direct and, in particular, indirect aerosol radiative forcing. To address this knowledge gap, we develop and evaluate a new setup for the ECHAM/MESSy Atmospheric Chemistry (EMAC) model. The evaluation is based on a comprehensive set of stationary, remote-sensing and aircraft observations of aerosol mass, number concentrations, and optical properties, with a focus on the FT, UT, and the lower stratosphere. The evaluation shows good agreement between observed and simulated aerosol properties across all layers and most regions of the lower atmosphere. The improved setup is used to simulate global distributions of key aerosol species, their composition, and number concentrations from the surface to the stratosphere. Aerosol mass concentrations exhibit a consistent global minimum between 400 and 200 hPa, marking the transition from the FT to the upper troposphere/lowermost stratosphere (UTLS) region, separating the boundary layer (BL) and stratosphere-influenced atmospheric layers, with consequent frequent new particle formation. Furthermore, we find that primary particles emitted in the BL are rarely transported into the UT and stratosphere, contrary to previous modelling studies but in closer agreement with recent observations. Stratospheric aerosol mass is dominated by sulfate, with a notable contribution from secondary organic aerosol (SOA) in the lower stratosphere, due to convective transport. This leads to a global tropical band of elevated SOA concentrations in the lower stratosphere that is transported to higher latitudes via the Brewer-Dobson circulation. This work is the basis for future studies on anthropogenic influence on aerosol distributions across all layers of the lower atmosphere, detailed aerosol process studies, and online aerosol-cloud-radiation-interaction. Ultimately, it shall contribute to the reduction

of uncertainties in radiative forcing estimates and to our understanding of aerosol-climate-health effects across all layers of the
20 lower and middle atmosphere.

1 Introduction

Atmospheric aerosols, composed of a mixture of condensed phase particles of varying size and composition, play a key role
in Earth's climate system (Szopa et al., 2021). They originate from both natural (e.g., dust, sea salt, volcanic ash) and an-
thropogenic (e.g., black and organic carbon) sources, either emitted directly or formed through gas-to-particle conversion of
25 oxidized precursors such as sulfur, nitrogen, or organic compounds (e.g., Pöschl, 2005; Boucher, 2015). Once airborne, they
undergo complex mixing, growth, transport, and removal processes, influencing Earth's radiative balance, the hydrological
cycle, atmospheric chemistry, and human health (e.g., Szopa et al., 2021; Lohmann and Feichter, 2005; Murray et al., 2020;
Lelieveld et al., 2015; Klobas et al., 2017).

Uncertainties in our knowledge about aerosol direct and indirect radiative forcing remain the dominant contributor to the
30 total anthropogenic forcing uncertainty in the latest IPCC report (Szopa et al., 2021), largely due to substantial discrepan-
cies between simulated and observed aerosol distributions, insufficient knowledge about vertical aerosol profiles, and limited
model–data intercomparisons. While some CMIP6 models rely on prescribed aerosol climatologies and others implement sim-
plified chemistry schemes, very few represent a comprehensive set of aerosol species that reflect the chemical diversity of the
atmosphere (Szopa et al., 2021). Accurately capturing vertical profiles of aerosol mass, composition, and number concentra-
35 tions from the surface to the stratosphere by numerical models is essential to reduce these uncertainties (Watson-Parris et al.,
2019; Reddington et al., 2017).

Aerosols in the boundary layer (BL) have been extensively studied through a combination of ground-based measurements,
satellite observations and numerical simulations (e.g., van Donkelaar et al., 2016; Lelieveld et al., 2015; Kohl et al., 2023;
Pozzer et al., 2012; Zhang et al., 2007; Gordon et al., 2017), with the aerosol composition and abundance strongly depending
40 on local sources due to the short lifetime of aerosols close to the surface. Stratospheric aerosol is dominated by sulfur species
originating from precursor gases emitted by natural sources (mostly OCS; e.g., Crutzen, 1976; Brühl et al., 2012) and occasional
volcanic injections (e.g., Schmidt et al., 2018; Schallock et al., 2023), forming the Junge layer (Junge et al., 1961), which
spans altitudes between 12 and 28 km. Stratospheric aerosol has been observed through in-situ balloon measurements (e.g.,
Deshler et al., 2003; Hertzog and Plougonven, 2021) and satellite observations (e.g., Vernier et al., 2009; Bourassa et al., 2012;
45 Kovilakam et al., 2023), which provide information about aerosol optical properties, however only offering limited insight into
particle composition and size. The contribution of non-sulfur species to the stratospheric aerosol burden remains particularly
uncertain (e.g., Kremser et al., 2016). For example, while model studies have suggested a dominant role of dust in the upper
troposphere (UT) and lowermost stratosphere (UTLS; e.g., Bossolasco et al., 2021; Ma et al., 2019; Brühl et al., 2018), recent
observations report a very low abundance of primary particles, particularly with mineral dust composition (Ebert et al., 2024;
50 Appel et al., 2022). A more comprehensive review on stratospheric aerosol particles is provided by Kremser et al. (2016).

Aerosol particles and their properties in the free troposphere (FT) and UTLS can only be studied through aircraft- and balloon-based measurements, both of which are spatially and temporally limited, or via satellite observations that provide vertical profiles of aerosol extinction (e.g., Maheshwarkar et al., 2024; Winker et al., 2013). As a result, no global long-term observations of aerosol composition and size distributions exist for these atmospheric regions. Nevertheless, an increasing number of aircraft campaigns have studied atmospheric aerosol populations in the FT and UTLS, focusing either on background conditions (e.g., NASA Atmospheric Tomography (ATom) Mission; Thompson et al., 2022) or on specific atmospheric processes. These studies have exemplarily revealed high concentrations of freshly formed particles in the tropical and subtropical UT (Williamson et al., 2019; Weigel et al., 2021; Curtius et al., 2024), the formation of the Asian Tropopause Aerosol Layer (ATAL) (Vernier et al., 2011; Höpfner et al., 2019; Appel et al., 2022), and widespread presence of biomass burning and dust particles in the troposphere (Schill et al., 2020; Froyd et al., 2022).

Given the spatial and temporal limitations of observations in the FT and UTLS, numerical models are essential to extrapolate these localized findings to global and seasonal scales and assess their broader implications. The vertical distribution of aerosols is particularly important, as their indirect radiative effect strongly depends on their profile relative to cloud-forming altitudes (Watson-Parris et al., 2019; Mishra et al., 2015; Zhu et al., 2024; Choi and Chung, 2014). However, to date, no detailed evaluation of key aerosol species in simultaneous simulations across all layers of the lowermost 20 to 30 km has been conducted.

We present the results of a global simulation of atmospheric aerosol distributions, composition, and number concentrations from the surface to the stratosphere using the ECHAM/MESSy Atmospheric Chemistry (EMAC) model, based on a newly developed model setup. EMAC has previously been applied to study BL and tropospheric aerosols (e.g., Pringle et al., 2010; Kohl et al., 2023; Pozzer et al., 2012; Lelieveld et al., 2015; Tsimpidi et al., 2024), mostly focusing on air pollution and associated health impacts, as well as for stratospheric aerosols (e.g., Schallack et al., 2023; Brühl et al., 2018; Kohl et al., 2024), with a focus on the formation of the Junge layer and the impacts of stratospheric volcanic eruptions. In this study, we combine and extend the tropospheric and stratospheric setups to represent aerosols across the entire lower atmosphere, from the surface to the stratosphere.

Accurate representation of aerosols in chemistry-climate models requires thorough evaluation against observations to ensure that aerosol properties and key processes are realistically captured. We comprehensively evaluate our simulations with a combination of ground-based observations, satellite remote sensing data, and data from a number of aircraft missions, especially focusing on the FT and UTLS, justifying its usage for the derivation of global aerosol distributions and composition.

Section 2 outlines the methods and experimental setup, including the observational datasets, the EMAC model, and the newly developed setup. The model evaluation is presented in Sect. 3, followed by the resulting global aerosol distributions and composition in Sect. 4. Section 5 discusses the findings and limitations of the simulations.

2 Methods & Model Setup

While the aerosol distributions presented in Sect. 4 are derived from numerical simulations, observational data are essential for a comprehensive model evaluation. Section 2.1 introduces the observational datasets used in this study, followed by a detailed description of the EMAC model and the newly developed setup in Sect. 2.2.

2.1 Observations

2.1.1 In-situ surface observations

The Globally Harmonised Observations in Space and Time (GHOST; Bowdalo et al., 2024) dataset compiles extensive in-situ surface observations of atmospheric composition from multiple networks, harmonized into a standardized format. We evaluate simulated surface aerosol mass concentrations using data from the following networks: EPA Air Quality (USA), EEA Air Quality and EMEP (Europe), Canada NAPS (Canada), UK Air (UK), Chile SINCA (Chile), MEXICO CDMX (Mexico), and MITECO (Spain). Further details on these networks and the data harmonization are provided by Bowdalo et al. (2024).

Although the Acid Deposition Monitoring Network in East Asia (EANET) is part of the GHOST project, its data is not included in the published GHOST dataset used in this study (Bowdalo, 2024) due to data rights limitations. Therefore, supplementary observations were obtained directly from EANET (2025).

2.1.2 Aircraft in-situ observations

We combine observational datasets from aircraft missions targeting both background conditions and specific regions or processes. Figure 1 provides a global overview of the flight paths from the aircraft campaigns used for this evaluation. Further details on the individual campaigns are provided below.

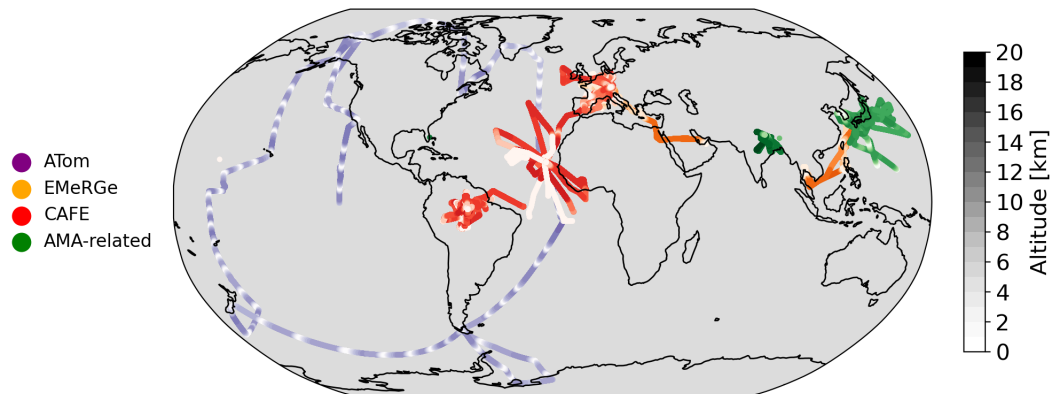


Figure 1. Flight paths of the aircraft missions used for the evaluation of the EMAC model. Asian Monsoon Anticyclone (AMA) related aircraft campaigns are StratoClim and ACCLIP. The altitude profiles are provided by the shading of the colour.

- 100 – **NASA ATom mission:** The NASA ATom mission (Thompson et al., 2022) sampled background atmospheric conditions over continental North America, the Pacific, Atlantic and Southern Ocean, in four two-months-campaigns (6 flights each) between July 2016 and May 2018. Numerous profiles between 200 m and 12 km altitude were recorded, providing the basis for the evaluation of the model’s capability to represent atmospheric background aerosol distributions in the FT and UTLS. Aerosol mass concentrations were measured with the High-Resolution Aerosol Mass Spectrometer (HR-AMS
 105 Jimenez et al., 2019; Canagaratna et al., 2007; DeCarlo et al., 2006), and the Single Particle Soot Photometer (NOAA SP2; Schwarz and Katich, 2019; Katich et al., 2018; Schwarz et al., 2010; Gao et al., 2007) for BC. Particle number concentrations and size distributions were taken from the Aerosol Microphysical Properties (AMP) instrument (Brock et al., 2019a, b; Williamson et al., 2019; Kupc et al., 2018). Gas phase mixing ratios are provided by the NOAA Nitrogen Oxides and Ozone (NO_yO₃) 4-channel chemiluminescence (CL) instrument (NO; Ryerson et al., 2019), the Picarro
 110 G2401 spectrometer (CO; McKain and Sweeney, 2021), the Airborne Tropospheric Hydrogen Oxides Sensor (ATHOS; OH; Brune et al., 2021), the California Institute of Technology Chemical Ionization Mass Spectrometer (CIT-CIMS; SO₂ & HNO₃; Allen et al., 2021), and the Unmanned Aircraft Systems (UAS) Chromatograph for Atmospheric Trace Species (UCATS; O₃; Elkins et al., 2020).
- **HALO campaigns (CAFE and EMeRGe)** The High Altitude and Long Range Research Aircraft (HALO) is regularly
 115 equipped for atmospheric studies. We use aerosol mass concentration measurements collected with the Compact Time-of-Flight Aerosol Mass Spectrometer (C-ToF-AMS; Drewnick et al., 2005; Schulz et al., 2018), covering the size range from 40-800 nm, onboard HALO during three CAFE (Chemistry of the Atmosphere: Field Experiments; altitudes from 0 to 15 km) and two EMeRGe (Effect of megacities on the transport and transformation of pollutants on the regional and global scale; altitudes from 0 to 14 km) missions. CAFE-Africa (2018; from Cape Verde) studied the influence
 120 of biomass burning from Africa on the tropical and subtropical Atlantic Ocean. CAFE-Brazil (Dec. 2022/ Jan. 2023; from Manaus) investigated the chemical processes in the remote atmosphere over the Amazon rainforest. CAFE-EU (May/June 2020; from Oberpfaffenhofen; Voigt et al., 2022) studied the impact of the emission reduction due to the COVID-related lockdown on the atmosphere over Europe. The EMeRGe missions aimed for polluted regions above megacities, and focused on lower altitudes in Europe (EMeRGe-EU; May to August 2017) and Asia (EMeRGe-Asia;
 125 February to April 2018).
- **Additional aircraft campaigns:** The Asian Monsoon Anticyclone (AMA) has been identified as a key source of UTLS aerosol, by transporting aerosol precursors and primary material in convective systems, building the ATAL (e.g., Vernier et al., 2011; Appel et al., 2022). We include data from two aircraft missions in our evaluation, studying this phenomenon in recent years. The StratoClim mission (July/August 2017; from Kathmandu, Nepal), directly probed ATAL air and the
 130 convective outflow, using the high-altitude research aircraft M-55 Geophysica, reaching up to 20 km, mostly sampling UTLS air. The ACCLIP mission (Sept. 2022; from South Korea) combined two research aircrafts, NASA WB-57 and the NCAR G-V, and studied transport pathways of ATAL aerosol over the Western Pacific. Aerosol mass concentration measurements have been performed with the ERC Instrument for the Chemical composition of Aerosols (ERICA-AMS;

Hünig et al., 2022; Dragoneas et al., 2022) in both missions. Additionally, BC measurements during the ACCLIP mission are taken from the NOAA SP2 instrument (see NASA ATom mission).

2.1.3 Remote sensing

As the available aircraft-based observations only extend into the lowermost stratosphere, we complement the evaluation of simulated stratospheric aerosol with the Global Space-based Stratospheric Aerosol Climatology (GloSSAC V2.21; Kovilakam et al., 2023, 2020; Thomason et al., 2018). GloSSAC merges remote sensing data from multiple satellite instruments and provides monthly mean aerosol extinction coefficients in the stratosphere, interpolated to four wavelengths (386, 452, 525, and 1020 nm). GloSSAC observations from 2016–2018, used for the evaluation, rely on the following satellite instruments:

- **The Optical Spectrograph and InfraRed Imaging System (OSIRIS)**, onboard the ODIN satellite, uses limb scattered sunlight for the retrieval of aerosol extinction at 750 nm (Bourassa et al., 2012). Here, the aerosol retrieval version 7.0 (Rieger et al., 2019) is applied. In GloSSAC, the aerosol extinction is converted to 525 nm using the Angstrom coefficient and bias-corrected (Kovilakam et al., 2020).
- **The Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP)** instrument, onboard the CALIPSO satellite observes back-scattered laser signals at near-nadir geometry, providing aerosol extinction at 532 and 1064 nm (Winker et al., 2009), extrapolated to 525 nm and 1020 nm for GloSSAC, respectively.
- **The Stratospheric Aerosol and Gas Experiment III (SAGE-III)**, externally mounted on the ISS is a solar occultation instrument, retrieving aerosol extinction at 9 different wavelengths, extrapolated to all 4 GloSSAC wavelength (Kovilakam et al., 2023).

Additionally, simulated column aerosol optical depth (AOD) at 550 nm is evaluated against observations from the MODerate Resolution Imaging Spectroradiometer (MODIS), onboard the Terra satellite. We use the MODIS Level 3 product (Platnick, 2015), applying the Deep Blue retrieval algorithm (Hsu et al., 2004).

2.2 Model setup

Model simulations are performed using the ECHAM/MESSy Atmospheric Chemistry (EMAC) model (Jöckel et al., 2006), which combines the general circulation model ECHAM (version 5.3.02, Roeckner et al., 2003) with submodels from the Modular Earth Submodel System (MESSy version v2.55.1, Jöckel et al., 2010). The model is operated at a spectral resolution of T63 ($1.875^\circ \times 1.875^\circ$) with 90 vertical levels extending up to 0.01 hPa (approximately 80 km), including additional layers in the middle atmosphere, with 7.5 minute timestep length. Temperature, surface pressure (logarithm), divergence and vorticity are “nudged” by Newtonian relaxation (more details by Jöckel et al., 2006; Jeuken et al., 1996) towards meteorological reanalysis data (ERA5, Hersbach et al., 2020) from the European Centre for Medium-Range weather forecasts (ECMWF) below 100 hPa, with prescribed transient sea surface temperatures and sea ice coverage. Zonal winds are weakly “nudged” to observations (Giorgetta et al., 2006) between 10 and 90 hPa to avoid a phase drift of the quasi-biennial oscillation (QBO).

The main simulations cover the years 2016 to 2018, following a half-year spin-up period. In addition, shorter simulations were conducted for the periods July 2019 to June 2020 (for comparison with the CAFE-EU campaign) and January 2022 to January 2023 (for comparison with the ACCLIP and CAFE-Brazil campaigns). Tracer initialization is based on ten-year simulations to ensure the establishment of a stratospheric equilibrium.

Aerosol microphysics are handled by the MESSy submodel GMXe (Pringle et al., 2010), which uses hydrophilic and hydrophobic lognormal modes to describe the particle size distribution (PSD). Detailed information about these modes in the context of this study, and the respective microphysics is provided in Sect. 2.2.1. In GMXe, inorganic aerosol thermodynamics are computed using ISORROPIA-II (Fountoukis and Nenes, 2007), whereas the phase partitioning of organic aerosols (OA) is processed by the ORACLE submodel (Tsimpidi et al., 2014, 2017), based on source and volatility. While organic and inorganic species are treated separately thermodynamically, they are internally mixed within each aerosol size mode. Aerosols can transfer between modes due to changes in size or solubility after each timestep. Sulfate evaporation in the UTLS at low humidity and in the absence of ammonia is computed based on molality following Tabazadeh et al. (1997), relying on the tabulation of Giauque et al. (1960).

The EMAC simulation results presented here capture key aerosol species across the lower atmosphere. Speciated aerosols include sulfur aerosol (SO_4^{2-} ; hereafter referred to as sulfate), ammonium (NH_4^+), nitrate (NO_3^-), sodium (Na^+), chloride (Cl^-), potassium (K^+), calcium (Ca^{2+}), and magnesium (Mg^{2+}). OA is categorized based on source, volatility, and primary (POA) or secondary (SOA) origin, amounting to 36 different organic species (more details by Tsimpidi et al., 2014, 2017). In addition, the model includes bulk aerosol species: black carbon (BC), desert dust (DU), and sea salt (SS). Desert dust emissions are primarily represented by the DU bulk species, which accounts for $\sim 90\%$ of total dust mass, with smaller contributions from speciated cationic components (Klingmüller et al., 2018). Sea salt aerosol is predominantly emitted as sodium chloride (86%), with additional emissions of sulfate (8%), bulk SS (0.8%), and other minor salt constituents.

Stratospheric heterogeneous reactions and the formation of Polar Stratospheric Clouds (PSCs) are simulated by the MESSy submodel MSBM (Jöckel et al., 2010), using a climatology for background liquid sulfur aerosol in the stratosphere (not considering volcanic eruptions) as a basis for heterogeneous reactions here. Neutral and ion-induced particle formation is simulated with the NAN submodel (Ehrhart et al., 2018) with ion concentrations provided by the IONS submodel (Ehrhart et al., 2018). We use new particle formation (NPF) parameterisations from the CLOUD experiment, including binary ($\text{H}_2\text{SO}_4\text{-H}_2\text{O}$) and ternary ($\text{H}_2\text{SO}_4\text{-NH}_3\text{-H}_2\text{O}$) nucleation (Dunne et al., 2016), enhanced with HNO_3 ($\text{H}_2\text{SO}_4\text{-NH}_3\text{-HNO}_3$; Wang et al., 2022), and pure organic nucleation (Kirkby et al., 2016) of highly oxygenated organic molecules (HOMs).

Global anthropogenic emissions of reactive gases and aerosols at the surface are sourced from the Community Emission Data System (CEDS; McDuffie et al., 2020), while aircraft emissions are taken from CAMS Global Aviation Emissions (CAMS-GLOB-AIR; Granier et al., 2019) up to 2019. For subsequent years, emissions are repeated due to the unavailability of updated datasets. Biomass burning and agricultural waste burning emissions are calculated based on observed dry matter burned and fire type, following Andreae (2019), using the BIOBURN submodel. Natural emissions of biogenic VOCs are computed using the MEGAN model (Guenther et al., 2012) implemented as MESSy submodel. Sea salt emissions (Guelle et al., 2001) and dust emissions (Klingmüller et al., 2018) are computed online with the ONEMIS submodel (Kerkweg et al., 2006), and are emitted

200 as a combination of bulk dust and sea salt, along with specific compounds. Size distributions for primary particle emissions are applied per sector based on available literature as described in Kohl et al. (2023).

Emissions from volcanic eruptions that reach the stratosphere are handled by the EVER submodel (Kohl et al., 2024), including volcanic eruptions from 1990 to 2023 based on the emission inventory from Schallrock et al. (2023). Degassing tropospheric volcanoes are applied based on a volcanic climatology from AEROCOM (Dentener et al., 2006).

205 Gas-phase chemistry is simulated using the MECCA submodel (Sander et al., 2019), utilizing the Mainz Isoprene Mechanism (MIM1; Pöschl et al., 2000; Jöckel et al., 2006), which includes over 100 gas-phase species and more than 250 chemical reactions. Removal of gas and aerosol species via sedimentation, dry deposition, and wet deposition is handled by the SEDI, DDEP (both Kerkweg et al., 2006), and SCAV submodels (Tost et al., 2006), respectively.

Aerosol optical properties are diagnosed using the newly developed MESSy submodel AOP (Tost et al., manuscript in preparation), which is comparable in concepts to the AEROPT scheme, described by Dietmüller et al. (2016). AOP computes mode-averaged refractive indices based on the chemical composition, and derives optical properties like extinction, single scattering albedo and asymmetry factor, using pre-calculated Mie parameters. The total optical effects per model grid cell are then obtained by weighting the contributions of the individual modes according to their number distributions and interpolating or calculating their optical properties at the target wavelengths.

215 Noteworthy, we did not implement any aerosol feedbacks here, as this study focuses on the establishment of aerosol distributions from the surface to the stratosphere without considering any direct implications for climate or the ozone layer. Heterogeneous reactions are calculated based on a stratospheric aerosol climatology (Luo, 2013), radiation calculations are based on a merged aerosol climatology for the troposphere (Tanré et al., 1984) and the stratosphere (Luo, 2013), while online aerosol optical properties based on simulated aerosol distributions are used diagnostically for comparison with observations (see above), and cloud properties are decoupled from the diagnostic cloud condensation nuclei (CCN) number simulated by GMXe.

2.2.1 Aerosol size distribution

An important aspect in the development of the new setup is the representation of aerosol size distributions. Due to differences in aerosol lifetime, ambient pressure, and pollution levels across the BL, FT, UTLS, and the stratospheric overworld, size distributions vary significantly. GMXe provides an adjustable modal size distribution, allowing the aerosol median diameter to evolve freely within user-defined mode boundaries, with particles transferred to neighboring modes when they grow too large or too small. Within each mode, particles are internally mixed, while the modes themselves remain externally mixed with respect to each other. The boundaries and standard deviation of the logarithmic modes are constant globally, resulting in varying microphysical behavior across different regions of the lower atmosphere.

230 Past studies on tropospheric aerosol (e.g., Kohl et al., 2023; Pozzer et al., 2012; Lelieveld et al., 2015; Tsimpidi et al., 2024) typically considered 4 soluble log-normal modes (see Table 1; here referred to as **TROPO**): a nucleation mode for freshly formed particles up to 12 nm, the Aitken mode for fine anthropogenic primary emissions and grown nucleation mode particles (12-120 nm), the accumulation mode for further grown particles and larger primary particles (120-1400 nm), and a coarse

Table 1. Modal size distribution in the new setup for the entire lower atmosphere (LA; used in this study) in contrast to size distributions used in former studies, which focussed only on the troposphere (TROPO) or stratosphere (STRATO). The "small coarse" mode was added between the accumulation and the coarse mode in the LA setup. The median diameter can adjust within the given diameter boundaries. Additionally, the globally constant sigma of the log-normal distribution is given for each mode.

Setup		Nucleation	Aitken	Accumulation	Small Coarse	Coarse
TROPO	Diameter [nm]	1 – 12	12 – 120	120 – 1400	–	> 1400
	Sigma	1.59	1.59	1.59	–	2.0
STRATO	Diameter [nm]	1 – 12	12 – 140	140 – 3200	–	> 3200
	Sigma	1.59	1.59	1.49	–	1.7
LA	Diameter [nm]	1 – 12	12 – 120	120 – 800	800 – 3200	> 3200
	Sigma	1.59	1.59	1.4	1.25	1.7

mode for particles larger than 1400 nm, primarily consisting of dust and sea salt particles. Additionally, three insoluble log-normal modes are used (Aitken, accumulation and coarse) with the same mode boundaries as the respective soluble modes. Insoluble particles can transfer to the soluble mode through coagulation with soluble particles or coating with soluble species (more details in Pringle et al., 2010).

Stratospheric studies (e.g., Schallcock et al., 2023; Brühl et al., 2018; Kohl et al., 2024) used the same number of modes, but with modified mode boundaries and widths (Table 1; here referred to as **STRATO**). While the nucleation mode remains identically defined, the Aitken mode is extended to slightly larger diameters (140 nm). The accumulation mode is expanded to include particles up to 3200 nm, with a narrower modal width (sigma = 1.49). This adjustment suppresses the rapid transfer of particles to the coarse mode, thus reducing sedimentation and prolonging aerosol lifetimes in the stratosphere. However, for the same reason this setup fails to reproduce BL aerosol distributions, where the inhibited transfer to the coarse mode leads to overestimated particle lifetimes. More details on the performance of the setups are provided in the evaluation (Sect. 3) and discussed in more detail in Sect. 5.

In this study, we present a new modal configuration for GMXe, combining elements of the TROPO and STRATO setups to represent the entire **Lower Atmosphere** (Table 1; here referred to as **LA**). This setup introduces an additional mode, termed the *small coarse mode*, positioned between the accumulation and the coarse modes, and spanning the size range from 800 to 3200 nm. This mode serves three main purposes. First, it prevents particles in the stratosphere from growing too large and sedimenting in the coarse mode, and thus reproducing stratospheric aerosol distributions similar to the STRATO setup. Second, in the BL, accumulation mode particles grow into the small coarse mode, reducing BL aerosol lifetimes compared to the STRATO setup. Third, it better represents sea salt and dust emissions, which typically peak between 800 and 1000 nm and are now emitted directly into the small coarse mode. In the STRATO and TROPO setups, these particles were instead emitted into the accumulation mode, internally mixing with particles freshly grown from the Aitken mode with median diameters

255 around 300 nm, thus underestimating the actual emission size. The three setups are comprehensively evaluated, compared and discussed in Sect. 3 and Sect. 5, further motivating the development and choice of the LA setup.

2.3 Spatial and temporal sampling

To ensure comparability between model data and observations, all datasets are homogenized through consistent spatial and temporal sampling. The specific sampling approach depends on the type of comparison.

260 For in-situ ground-based observations, annual averages are calculated and compared to the corresponding annual mean of the lowermost model grid box that contains the measurement site. In the case of AOD, MODIS observations are first regridded to the model resolution before comparing annual averages to simulated values.

Aircraft observations are typically available at sub-minute resolution, whereas the model output is provided every 7.5 minutes. During a single model timestep, the aircraft may cover a considerable vertical range. Therefore, the complete vertical
265 column of model output along the flight tracks is sampled at each timestep using the S4D submodel (Jöckel et al., 2010). Both modelled and observed data are subsequently binned by altitude and timestep, and the resulting averages are compared.

For comparisons with GloSSAC satellite observations, simulated daily averaged aerosol extinction is regridded to match the horizontal and vertical resolution of the satellite data prior to evaluation.

3 Evaluation

270 Model simulations are evaluated using in-situ ground and aircraft observations, as well as remote sensing data from satellite instruments. The evaluation primarily focuses on the new LA setup, but comparisons to simulations using the modal size distributions from the TROPO and STRATO setups are also presented.

For statistical comparison with observations, the following statistical metrics are applied:

- $\overline{M/O}$: Geometric mean of the ratio between model simulated (M) and observed (O) means
- 275 – PF2: Percentage of simulated values within a factor of two of the observed values
- Corr: Pearson correlation coefficient between observations and simulation results
- RMSE: Root Mean Squared Error

3.1 Comparison to in-situ ground observations

Figure 2 compares total observed and simulated (LA setup) $PM_{2.5}$ mass concentrations, along with the most dominant components (excluding desert dust due to missing observations). Table 2 provides a statistical summary of the comparison, also
280 including results from simulations with the STRATO and TROPO setups. Total $PM_{2.5}$ mass concentrations are slightly underestimated in the LA simulation ($\overline{M/O} = 0.92$); however, 91% of simulated values fall within a factor of two of the observations.

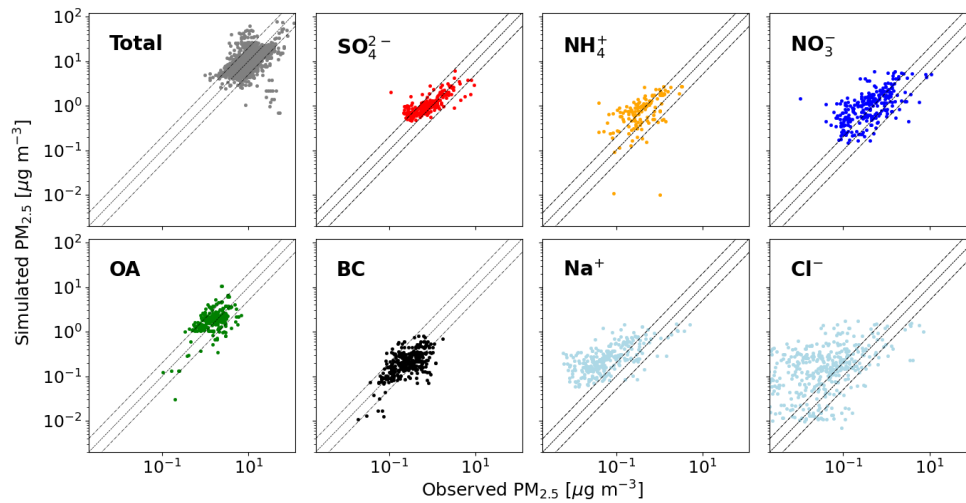


Figure 2. Comparison of annually averaged (2017) $\text{PM}_{2.5}$ mass concentrations at the Earth’s surface from EMAC simulations (LA setup) with stationary observations from the GHOST (Bowdalo, 2024) and EANET (2025) databases, showing total $\text{PM}_{2.5}$ and contributions from key aerosol species. The central dotted line indicates 1:1 agreement between observations and simulations, while the surrounding dashed lines represent overestimation and underestimation by a factor of two.

The TROPO setup performs similarly, whereas the STRATO setup simulates higher $\text{PM}_{2.5}$ values, and overestimates observations ($\overline{\text{M}}/\overline{\text{O}} = 1.28$), though with comparable correlation and RMSE.

285 The LA and TROPO simulations show comparable performance in predicting surface mass concentrations of key aerosol species (see Table 2), confirming the suitability of the LA setup for surface-level studies. In both setups, NO_3^- is overestimated by about a factor of 3 (lower than in previous EMAC studies (Pozzer et al., 2022)), but is well correlated with observations. Simulated SO_4^{2-} and OA match observations closely, with a slight high bias at low concentrations. The STRATO setup tends to overestimate these species (Table 2), mainly due to reduced transfer of accumulation mode particles to the coarse mode, leading to longer particle lifetimes and making it less suitable for BL applications.

290 Speciated dust and sea-salt compounds show poor surface-level agreement with stationary observations, as reported in previous studies (Pozzer et al., 2022). Both LA and TROPO simulations overestimate sodium (by a factor of 4 in the LA setup), chloride (factor 5), and magnesium (factor 1.52), while underestimating potassium and calcium (both by a factor of 4-5). The STRATO setup generally produces larger values for those 5 species leading to a strong overestimation of sodium and chloride by a factor of 18 and 30, respectively. A detailed investigation of the composition, abundance, and size distribution of dust and sea-salt aerosol emissions is needed.

Table 2. Summary of annually averaged (2017) observed (Obs) surface PM_{2.5} concentrations and composition (Obs), based on data from the GHOST (Bowdalo, 2024) and EANET (2025) databases. Observations (in $\mu\text{g m}^{-3}$) are compared to EMAC simulations using the LA, TROPO, and STRATO setups, evaluated with the statistical metrics defined earlier. The number of available observational stations is indicated by N_{obs}.

Species	Observations		LA				TROPO				STRATO			
	N _{obs}	Obs	M/O	PF2	Corr	RMSE	M/O	PF2	Corr	RMSE	M/O	PF2	Corr	RMSE
NH ₄ ⁺	100	0.44	2.07	38	0.64	0.55	2.09	39	0.62	0.53	2.52	29	0.64	0.71
SO ₄ ²⁻	238	0.8	1.39	83	0.73	0.38	1.16	86	0.75	0.31	1.89	60	0.67	0.65
NO ₃ ⁻	237	0.57	2.45	34	0.67	1.07	3.04	28	0.66	1.17	4.99	8	0.69	1.86
OA	307	1.79	1.16	77	0.59	1.25	1.05	81	0.59	1.2	1.23	76	0.57	1.27
BC	307	0.34	0.76	70	0.6	0.24	0.67	66	0.61	0.25	0.76	72	0.6	0.24
Cl ⁻	393	0.07	5.29	22	0.3	0.28	10.19	13	0.29	0.46	29.81	3	0.27	1.18
K ⁺	310	0.05	0.19	7	0.12	0.05	0.24	12	0.04	0.05	0.71	62	0.06	0.04
Na ⁺	252	0.08	4.85	13	0.62	0.27	6.29	8	0.55	0.33	18.13	0	0.48	0.98
Mg ²⁺	169	0.02	1.7	48	0.22	0.03	2.25	43	0.17	0.04	6.42	11	0.1	0.11
Ca ²⁺	233	0.05	0.24	21	-0.19	0.06	0.31	24	-0.3	0.06	0.89	49	-0.27	0.05
PM _{2.5}	3461	11.63	0.92	91	0.53	6.25	0.85	91	0.54	6.08	1.28	87	0.53	6.04

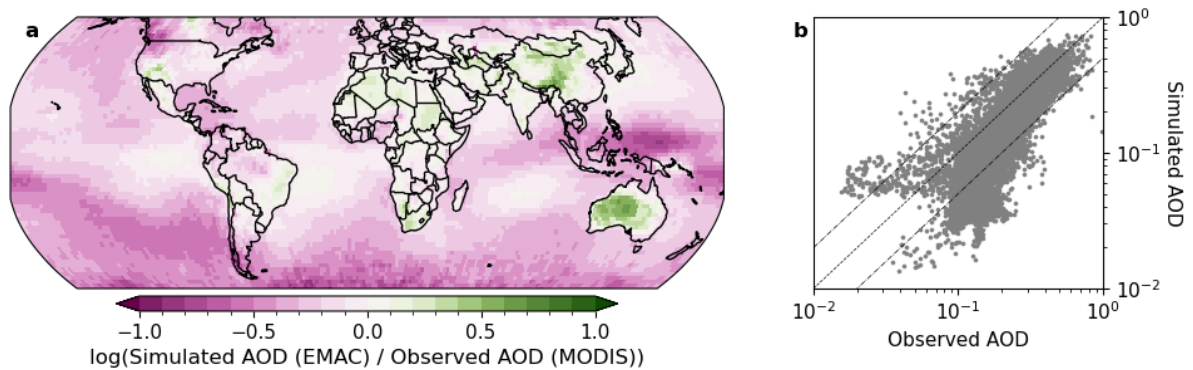


Figure 3. Comparison of annually averaged (2017) simulated column Aerosol Optical Depth (AOD) from EMAC (LA setup) with MODIS observations at 550 nm. (a) Global map of the logarithmic ratio of simulated to observed AOD; green indicates overestimation by the model, pink indicates underestimation. (b) Scatter plot comparing simulated and observed AOD globally. Each dot represents one model grid box. The central dotted line indicates 1:1 agreement between observations and simulations, while the surrounding dashed lines represent overestimation and underestimation by a factor of two.

3.2 Aerosol Optical Depth (AOD)

Figure 3 compares the annually averaged simulated column AOD (LA setup) with MODIS observations at 550 nm for the year 2017. AOD over continental regions is generally well represented, with a slight underestimation in North America and Southeast Asia, and an overestimation over Australia and remote parts of China. Marine AOD is underestimated over unpolluted oceans, but the agreement improves in coastal and biogenic- or dust-influenced regions of the tropical Pacific and Atlantic. The underestimation in remote marine regions may be attributed to excessive particle growth from the accumulation mode into the small coarse mode in the LA setup, or a misrepresentation of the sea salt emission size distribution.

Simulated column AOD varies markedly across the three setups (see also Supplementary Fig. S7). TROPO shows the best overall agreement, except for a marked underestimation over Southeast Asia. STRATO strongly overestimates the column AOD, particularly at large observed AOD values, likely due to the increased aerosol lifetime, resulting from less efficient growth from the accumulation to the coarse mode (see above). Notably, there is a coincidence of the mode transition in the LA setup at 550 nm, making AOD underestimation more likely. In comparison, the minima in the other size distributions have less influence at 550 nm.

3.3 Vertical profiles

Vertical profiles of atmospheric aerosol composition for the evaluation of the EMAC simulation results are derived from multiple aircraft campaigns. We primarily focus on the period from January 2016 to December 2018, with additional simulations used for comparisons to the CAFE-EU (2020), ACCLIP (2022), and CAFE-Brazil (2022/2023) missions.

3.3.1 Atmospheric background conditions – ATom campaign

Figure 4 compares simulated mass concentrations of sulfate, ammonium, nitrate, total OA (all PM_{10}), BC (90–550 nm), and dust (100 nm–4.8 μm), as well as particle number concentrations (2.7 nm–4.8 μm), to corresponding ATom observations. Observations were divided into 10 geographic regions, detailed in Appendix Fig. C1, five of which are shown here. The remaining regions are presented in Appendix Fig. C2. Table 3 shows a statistical comparison of the three model setups to observations, using observed and simulated median values at each region, season and altitude. Additional evaluations of key gas-phase species are shown in Supplementary Figs. S5 and S6. Appendix Fig. C3 presents PSDs across the observed layers in the different regions.

Gas-phase species generally agree well with observations, especially CO, OH, O_3 , and NO, with minor discrepancies in O_3 at higher altitudes. SO_2 mixing ratios are slightly underestimated in the East Pacific but overestimated in the northern regions and the North Pacific. The underestimation may be due to local volcanic degassing events, which are included in the model based on climatological data, which, however, do not represent irregular and unpredictable emissions.

Overall, the model captures the order of magnitude of aerosol observations, though the level of agreement varies by region and season. Low $\overline{\text{M/O}}$ values partly reflect detection limits of the observations. The largest discrepancies occur over the Southern Ocean and in the northern FT (Fig. C2), where aerosol mass is underestimated across all species. This underestimation

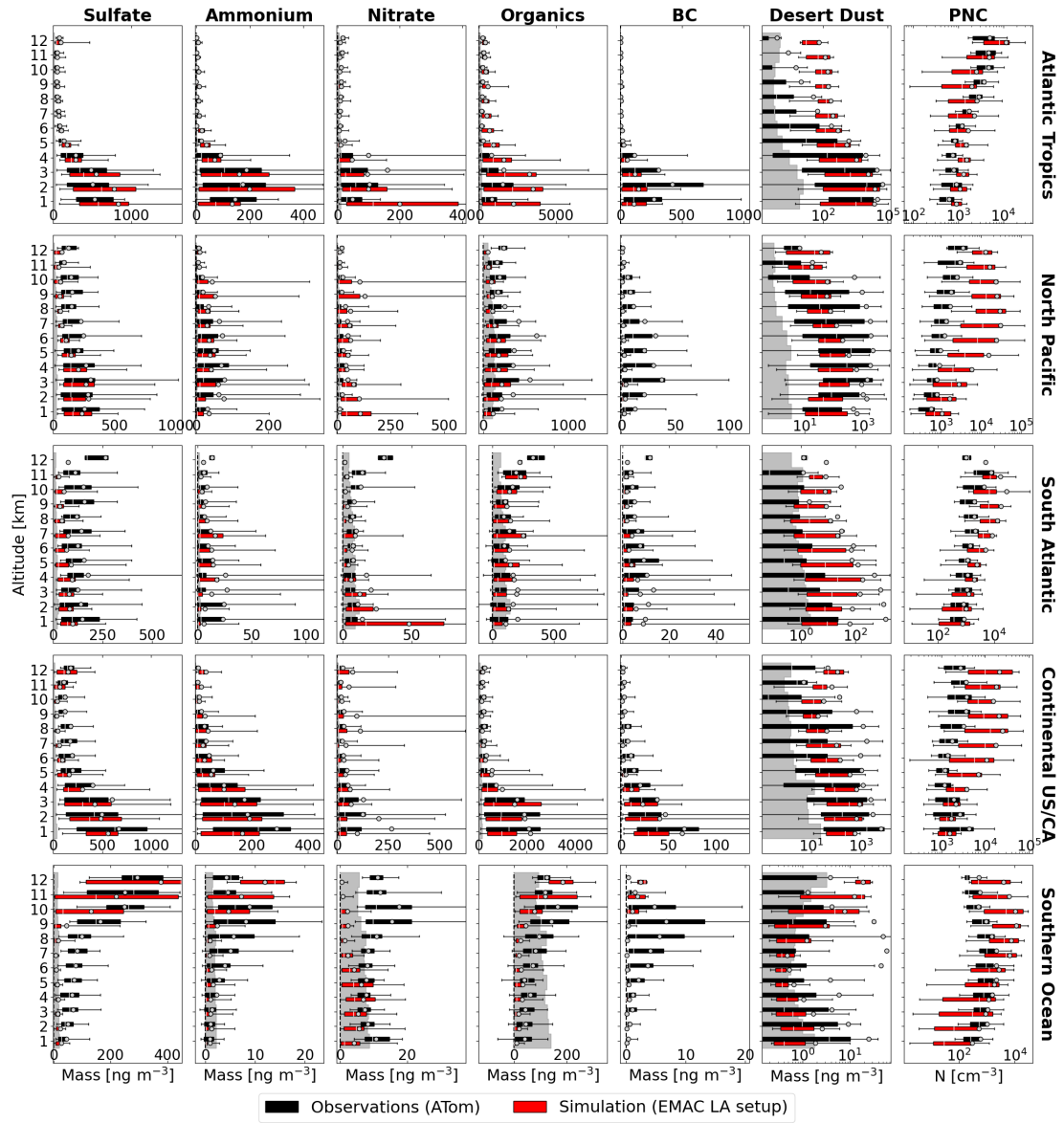


Figure 4. Comparison of simulated (red) and observed (black) mass concentrations of sulfate, ammonium, nitrate, organic aerosol (all PM_{10}), black carbon (BC; 90–550 nm), dust (0.1–4.8 μm), and particle number concentrations (PNC; 2.7 nm–4.8 μm) from the ATom mission across selected regions, as defined in Appendix Fig. C1. Grey points indicate mean values, white vertical lines represent medians, boxes show the interquartile range, and whiskers denote the 5th and 95th percentiles. The grey shaded area indicates the detection limit of the instrument. All concentrations are provided at standard temperature and pressure (STP; $T = 273.15 \text{ K}$ & $p = 1013.25 \text{ hPa}$).

Table 3. Summary of observed (Obs) mass concentrations of key aerosol species and number concentrations during the ATom campaign. Observations (in ng m^{-3} for mass concentrations, and cm^{-3} for particle number concentration) are compared to EMAC simulation results (Mod) using the LA, TROPO, and STRATO setups, evaluated with the statistical metrics defined earlier. For statistical comparison, the median of observed and simulated values at each region, season and altitude (1 km-binned; see Fig. 4) is used.

	ATom	LA					TROPO					STRATO				
Spec.	Obs	Mod	$\overline{\text{M/O}}$	PF2	Corr	RMSE	Mod	$\overline{\text{M/O}}$	PF2	Corr	RMSE	Mod	$\overline{\text{M/O}}$	PF2	Corr	RMSE
SO_4^{2-}	152	117	0.49	50.7	0.61	148.4	121	0.55	49.7	0.67	130.6	225	0.88	48.6	0.64	298.4
NH_4^+	19	20	0.68	30.8	0.65	37.3	20	0.69	33.1	0.73	32.6	25	0.76	32.5	0.66	43.1
NO_3^-	14	28	0.48	22.9	0.42	59.6	54	0.69	20.0	0.4	130.0	110	1.8	16.1	0.38	255.2
OA	177	217	0.52	41.0	0.62	399.3	230	0.53	38.9	0.62	417.4	312	0.71	39.9	0.62	676.7
BC	13	5	0.52	27.6	0.77	58.4	4	0.49	31.0	0.62	64.0	6	0.75	31.2	0.74	59.0
DU	833	931	1.69	15.5	0.94	2874.5	481	1.49	15.1	0.94	5477.3	630	2.46	15.3	0.93	4750.9
PNC	1573	5523	2.13	43.5	0.24	10041.5	5193	1.93	44.8	0.22	9531.4	3580	1.35	52.0	0.2	6926.0

is most likely due to a combination of underestimated transport from the stratosphere to the troposphere and insufficient long-range transport of biomass burning aerosols to remote regions. Schill et al. (2020) showed that biomass burning pollution is widespread throughout the FT, with enhanced levels in the regions and seasons where the model most strongly underestimates FT aerosol mass.

Simulated sulfate aerosol concentrations in the lower 1 to 3 km generally agree well in both continental and marine regions, while the STRATO setup overestimates BL values. In the FT, the LA setup tends to underestimate sulfate in background regions, with better agreement near the UTLS. Statistically, all setups show similar correlations ($r > 0.6$) and PF2 ($\sim 50\%$), but STRATO has twice the RMSE, mostly due to the overestimation at low altitudes.

Ammonium concentrations correlate well with observations throughout the troposphere, though PF2 values remain low due to high variability. Over the Southern Ocean and high northern latitudes (Fig. C2), concentrations rise above 9 km, indicating an overestimation in the stratosphere. Ammonium levels in the lowermost stratosphere range from 10–20 ng m^{-3} in the Southern Hemisphere to $\sim 50 \text{ ng m}^{-3}$ in the Northern Hemisphere. As shown in Sect. 3.1, surface nitrate is overestimated by a factor of ~ 3 , increasing to ~ 5 over the Atlantic Ocean. This bias is stronger in the TROPO and STRATO setups (resulting in a higher RMSE), particularly in dust-dominated regions (see Supplementary Figs. S1 and S2).

OA and BC are well represented in most regions of the background atmosphere, except for the Southern Ocean and northern latitudes, where missing biomass burning transport (as mentioned above) leads to discrepancies, and simulated mass concentrations of BC increase towards the stratosphere (not observed), suggesting an overestimation of stratospheric values. This overestimation is also evident in stratospheric dust concentrations. This likely results from overly efficient transport or insufficient scavenging of at least partially hydrophobic aged primary particles. Notably, the stratospheric overestimation over the

Southern Ocean coincides with an overestimation of O_3 above 10 km (Supplementary Fig. S5), possibly due to the aircraft sampling of more tropospheric air than simulated.

350 Near the Earth's surface, the model captures the strong contrast of high BC and OA mass concentrations from the Atlantic Ocean (influenced from Biomass burning in Central Africa) and low values over the Pacific, as observed by Katich et al. (2018). However, at higher altitudes over the Pacific, concentrations are underestimated, likely due to missing transport pathways. Dust concentrations generally match observations near the surface, and horizontal dust transport is well represented. The improved dust emission (emitting into the small coarse mode instead of the accumulation mode) scheme in the LA setup reduces RMSE
365 compared to the TROPO and STRATO setups.

PNC exhibit varying patterns across different regions. Near the Earth's surface, observed and simulated PNC agree well, with values ranging from 100 to 1000 particles per cm^3 over the ocean, and slightly higher PNC over background continental regions. However, surface PNC are underestimated over the Southern Ocean. The current model setup does not include recently proposed NPF from MSA and iodic acid (Baccarini et al., 2021; He et al., 2021; Zhang et al., 2025), which may contribute
360 to this underestimation. In most regions, UT NPF is overestimated, likely due to overestimated nucleation rates from ternary or synergistic nucleation, which dominate at these altitudes, or from the underestimated condensational sink (sulfate concentrations). However, over the Atlantic Tropics and the East Pacific, PNC are slightly underestimated. These regions are closest to the Amazon rainforest and may be influenced by NPF from isoprene oxidation products, as recently discovered by Curtius et al. (2024) and Shen et al. (2024a), which is not yet incorporated into the EMAC model, but will be addressed in follow-up
365 studies. In general, the LA setup shows worse agreement with observations for PNC compared to the TROPO and STRATO setups.

The AMP instrument also measured PSDs ranging from 2.7 nm to 4.8 μm . Appendix Fig. C3 compares observed PSDs with those simulated using the three different model setups. Due to their modal structure, the simulated PSDs exhibit local minima between modes, which introduces deviations from the observed distributions. In most regions, the LA setup reproduces
370 the observed PSDs well. Near the surface, nucleation mode particles are generally overestimated, except over the South Atlantic. Over the Southern Ocean, the Aitken and accumulation modes are underestimated, likely a result of missing long-range transport of biomass burning aerosols as described above. Coarse mode particles appear underestimated in most FT regions; however, observational data in this size range are close to the detection limit.

3.3.2 HALO campaigns

375 While the ATom mission focused on atmospheric background conditions, the HALO campaigns primarily aimed at investigating specific processes or regions. Figure 5 summarizes the comparison of the LA simulation with CAFE and EMERGE observations, while Table 4 statistically compares the simulations with the three setups to the observations. The LA setup exhibits lowest RMSEs compared to observations across all species, with similar or higher correlations and PF2 compared to the TROPO and STRATO setups, respectively, with the exception of ammonium. This underlines the capacity of the LA setup to
380 simulate the entire profiles encompassing the BL, FT and UTLS.

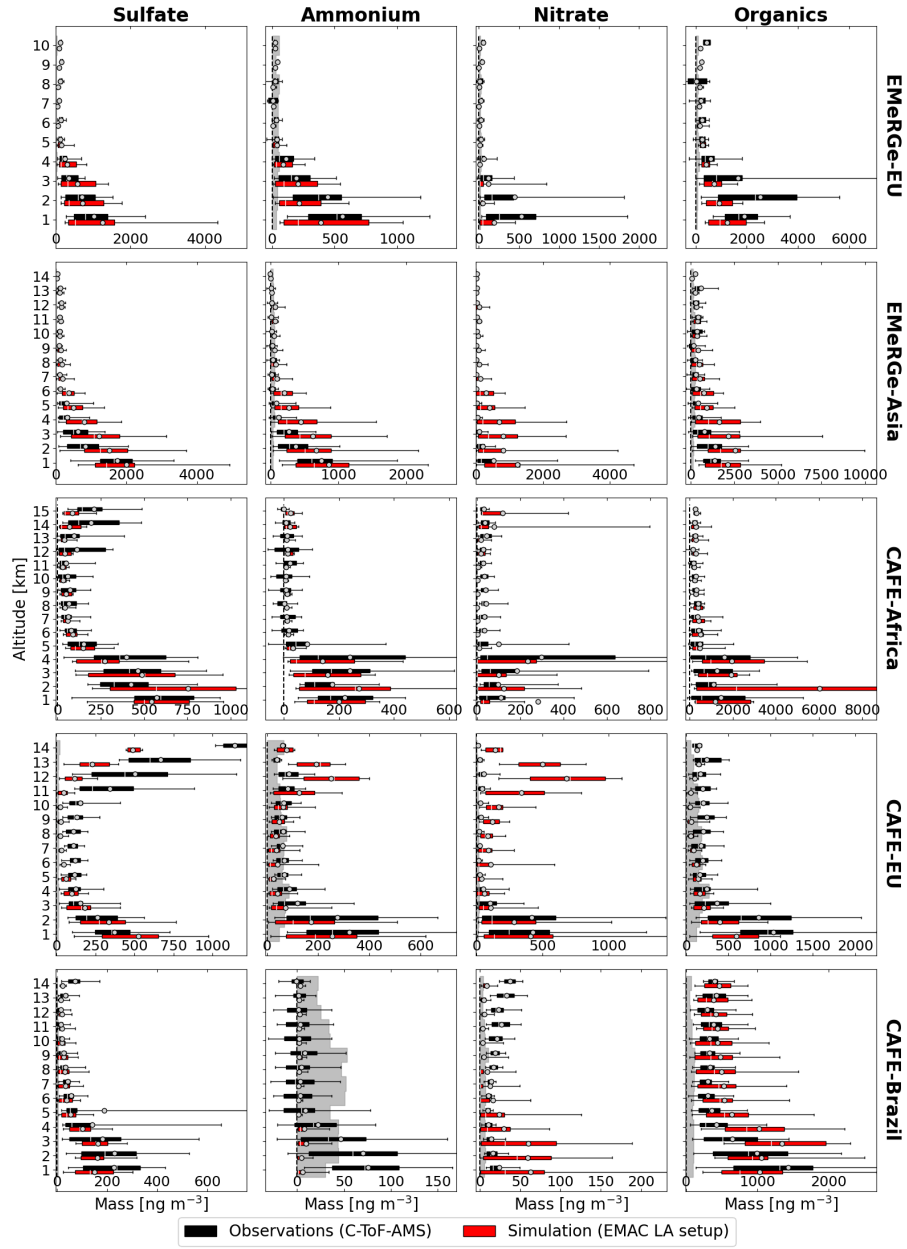


Figure 5. Comparison of simulated (red) and observed (black) mass concentrations of sulfate, ammonium, nitrate, and organic aerosol (all PM_{10}) from C-ToF-AMS measurements during recent HALO aircraft campaigns. Box-whisker plots follow the format of Fig. 4. All concentrations are provided at standard temperature and pressure (STP; $T = 273.15 \text{ K}$ & $p = 1013.25 \text{ hPa}$).

Table 4. Summary of observed (Obs) mass concentrations of key aerosol species from various HALO campaigns. Observations (in ng m^{-3}) are compared to EMAC simulation results (Mod) using the LA, TROPO, and STRATO setups, evaluated with the statistical metrics defined earlier. For statistical comparison, the median of observed and simulated values of each campaign and altitude (1 km-binned; see Fig. 4) is used.

Spec.	Obs	LA					TROPO					STRATO				
		Mod	$\overline{\text{M/O}}$	PF2	Corr	RMSE	Mod	$\overline{\text{M/O}}$	PF2	Corr	RMSE	Mod	$\overline{\text{M/O}}$	PF2	Corr	RMSE
SO_4^{2-}	207	188	0.74	67.2	0.77	191.6	500	1.62	41.8	0.77	729.1	295	0.99	64.2	0.73	367.2
NH_4^+	69	72	0.76	38.8	0.74	84.9	139	1.27	47.8	0.72	203.8	109	1.08	52.2	0.73	150.6
NO_3^-	37	73	0.44	23.9	0.21	143.0	302	2.39	22.4	0.52	654.6	201	1.18	16.4	0.42	456.5
OA	411	444	0.95	74.6	0.6	360.6	776	1.45	58.2	0.57	801.0	572	1.09	73.1	0.57	523.6

EMeRGe-EU and EMeRGe-Asia focused particularly on polluted megacities and their outflow at lower altitudes, resulting in large BL and FT aerosol concentrations. Observed sulfate concentrations are well reproduced by the model in the lower 5 km, indicating adequate vertical transport at these altitudes. Ammonium, nitrate, and OA are slightly underestimated compared to EMeRGe-EU, but overestimated compared to EMeRGe-Asia. These discrepancies are likely due to biases in the emission inventories or the limited horizontal resolution.

The CAFE-Africa mission aimed at investigating biomass burning emissions and long-range transport. Simulated mass concentrations in the lower 5 km generally agree well with observations, except for high simulated outliers in OA mass concentrations around 2 km. OA and ammonium (near zero) are well reproduced at higher altitudes, while sulfate and nitrate mass concentrations are slightly underestimated.

The CAFE-EU mission (2020) investigated atmospheric impacts of the Covid lockdown, and we applied lockdown-induced anthropogenic emission reductions following Doumbia et al. (2021). Observations in the BL and lower FT are well reproduced. However, above 4 km, the model underestimates sulfate concentrations. Joppe et al. (2024) found that CAFE-EU observations were likely influenced by SO_2 emissions from the Shiveluch eruption, which mixed into the tropopause region and were zonally transported. These emissions were not included in our simulations, as they initially failed to reach the stratosphere. A dedicated model study is planned. The ammonium overestimation in the UT may result from exaggerated detrainment from the South Asian monsoon system (see next section), with a concurrent nitrate overestimation driven by both excess ammonium and low sulfate, promoting ammonium nitrate formation. Enhancements in these altitude regions are also observed, albeit at lower magnitude.

The CAFE-Brazil mission aimed at investigating the Amazon forest-atmosphere system, focusing on the UT. Sulfate concentrations are well reproduced, and ammonium concentrations fall below the detection limit above 4 km. However, BL ammonium concentrations are strongly underestimated, probably due to missing local ammonia sources, e.g., from inundated areas. Simulated nitrate concentrations deviate from observations, with overestimation at lower altitudes and underestimation in the

UT. OC concentrations are overestimated in the FT, but show good agreement in the UT and BL. The FT overestimation likely stems from the missing implementation of the kinetic limitation for condensation for organic species, discussed in Sect. 5.

3.3.3 Additional aircraft campaigns

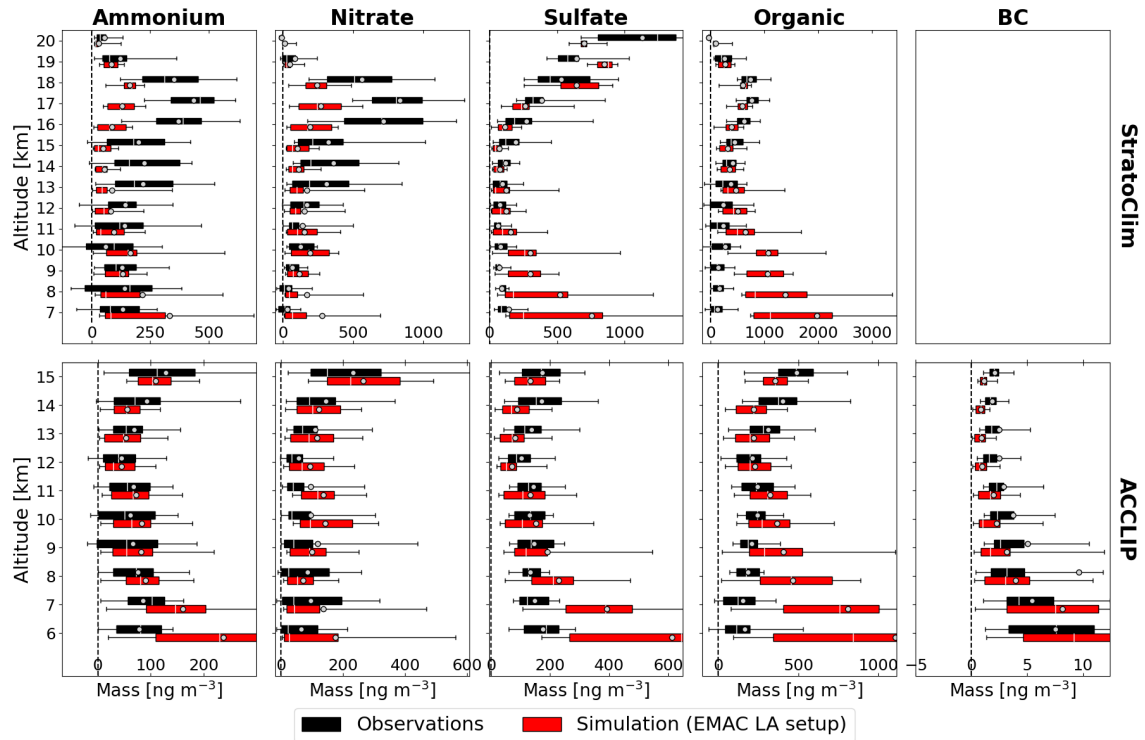


Figure 6. Comparison of simulated (red) and observed (black) mass concentrations of sulfate, ammonium, nitrate, and organic aerosol (all PM₁) from ERICA-AMS measurements, during the StratoClim and ACCLIP missions, and black carbon (BC) measurements from the SP-2 instrument. Box-whisker plots follow the format of Fig. 4. All concentrations are provided at standard temperature and pressure (STP; $T = 273.15$ K & $p = 1013.25$ hPa).

Figure 6 compares simulated aerosol mass concentrations with ERICA-AMS measurements from the StratoClim and ACCLIP campaigns, targeting the ATAL. The evaluation focuses on the UTLS region (12–19 km), where the ATAL forms. While OA and sulfate are overestimated in the BL and lower FT, the ATAL itself is well captured. During StratoClim, enhancements in ammonium, nitrate, and OA were observed but are underestimated in the simulation, likely due to flight planning targeting convective outflow, which the model’s coarse horizontal resolution cannot fully resolve. Notably, the upper limit of the ATAL is also well captured in the simulations. At 20 km, simulated ammonium, nitrate and OA mass concentrations tend towards zero, in close agreement with observations.

The ATAL enhancement observed during the ACCLIP mission is better reproduced by the model. Unlike StratoClim, ACCLIP sampled the ATAL at various stages of its development, making it more representative of the average ATAL, and better reproducible by model simulations. The overestimation of FT OA can be attributed to the same reason as before and will be discussed in Sect. 5. Both campaigns observed low concentrations of primary particles, especially dust and BC, in the ATAL (e.g., Appel et al., 2022; Ebert et al., 2024; Berberich et al., 2025). The model successfully reproduces these low UTLS BC concentrations, with values below 5 ng m^{-3} .

3.4 Stratosphere

For the evaluation of stratospheric aerosol, simulated sAOD and extinction from the three setups are compared with satellite observations merged in GloSSAC. Figure 7 presents a timeline of sAOD from January 2016 to December 2018, derived from GloSSAC observations and model simulation results at the four available GloSSAC wavelengths. The sAOD at the two lower GloSSAC wavelengths depend on observations from SAGE-III/ISS, which became operational in mid-2017 (Kovilakam et al., 2023). Additionally, we provide a comparison of zonally averaged aerosol extinction for the considered time period in Appendix Fig. B1. The displayed period was relatively volcanically quiescent (e.g., Schallcock et al., 2023), mostly representing background stratospheric conditions. A detailed evaluation of the model's skill to reproduce stratospheric SO_2 injections from explosive volcanic eruptions using the EVER submodel is provided by Kohl et al. (2024).

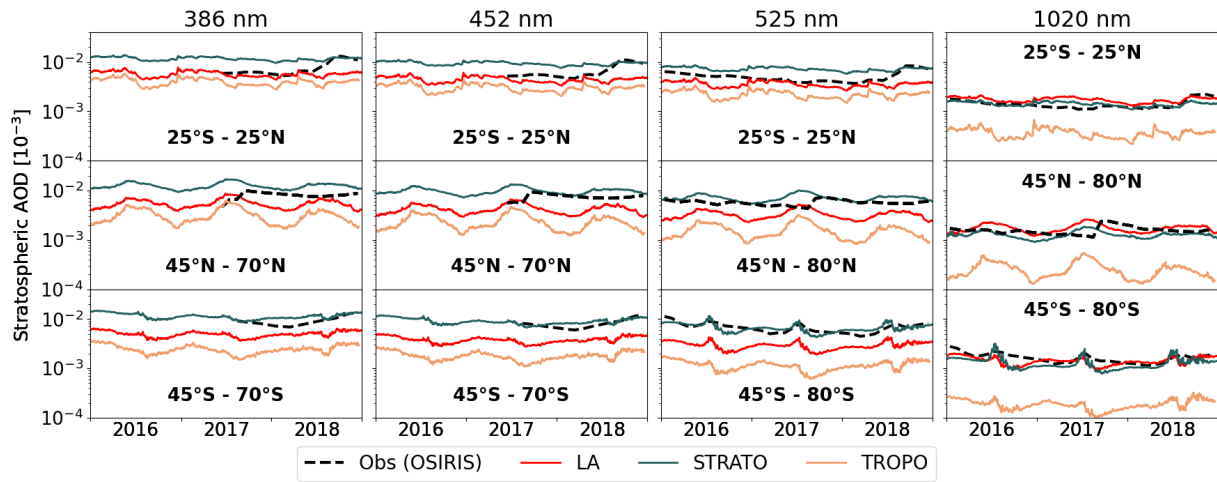


Figure 7. Timeline of simulated stratospheric Aerosol Optical Depth (sAOD) of the three defined setups compared to observations from the GloSSAC database at the four GloSSAC wavelengths, in the tropics (top), NH higher latitudes (middle), and SH higher latitudes (bottom).

In general, the simulation with the stratospheric setup produces the largest AOD, while the tropospheric setup produces the lowest AOD. This is explained by the underlying size distribution, shown in appendix Fig. A2. In the tropospheric setup, aerosols rapidly grow into the coarse mode and subsequently sediment from the stratosphere. In contrast, the accumulation

mode in the STRATO setup has wider mode boundaries and narrower sigma of the log-normal distribution, which limits growth into the coarse mode, resulting in a more realistic stratospheric size distribution.

The introduction of the intermediate mode in the LA setup buffers this effect. While particles still grow into the small coarse modes, sedimentation is reduced compared to the tropospheric setup, although the stratospheric lifetime of the particles slightly decreases compared to the STRATO setup. The lower AOD at shorter wavelengths in the LA setup, compared to STRATO, can also be partially attributed to another effect. According to Mie theory, particles with diameters similar to the wavelength of incident radiation scatter most efficiently, depending on aerosol composition (e.g., Bohren and Huffman, 1998). The newly adjusted size distribution shows a local minimum around 500 nm (see Appendix Fig.A2), caused by the mode boundary at 800nm. This results in a slight reduction in AOD between 400 and 700 nm, a purely computational artifact. As a consequence, both setups show much better agreement at 1020 nm, consistent with observational data.

In addition to the differences between the setups, simulations show systematic deviations from observations (Fig. 7). At northern midlatitudes, the simulated sAOD exhibits seasonality across all wavelengths. This seasonality results from the seasonal stratospheric input of the AMA, which is not observed and is likely overestimated in the model. Moreover, a sudden rise in sAOD is observed at northern midlatitudes, which is not reproduced by the simulations. The observed increase is attributed to the British Columbia wildfires in 2017 (Daniels et al., 2020; Healy et al., 2019). While volcanic eruptions are included in the simulations, we did not account for strong wildfires that directly inject particles and precursor gases into the stratosphere. Another observed feature is the increase of sAOD in the tropics in mid-2018, coinciding with the eruption of the Ambae volcano (Kloss et al., 2020). This eruption is included in the simulations based on the emission inventory by Schallcock et al. (2023), resulting in a slight sAOD enhancement, particularly at 1020 nm. However, the model may underestimate the magnitude and plume top altitude.

Finally, in the northern mid-to-high latitudes, the formation of Polar Stratospheric Clouds (PSCs) is evident in the simulations, leading to enhanced sAOD in the Southern Hemisphere summer. This enhancement is only visible in the GloSSAC datasets at 525 and 1020 nm, as the 386 and 425 nm datasets (based on SAGE-III observations) cover only 70°S to 70°N. The LA and STRATO simulations reproduce this enhancement, while the TROPO simulation results only show a weak signal, at low absolute sAOD.

4 Results

Following the comprehensive evaluation, global aerosol distributions and composition from the surface to the stratosphere are derived from EMAC simulations using the new LA setup¹. For this purpose, the lower atmosphere is subdivided into four layers based on pressure ranges: the BL (1000 – 750 hPa), the FT (750 – 300 hPa), the UTLS (300 – 80 hPa), and the stratospheric Overworld (OW; 80 – 30 hPa).

¹The LA setup was developed and chosen for these simulations, as it performs similar to the TROPO setup for BL conditions, and to the STRATO setup for stratospheric conditions, while representing the total column.

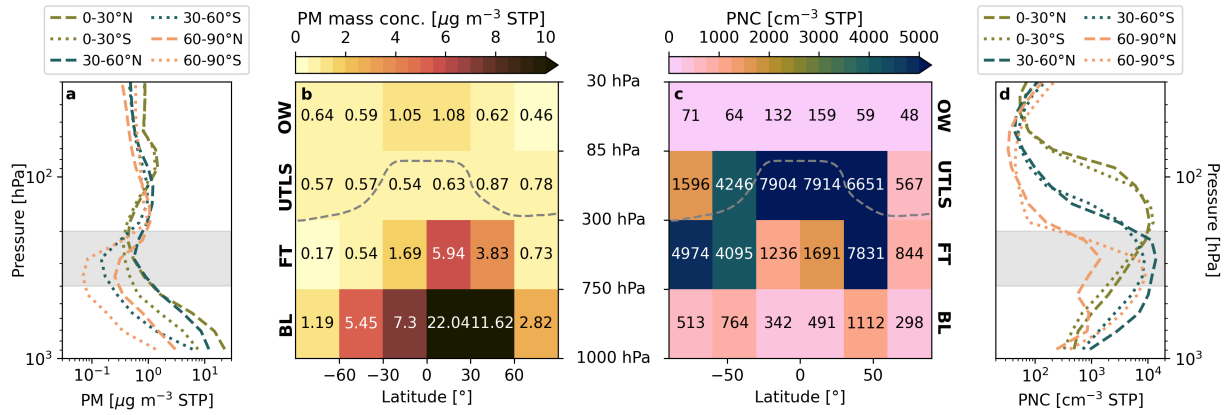


Figure 8. Zonally and annually (2017) averaged global aerosol mass (b) and particle number concentrations (PNC; c) in the boundary layer (BL), the free troposphere (FT), the upper troposphere / lower stratosphere (UTLS), and the stratospheric overworld (OW), along with the corresponding averaged profiles (a,d). The grey shaded area between 400 and 200 hPa marks the transition region between the FT and the UTLS, where lowest mass concentrations are simulated.

Figure 8 presents global particulate matter (PM) mass concentrations (b) and particle number concentrations (PNC; c) within the defined atmospheric layers and across six latitudinal bins, along with the corresponding averaged profiles (a and d, respectively). The composition of PM in each region and layer is shown in Fig. 9, while the horizontal distribution of individual aerosol components is displayed in Fig. 10.

BL aerosol mass concentrations and composition have been extensively studied using both observations (e.g., Zhang et al., 2007; Putaud et al., 2004; Nault et al., 2021) and models (e.g., van Donkelaar et al., 2016; Shen et al., 2024b; Lelieveld et al., 2015). Our simulations agree with these previous studies. Dust dominates aerosol mass in tropical and subtropical northern latitudes (up to 70%), primarily due to emissions from the Sahara, Middle East and Gobi deserts (see Fig. 10). In densely populated regions, SOA is the predominant component, accompanied by POA, sulfate, nitrate (which is overestimated in our model, as discussed above), and smaller contributions from ammonium and BC. Over tropical rainforests, organic matter dominates, either directly from biomass burning as POA or formed via oxidation of biogenic volatile organic compounds (BVOCs) as SOA. Marine regions are characterized by a mixture of sea salt (defined here as bulk sea salt + sodium chloride; exceeding 70% at high southern latitudes) and sulfate aerosol. For global particle number concentrations (PNC), specifically ultrafine particles (UFPs) at the surface, we refer the reader to Kohl et al. (2023) that presented high-resolution (10×10 km) global distributions based on a similar tropospheric setup.

The FT remains dominated by dust, exhibiting similar spatial patterns to those in the BL, yet more diffuse and reduced in absolute mass concentrations (Fig. 10). The FT is still strongly influenced by surface emissions, mostly driven by convective transport of intermediate strength and outflow altitude (congestus type or weak cumulonimbus) without substantial removal

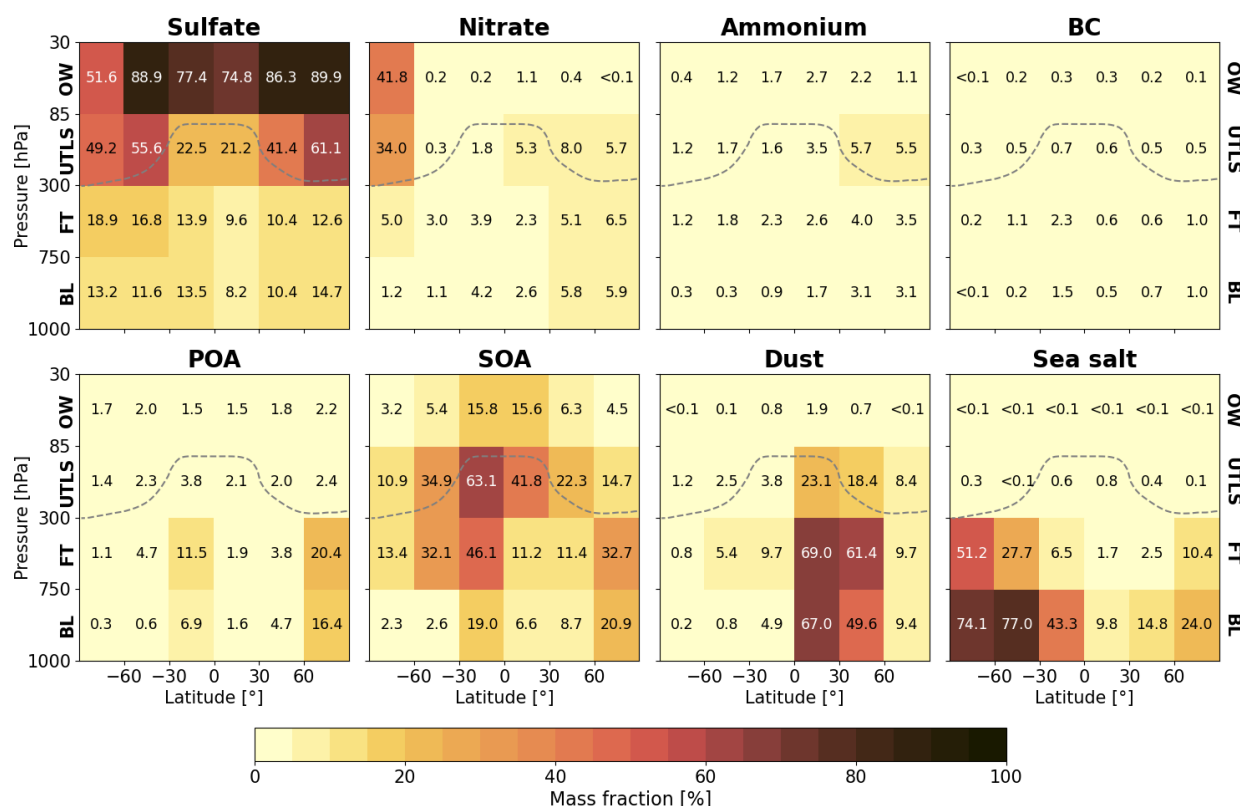


Figure 9. Zonally and annually (2017) averaged aerosol composition, subdivided into four vertical layers and six latitudinal bins, as defined in Fig. 8, showing the major aerosol species in the Lower Atmosphere.

of particles by precipitation. The importance of SOA increases in the FT, as oxygenated volatile organic compounds start to condense at decreasing temperatures.

The UTLS region can be subdivided into the UT in the tropics and the lowermost stratosphere at higher latitudes, that are separated by transport barriers (e.g., Gettelman et al., 2011). The tropical UT is mostly influenced by deep convective updraft of anthropogenic or biogenic precursor gases, that form new particles (mostly SOA here, with additional contributions of ammonium nitrate particularly in the ATAL). Primary particles, such as dust, BC and POA are reduced in the tropical UT, as they are too heavy, or washed out in convective clouds (Ebert et al., 2024; Appel et al., 2022; Berberich et al., 2025; Lund et al., 2018). However, in the NH tropical and subtropical UTLS, significant amounts of dust are present, likely overestimated (see Sect. 3) due to underestimated wet removal originating from the representation of parametrised convection as grid box mean mass fluxes.

The stratosphere is strongly dominated by liquid sulfur aerosol, that forms either as a consequence of OCS transport from the surface, photolytically converted to SO_2 , and subsequently oxidized to H_2SO_4 (Brühl et al., 2012), or from explosive volcanic eruptions (Schallock et al., 2023; Schmidt et al., 2018; Kohl et al., 2024). In the tropical stratosphere, sulfate accounts for about 75% of the aerosol mass, with a notable 15% contribution from SOA, which decreases rapidly with altitude (see Fig. 11). In the subtropical and polar stratospheric overworld, sulfate fractions increase to around 90%. However, in the southern polar regions, this dominance is reduced due to polar stratospheric cloud formation, leading to an annual average nitrate contribution of 42%.

Primary particle abundance is limited in the stratosphere, falling below 2.5% globally in the stratospheric overworld. This is in contrast to previous model studies (e.g., Bossolasco et al., 2021; Ma et al., 2019; Brühl et al., 2018), that found dust to be an important species in the tropical and subtropical UTLS, and the lower stratosphere. However, recent observations revealed very small amounts of dust in the UTLS and lower stratosphere, below 10 ng m^{-3} (Ebert et al., 2024; Appel et al., 2022). We still simulate larger values compared to these estimates (20 ng m^{-3} in the tropical stratosphere), yet much lower than Bossolasco et al. (2021), Ma et al. (2019) and Brühl et al. (2018). More details will be provided in Sect. 5.

Lowest aerosol mass concentrations are simulated in the transition from the FT to the UTLS, typically between 400 and 200 hPa (grey shaded area in Fig. 8a). The surface, BL, and the stratosphere serve as primary and secondary sources of atmospheric aerosols, respectively. The primary aerosols originating from the surface are largely removed by wet deposition during convective uplift, while stratospheric aerosol influence generally extends only down to the upper FT. This global minimum in PM mass concentrations reduces the condensational sink, and thereby enables strong NPF (see Fig. 8d). Maximum NPF is simulated at the altitude of the mass minimum at higher latitudes, whereas in the tropics, the peak in particle number concentration (PNC) is slightly elevated, likely due to increased availability of precursor gases in the deep convective outflow of the UT.

Figure 11 provides a more detailed view of aerosol composition profiles across global regions. While BL composition reflects regional surface sources, some consistent global patterns emerge. As previously discussed, all regions exhibit a distinct minimum in PM mass concentrations between 400 and 200 hPa, separating the surface-dominated lower atmosphere from the sulfur chemistry-dominated UT and stratosphere. Only a few intense source regions, such as the Sahara, South and East Asia, South America, and Central Africa, significantly impact the UTLS and the stratosphere. Above the tropopause, the global Junge layer forms, composed primarily of liquid sulfur aerosols.

Elevated OA concentrations are simulated in the UTLS across most regions, diminishing near 70–80 hPa, consistent with StratoClim observations showing organic contributions of $\sim 40\%$ up to 19 km (Appel et al., 2022). Additionally, Figs. 6 and 4 show that lower stratospheric OA mass concentrations are well reproduced, both at the top of the ATAL, and in the lowermost stratosphere over the Southern Ocean and at high northern latitudes. These elevated SOA concentrations in the tropical lower stratosphere (originating from key source regions such as the Amazon Basin, Central Africa, and South & East Asia) form a global band (Fig. 10) driven by strong horizontal winds and are transported throughout the UTLS.

5 Discussion & Limitations

We present atmospheric aerosol distributions and composition from the surface to the stratosphere derived from EMAC simulations, based on a new model setup and thorough evaluation. The new setup was developed by optimizing former tropospheric and stratospheric setups, and introducing an additional mode between the accumulation and coarse mode (small coarse mode).

The new setup provides the best overall agreement for aerosol simulations in the lower atmosphere. It performs similarly to the TROPO setup in the BL and to the STRATO setup in the stratosphere, while most accurately capturing vertical profiles. Key differences between the original setups lie in the width and sigma of the accumulation mode: the TROPO setup allows rapid growth into the coarse mode, shortening aerosol lifetimes in the stratosphere, while the STRATO setup suppresses this growth, extending lifetimes. Using either setup beyond its intended domain leads to an overestimation or underestimation of aerosol lifetimes, respectively. The additional small coarse mode in the new configuration buffers this issue: near the surface, it allows modest growth and slightly shorter lifetimes than the STRATO setup; in the stratosphere, it slows coarse mode formation and enhances lifetimes compared to TROPO setup. Thus, the new LA setup offers a balanced representation across the lower atmosphere. Future developments could consider making the growth into larger modes dependent on the atmospheric domain.

This evaluation also revealed some discrepancies, attributable to general limitations in global atmospheric aerosol modelling or shortcomings of the EMAC model in particular.

The evaluation revealed two key global-scale weaknesses of the LA setup, likely interconnected. First, sulfate concentrations are systematically underestimated in the remote FT, while closer to source regions they are well-captured. This underestimation is more pronounced than in the STRATO setup. We hypothesize that overly efficient growth from the accumulation to the small coarse mode during long-range transport, followed by sedimentation or reduced transport efficiency, contributes to this bias. Second, in regions with the strongest underestimation of aerosol mass, PNC is most strongly overestimated. In our model, FT and UT NPF is primarily driven by ammonia, sulfuric acid, and nitric acid (Xenofontos et al., 2024; Wang et al., 2022). While the nucleation rates may be overestimated, the stronger bias in the LA setup compared to TROPO and STRATO suggests that the main issue lies in the underestimation of the condensational sink. Future work will aim to improve background sulfur representation in the FT to better constrain PNC and CCN formation in remote regions.

The modelling of OA formation remains challenging. Here, gas-aerosol partitioning is treated with the ORACLE submodel (Tsimpidi et al., 2014, 2017), that was specifically developed and evaluated for the lower troposphere. In its current implementation, ORACLE is not considering the Kelvin effect and the kinetic limitation for condensation on available aerosol surfaces, which is in contrast to the inorganic phase partitioning, that takes the kinetic limitations into account. Equilibrium gas-aerosol partitioning is calculated, and distributed onto the soluble modes. However, particularly in the FT and UTLS, where aerosol surface areas are minimal, equilibrium may not be reached due to kinetically limited condensation. In addition, the mixing times of organic compounds within SOA are crucial for predicting aerosol growth and evaporation. While organic mixing times in most of the BL are below one hour, supporting the use of equilibrium partitioning, this assumption may not hold in the FT and UTLS. In these regions, mixing times can extend to several hours, which is significantly longer than typical model

555 timesteps, and SOA may exist in a glassy, highly viscous state (Maclean et al., 2021). These combined effects potentially lead to an overestimation of OA in the FT, as is evident in the model evaluation near source regions.

Moreover, a set of simplifications is applied for the modelling of organic gas phase and particulate species, as the individual modelling of all organic species is not computationally feasible. First, gas phase oxidation products of primary emitted VOCs are combined in lumped species, ageing with fixed yields, as described by Tsimpidi et al. (2014). Second, these already
560 lumped oxidation products are further combined in volatility bins in the particulate phase. Reaction rates, photochemical removal, solubility, and volatility along with its temperature dependence may vary for the individual species, leading to a more complex picture of OA, and its transport and formation from the surface to the stratosphere. In future simulations, the more comprehensive chemical mechanism MOM (Pozzer et al., 2022; Sander et al., 2019) can be applied, with greatly increased computational costs. The transition to the new base model ICON will facilitate global simulations with more complex
565 chemistry, using nesting, and allowing for variable horizontal resolution around the globe.

The transport of primary particles into the stratosphere remains a subject of ongoing debate. Earlier modelling studies (e.g., Bossolasco et al., 2021; Ma et al., 2019; Brühl et al., 2018) concluded that dust plays a dominant role in the ATAL, and is subsequently transported to the stratosphere. In contrast, recent observational studies by Appel et al. (2022) and Ebert et al. (2024) reported very low concentrations of primary material within the ATAL with upper limits of 10 ng m^{-3} .

570 In our simulations, desert dust concentrations in the UTLS were reduced by adjusting emission size distribution in accordance with the new model setup. In the previous TROPO and STRATO setups, desert dust was emitted partly in the accumulation mode and upon ageing became internally mixed with particles that had recently grown into this mode, averaging at around 300 nm diameter. This configuration led to an overestimation of vertical transport, as it effectively downscaled the typical size of dust particles, originally around 900 nm, thus allowing them to reach higher altitudes more readily. Emitting desert dust
575 to the additional small coarse modes reduces this vertical transport and stratospheric dust concentrations. Appendix Fig. A1a illustrates this difference in PSD in desert regions. The accumulation mode in the LA setup is less pronounced compared to the STRATO and TROPO setup, and the particles shift to larger sizes. However, dust in the UTLS remains slightly overestimated with the LA setup. The good agreement between simulated and observed gas-phase tracers such as CO, indicating reasonable overall vertical transport, suggests that insufficient wet scavenging is most likely the cause of this overestimation, likely
580 attributed to the grid-box averaged parametrisation of convective transport in the model

The presented simulation results do not include meteoric material. While being most important in the mesosphere, meteoric material is also observed in the lower stratosphere, especially in the polar vortex, and down to the UTLS (Murphy et al., 1998; Curtius et al., 2005; Murphy et al., 2007; Schneider et al., 2021). Despite its low abundance in mass fraction, it can influence microphysics and chemical properties of stratospheric aerosol by suppressing NPF, and forming condensational sinks. The
585 global burden of meteoric material, and spacecraft debris (Murphy et al., 2023) are still mostly unknown. Future modelling studies will investigate these processes for the stratosphere.

Despite the discussed model shortcomings, simulations generally show reasonable agreement with observations across all layers and most regions in the atmosphere. We find that vertical profiles of total aerosol mass concentrations exhibit a minimum between 400 hPa and 200 hPa, consistently around the globe, representing the intersection of BL and stratospheric transport,

and enabling frequent NPF and high PNC not only in the tropics and subtropics as observed by Williamson et al. (2019), Weigel et al. (2021) and Curtius et al. (2024), but across all latitudes. As this transition region between the FT and UTLS is important for cloud formation, it is sensitive to indirect climate effects due to aerosol-cloud interactions.

The evaluation of simulated Particle Number Concentrations (PNC) during the ATom campaign revealed several noteworthy patterns. In most regions of the FT and UTLS, PNC is slightly overestimated (see above). However, some regions also exhibit an underestimation in PNC, likely due to missing NPF pathways in the model. In contrast to the UT regions, the PNC is underestimated over the Atlantic Tropics and the East Pacific, the regions closest to the Amazon rainforest, where recent studies suggest that NPF from isoprene oxidation products can occur in the UT (Curtius et al., 2024; Shen et al., 2024a), a process not yet implemented in the model. Additionally, PNC over the Southern Ocean is underestimated, potentially due to the absence of NPF pathways involving iodic acid and methanesulfonic acid (MSA) (Baccarini et al., 2021; He et al., 2021; Zhang et al., 2025).

This work demonstrated the capability of the new LA setup for the EMAC model to simulate atmospheric aerosols from the surface to the stratosphere. For that purpose, no aerosol feedbacks with the chemistry and climate were included in the presented simulations. For example, heterogeneous chemistry and the direct aerosol radiative effect are calculated based on a sulfuric acid aerosol and optical properties climatology, respectively. Further, cloud formation is treated independently on online CCN availability. Future coupling will allow online radiative forcing calculations based on global aerosol distributions, and studies on stratospheric ozone variability induced by aerosol. Moreover, the anthropogenic influence on aerosol distributions across all layers of the lower atmosphere can be calculated, and aircraft observations can be supported by detailed model process studies.

6 Conclusions

Global aerosol distributions and composition have been presented from the surface to the stratosphere, based on numerical simulations performed with the ECHAM/MESSy Atmospheric Chemistry (EMAC) model. These simulations represent a strongly evaluated assessment of global distributions of key aerosol species across all layers of the lower and middle atmosphere.

The global simulations are based on the development of a new setup for the EMAC model, combining previous tropospheric and stratospheric aerosol setups, and applying a newly customized modal aerosol size distribution. An additional intermediate mode between the accumulation and coarse size buffers growth into the coarse mode in the stratosphere, while accurately representing boundary layer (BL) removal processes. The new setup has been comprehensively evaluated against observations, showing generally good agreement across layers and regions.

Discrepancies include overestimated stratospheric concentrations of black carbon (BC) and dust, likely due to underestimated removal processes, as well as overestimated organic aerosol in the free troposphere (FT) due to the lack of an implemented kinetic limitation for condensation on available aerosol surfaces, but also because the equilibrium assumption for phase partitioning may not hold in the FT. Additionally, FT sulfate is underestimated, leading to an overestimation of particle number concentrations (PNC), and surface-level nitrate is overestimated.

The simulations reveal a global minimum in aerosol mass concentrations between 400 and 200 hPa, marking the transition from the FT to the upper troposphere and lowermost stratosphere (UTLS), and separates BL and stratospheric influenced atmospheric domains. This minimum facilitates the formation of new particles, leading to a peak in PNC at altitudes close to or slightly above this minimum. Underestimations of PNC in regional UT and over the Southern Ocean align with proposed NPF pathways from isoprene oxidation products, iodic acid, and methane sulfonic acid, respectively.

In the stratosphere, liquid sulfur aerosol dominates the Junge layer, consistent with previous findings. Secondary OA (SOA) contributes $\sim 15\%$ to the tropical stratospheric aerosol burden, but declines sharply with altitude, in line with recent observations. These elevated SOA concentrations form a tropical band and are transported throughout the UTLS.

We find that primary particles contribute less than 2.5% to aerosol mass in the stratospheric overworld, significantly lower than reported by previous model studies, but more consistent with recent observations. The lower abundance of primary particles in the stratosphere is attributed to a revised emission size distribution tailored to the new modal aerosol setup. Further in-situ observations of stratospheric aerosol composition are needed to validate these findings.

The simulation results presented here represent a first assessment of lower atmospheric aerosol burdens. With additional observational constraints and continued model development, these simulations can be further improved. Using this framework, a number of additional studies can be performed, such as assessments of anthropogenic influence on aerosol distributions across all layers of the lower atmosphere, detailed aerosol process studies, and simulations including online aerosol–cloud–radiation interactions. These studies may contribute to a better understanding of the role of aerosols in cloud formation and help reduce uncertainties in our knowledge about the indirect aerosol radiative forcing of the climate.

Appendix A: Particle size distributions in model setups.

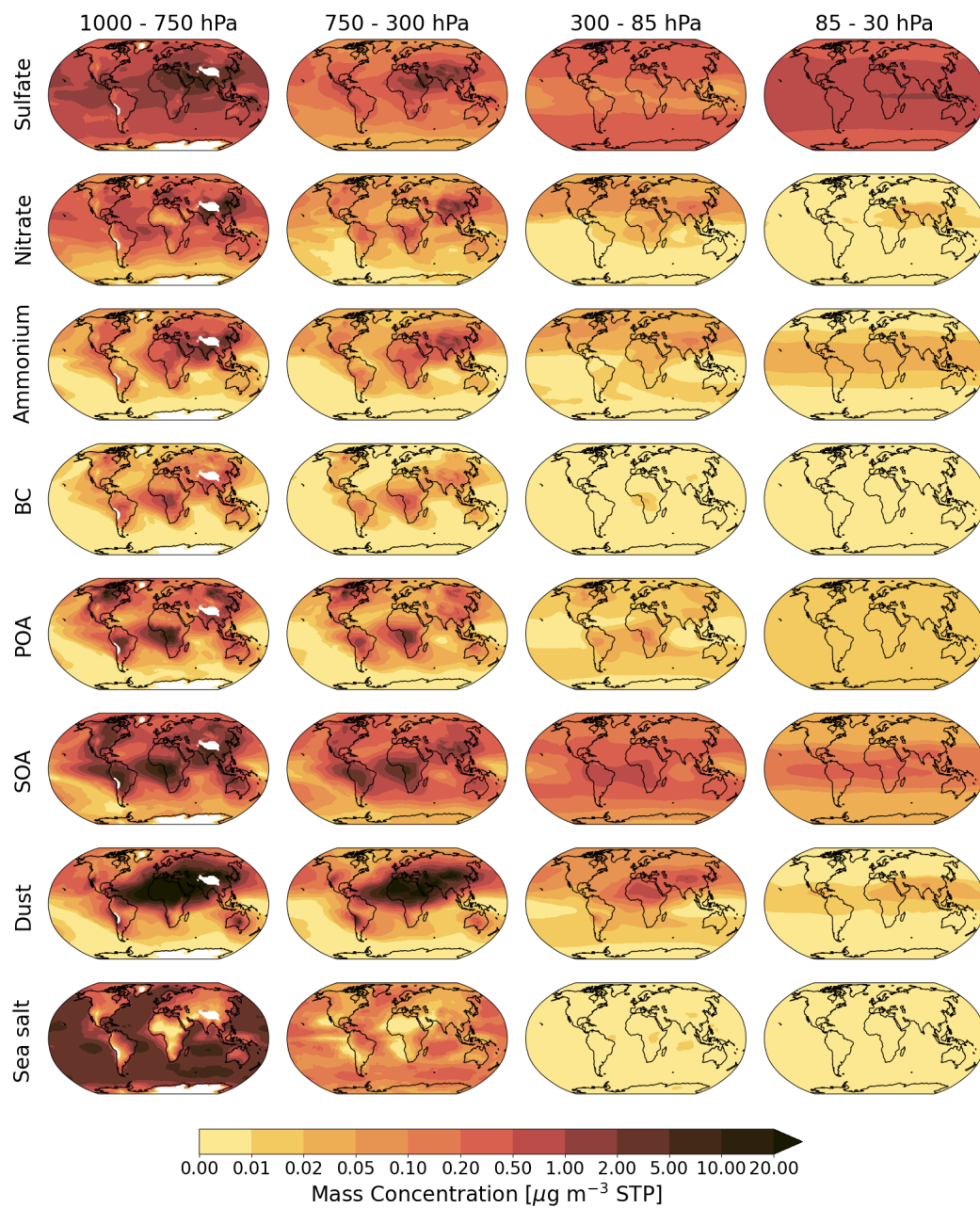


Figure 10. Annually averaged (2017) horizontal distributions of mass concentrations of the dominant aerosol species in the four vertical layers as defined in Fig. 8.

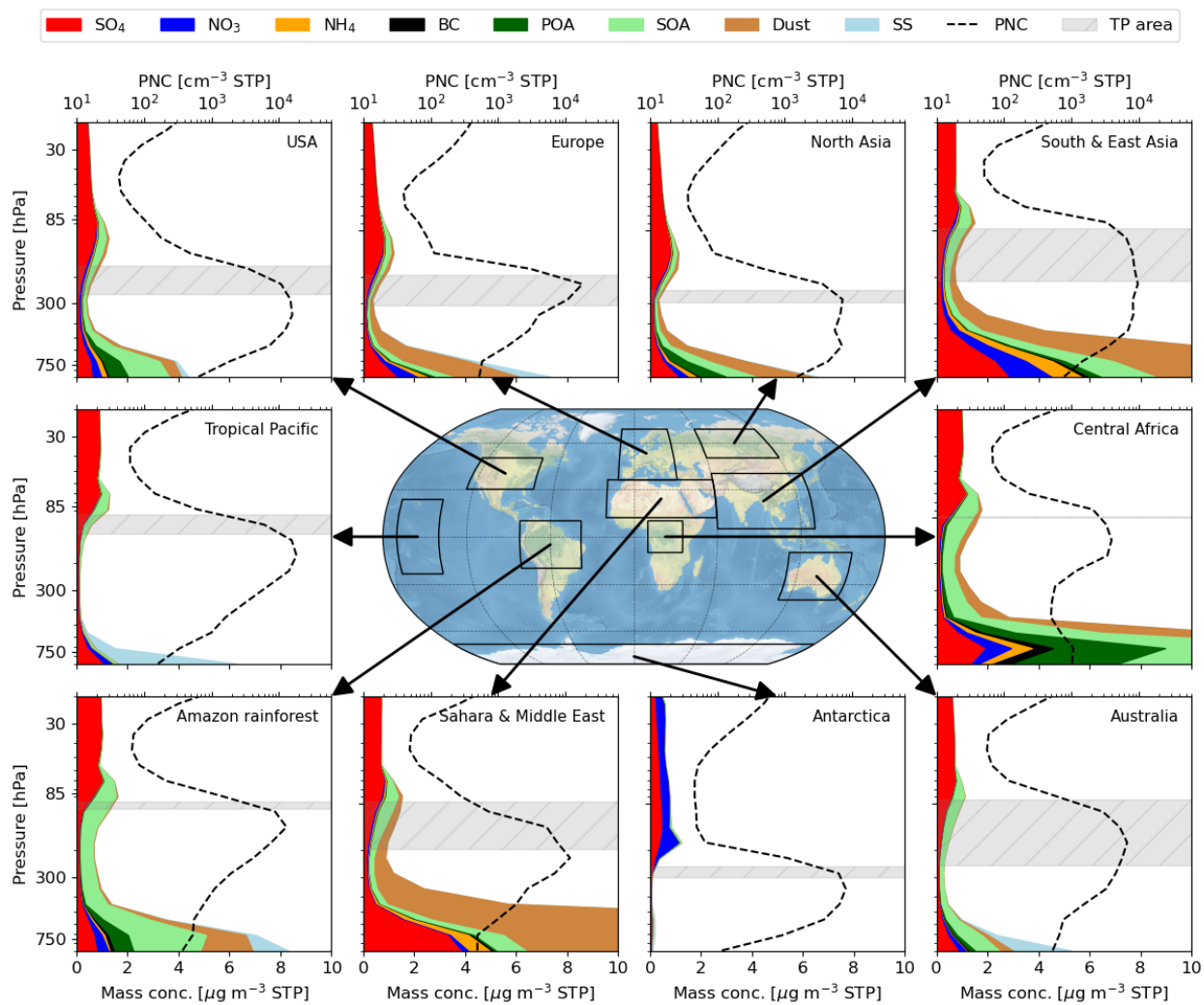


Figure 11. Profiles of annually averaged (2017) aerosol mass concentrations and corresponding composition in selected regions worldwide, spanning from the boundary layer (BL) to the stratospheric overworld (OW). Particle Number Concentrations (PNC) are shown as black dashed lines. The grey shaded area indicates the range of monthly averaged tropopause altitudes.

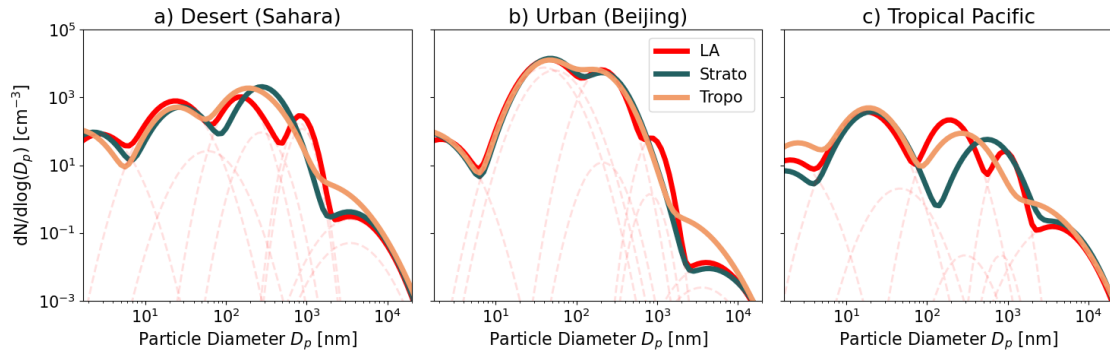


Figure A1. Simulated Particle Size Distributions (PSD) at the Earth's surface (averaged over June 2017) for the different model setups in: a) the Sahara Desert, b) an urban region (central Beijing), and c) the Tropical Pacific. The red dashed lines indicate the different modes in the LA setup.

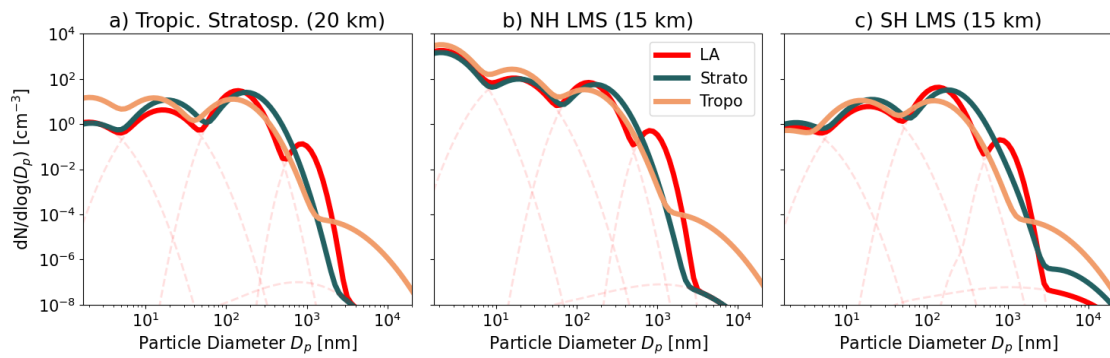


Figure A2. Simulated stratospheric Particle Size Distributions (PSD; averaged over June 2017) for the different model setups in: a) the tropical stratosphere (20°S–20°N) at 20 km, b) the Northern Hemisphere (NH) lowermost stratosphere (LMS; 45°–80°N) at 15 km altitude, and c) the Southern Hemisphere (SH) lowermost stratosphere (LMS; 45°–80°S) at 15 km altitude. The red dashed lines indicate the different modes in the LA setup. Note the differing y axis from Fig. A1.

Appendix B: Simulated stratospheric extinction compared to the GLoSSAC dataset

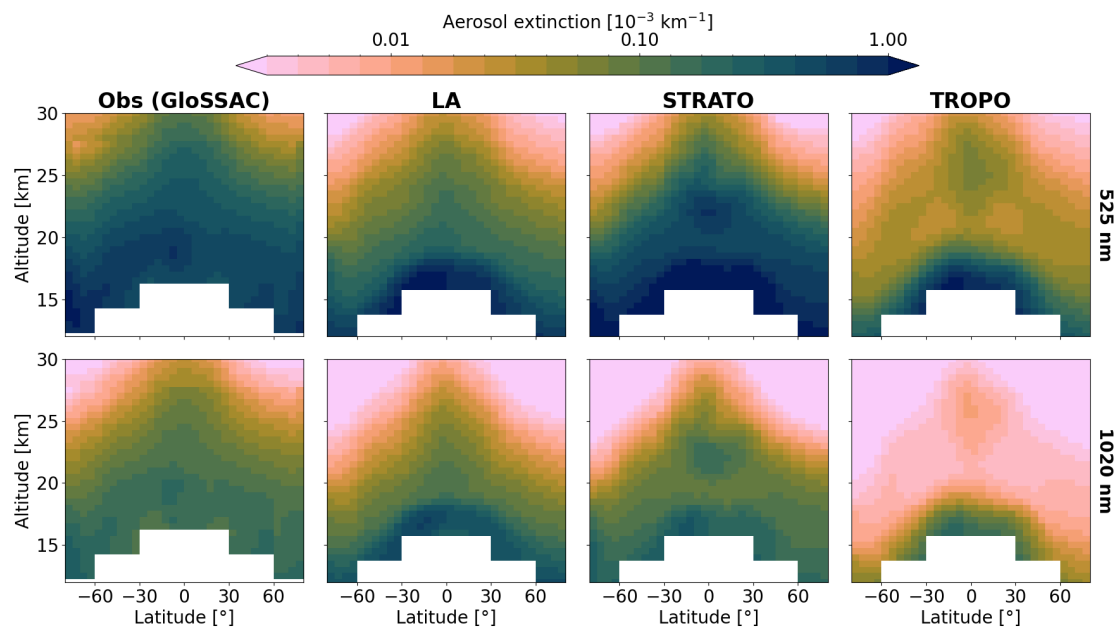


Figure B1. Zonally and temporally (2016–2018) averaged profiles of aerosol extinction, derived from EMAC model simulations compared to GLoSSAC observations at 525 and 1020 nm.

Appendix C: ATom flight paths and additional evaluation

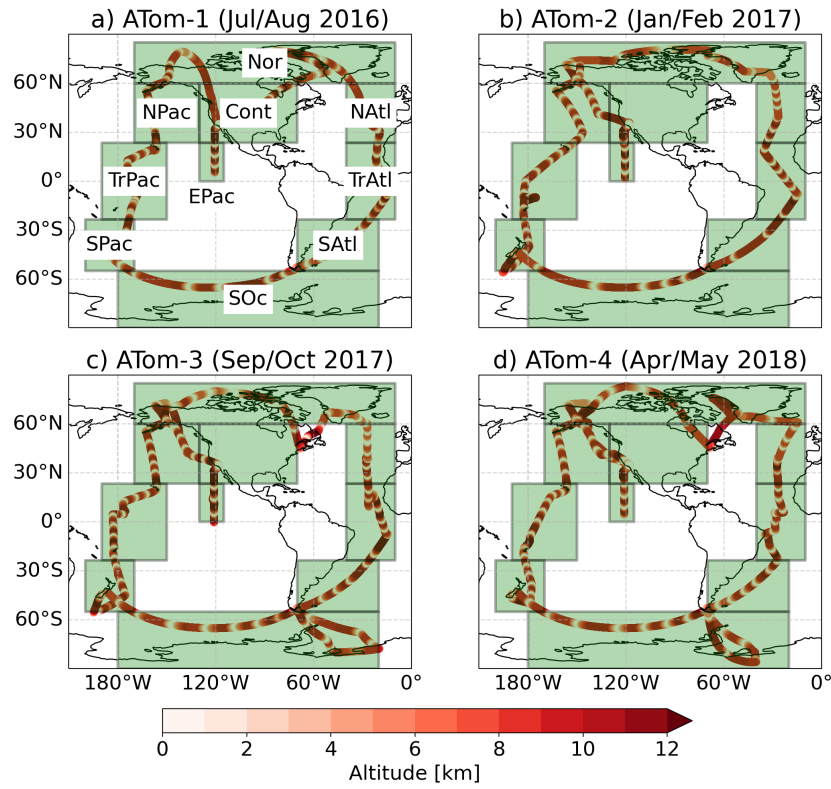


Figure C1. Flight paths during the four seasonal sub-campaigns of the NASA Atmospheric Tomography (ATom) mission: a) ATom-1 (July/August 2016), b) ATom-2 (January/February 2017), c) ATom-3 (September/October 2017), and d) ATom-4 (April/May 2018). Numerous vertical profiles were recorded, with altitude distributions indicated by red shading. For evaluation, flight paths were categorized into 10 characteristic regions: Tropical Pacific (TrPac), Tropical Atlantic (TrAtl), North Pacific (NPac), North Atlantic (NAtl), South Pacific (SPac), South Atlantic (SATl), East Pacific (EPac), Continental US/Canada (Cont), Northern Polar (Nor), and Southern Ocean (SOc).

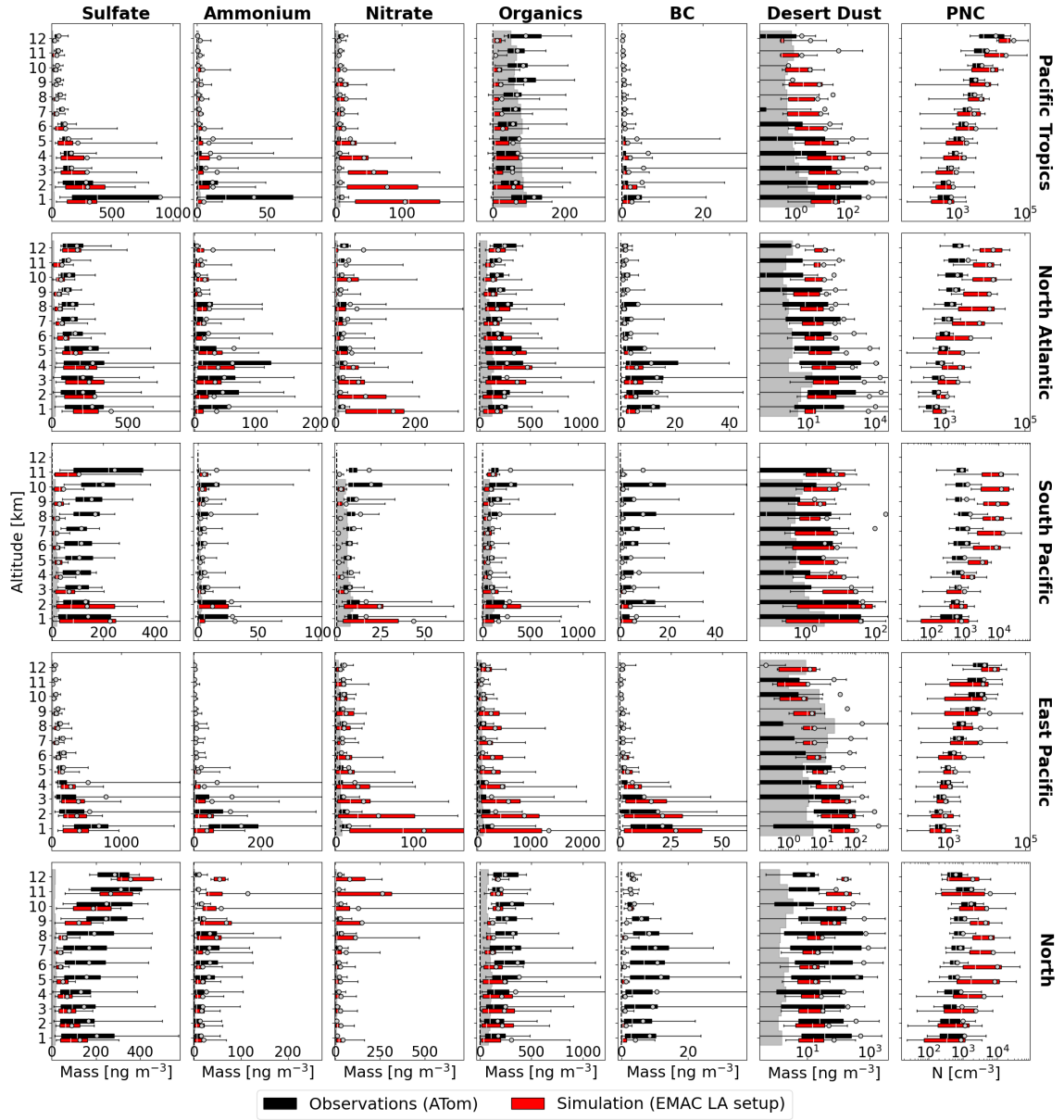


Figure C2. As Fig. 4, but for the remaining regions.

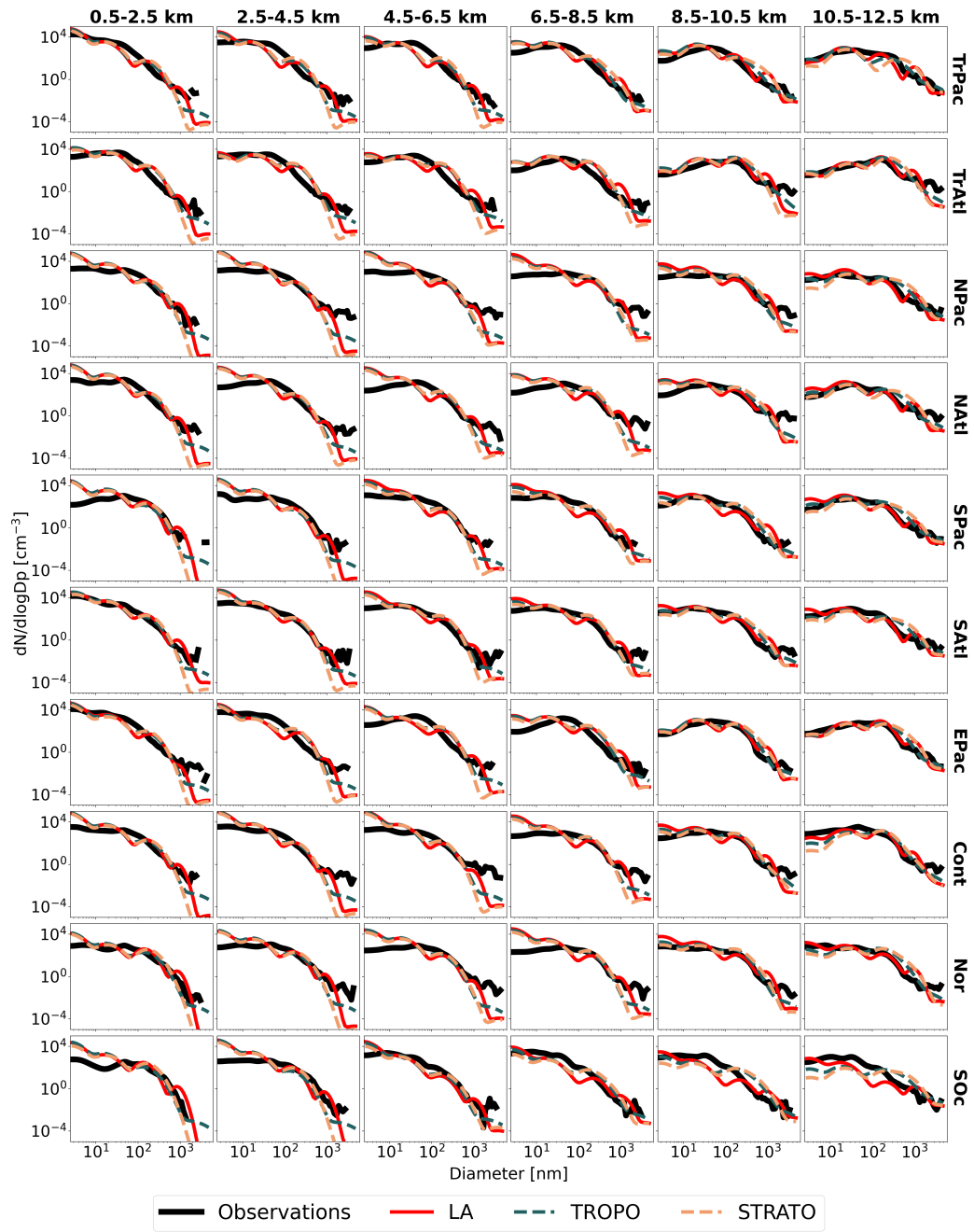


Figure C3. Comparison of simulated particle size distributions from the three model setups with ATom observations (black) across selected regions, as defined in Fig. C1.

Code and data availability. Will be provided upon submission of the manuscript.

Author contributions. M.K. led the development of the new model setup, performed and evaluated the simulations, analysed the results, and wrote the manuscript. A.P., C.Brü., H.T., C.X., T.C., and S.G. contributed to the model development. O.A., C.Bro., O.E., K.F., J.J., P.Jop., K.Ka., F.K., J.Schn., C.S., and J.Schw. provided observational datasets. P.Jöc., K.Kl., A.M., S.R., A.T., and R.V. contributed via testing and discussion on the new setup. J.L., H.T., and A.P. supervised the work. All authors contributed to the editing of the manuscript.

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References

- 655 Allen, H., Crounse, J., Kim, M., Teng, A., Xu, L., and Wennberg, P.: ATom: In Situ Data from Caltech Chemical Ionization Mass Spectrometer (CIT-CIMS), V2, ORNL Distributed Active Archive Center [data set], <https://doi.org/10.3334/ORNLDAAAC/1927>, 2021.
- Andreae, M. O.: Emission of trace gases and aerosols from biomass burning – an updated assessment, *Atmospheric Chemistry and Physics*, 19, 8523–8546, <https://doi.org/10.5194/acp-19-8523-2019>, 2019.
- Appel, O., Köllner, F., Dragoneas, A., Hünig, A., Molleker, S., Schlager, H., Mahnke, C., Weigel, R., Port, M., Schulz, C., Drewnick, F., Vogel, B., Stroh, F., and Borrmann, S.: Chemical analysis of the Asian tropopause aerosol layer (ATAL) with emphasis on secondary aerosol particles using aircraft-based in situ aerosol mass spectrometry, *Atmospheric Chemistry and Physics*, 22, 13 607–13 630, <https://doi.org/10.5194/acp-22-13607-2022>, 2022.
- 660 Baccarini, A., Dommen, J., Lehtipalo, K., Henning, S., Modini, R. L., Gysel-Beer, M., Baltensperger, U., and Schmale, J.: Low-Volatility Vapors and New Particle Formation Over the Southern Ocean During the Antarctic Circumnavigation Expedition, *Journal of Geophysical Research: Atmospheres*, 126, e2021JD035 126, <https://doi.org/https://doi.org/10.1029/2021JD035126>, 2021.
- 665 Berberich, J., Jacoby, S. A., Michailoudi, G., Schwarz, J. P., Viciani, S., D’Amato, F., Bianchini, G., Barucci, M., Campos, T., Ullman, K., Podolske, J. R., Gurganus, C., Smith, W. P., Ueyama, R., Honomichl, S. B., Pan, L. L., Woods, S., Wienzierl, B., Dollner, M., and Perring, A. E.: Black Carbon Reflects Extremely Efficient Aerosol Wet Removal in Monsoonal Convective Transport, *Journal of Geophysical Research: Atmospheres*, 130, e2024JD042 692, <https://doi.org/https://doi.org/10.1029/2024JD042692>, 2025.
- 670 Bohren, C. F. and Huffman, D. R.: Absorption and Scattering of Light by Small Particles, chap. 4, pp. 82–129, John Wiley & Sons, Ltd, ISBN 9783527618156, <https://doi.org/https://doi.org/10.1002/9783527618156.ch4>, 1998.
- Bossolasco, A., Jegou, F., Sellitto, P., Berthet, G., Kloss, C., and Legras, B.: Global modeling studies of composition and decadal trends of the Asian Tropopause Aerosol Layer, *Atmospheric Chemistry and Physics*, 21, 2745–2764, <https://doi.org/10.5194/acp-21-2745-2021>, 2021.
- 675 Boucher, O.: Atmospheric Aerosols, pp. 9–24, Springer Netherlands, Dordrecht, ISBN 978-94-017-9649-1, https://doi.org/10.1007/978-94-017-9649-1_2, 2015.
- Bourassa, A. E., Rieger, L. A., Lloyd, N. D., and Degenstein, D. A.: Odin-OSIRIS stratospheric aerosol data product and SAGE III inter-comparison, *Atmospheric Chemistry and Physics*, 12, 605–614, <https://doi.org/10.5194/acp-12-605-2012>, 2012.
- Bowdalo, D.: GHOST: A globally harmonised dataset of surface atmospheric composition measurements, Zenodo [data set], <https://doi.org/10.5281/zenodo.10637450>, 2024.
- 680 Bowdalo, D., Basart, S., Guevara, M., Jorba, O., Pérez García-Pando, C., Jaimes Palomera, M., Rivera Hernandez, O., Puchalski, M., Gay, D., Klausen, J., Moreno, S., Netcheva, S., and Tarasova, O.: GHOST: a globally harmonised dataset of surface atmospheric composition measurements, *Earth System Science Data*, 16, 4417–4495, <https://doi.org/10.5194/essd-16-4417-2024>, 2024.
- Brock, C., Kupc, A., Williamson, C., Froyd, K., Erdesz, F., Murphy, D., Schill, G., Gesler, D., McLaughlin, R., Richardson, M., Wagner, N., and Wilson, J.: ATom: L2 In Situ Measurements of Aerosol Microphysical Properties (AMP), ORNL Distributed Active Archive Center [data set, <https://doi.org/10.3334/ORNLDAAAC/1671>, 2019a.
- 685 Brock, C. A., Williamson, C., Kupc, A., Froyd, K. D., Erdesz, F., Wagner, N., Richardson, M., Schwarz, J. P., Gao, R.-S., Katich, J. M., Campuzano-Jost, P., Nault, B. A., Schroder, J. C., Jimenez, J. L., Weinzierl, B., Dollner, M., Bui, T., and Murphy, D. M.: Aerosol size distributions during the Atmospheric Tomography Mission (ATom): methods, uncertainties, and data products, *Atmospheric Measurement Techniques*, 12, 3081–3099, <https://doi.org/10.5194/amt-12-3081-2019>, 2019b.
- 690

- Brühl, C., Lelieveld, J., Crutzen, P. J., and Tost, H.: The role of carbonyl sulphide as a source of stratospheric sulphate aerosol and its impact on climate, *Atmospheric Chemistry and Physics*, 12, 1239–1253, <https://doi.org/10.5194/acp-12-1239-2012>, 2012.
- Brühl, C., Schallcock, J., Klingmüller, K., Robert, C., Bingen, C., Clarisse, L., Heckel, A., North, P., and Rieger, L.: Stratospheric aerosol radiative forcing simulated by the chemistry climate model EMAC using Aerosol CCI satellite data, *Atmospheric Chemistry and Physics*, 18, 12 845–12 857, <https://doi.org/10.5194/acp-18-12845-2018>, 2018.
- Brune, W., Miller, D., and Thames, A.: ATom: Measurements from Airborne Tropospheric Hydrogen Oxides Sensor (ATHOS), V2, ORNL Distributed Active Archive Center[data set], <https://doi.org/10.3334/ORNLDAAAC/1930>, 2021.
- Canagaratna, M., Jayne, J., Jimenez, J., Allan, J., Alfarra, M., Zhang, Q., Onasch, T., Drewnick, F., Coe, H., Middlebrook, A., Delia, A., Williams, L., Trimborn, A., Northway, M., DeCarlo, P., Kolb, C., Davidovits, P., and Worsnop, D.: Chemical and microphysical characterization of ambient aerosols with the aerodyne aerosol mass spectrometer, *Mass Spectrometry Reviews*, 26, 185–222, <https://doi.org/https://doi.org/10.1002/mas.20115>, 2007.
- Choi, J.-O. and Chung, C. E.: Sensitivity of aerosol direct radiative forcing to aerosol vertical profile, *Tellus B: Chemical and Physical Meteorology*, 66, 24 376, <https://doi.org/10.3402/tellusb.v66.24376>, 2014.
- Crutzen, P. J.: The possible importance of CSO for the sulphatelaye of the stratosphere, *Geophysical Research Letters*, 3, 73–76, 1976.
- Curtius, J., Weigel, R., Vössing, H.-J., Wernli, H., Werner, A., Volk, C.-M., Konopka, P., Krebsbach, M., Schiller, C., Roiger, A., Schlager, H., Dreiling, V., and Borrmann, S.: Observations of meteoric material and implications for aerosol nucleation in the winter Arctic lower stratosphere derived from in situ particle measurements, *Atmospheric Chemistry and Physics*, 5, 3053–3069, <https://doi.org/10.5194/acp-5-3053-2005>, 2005.
- Curtius, J., Heinritzi, M., Beck, L. J., Pöhlker, M. L., Tripathi, N., Krumm, B. E., Holzbeck, P., Nussbaumer, C. M., Hernández Pardo, L., Klimach, T., Barmounis, K., Andersen, S. T., Bardakov, R., Bohn, B., Cecchini, M. A., Chaboureaud, J.-P., Dauhut, T., Dienhart, D., Dörich, R., Edtbauer, A., Giez, A., Hartmann, A., Holanda, B. A., Joppe, P., Kaiser, K., Keber, T., Klebach, H., Krüger, O. O., Kürten, A., Mallaun, C., Marno, D., Martinez, M., Monteiro, C., Nelson, C., Ort, L., Raj, S. S., Richter, S., Ringsdorf, A., Rocha, F., Simon, M., Sree Kumar, S., Tsokankunku, A., Unfer, G. R., Valenti, I. D., Wang, N., Zahn, A., Zauner-Wieczorek, M., Albrecht, R. I., Andreae, M. O., Artaxo, P., Crowley, J. N., Fischer, H., Harder, H., Herdies, D. L., Machado, L. A. T., Pöhlker, C., Pöschl, U., Possner, A., Pozzer, A., Schneider, J., Williams, J., and Lelieveld, J.: Isoprene nitrates drive new particle formation in Amazon’s upper troposphere, *Nature*, 636, 124–130, <https://doi.org/10.1038/s41586-024-08192-4>, publisher: Nature Publishing Group, 2024.
- Daniels, L. D., Gray, R. W., and Burton, P. J.: 2017 megafires in British Columbia: Urgent need to adapt and improve resilience to wildfire, In: Hood, Sharon M.; Drury, Stacy; Steelman, Toddi; Steffens, Ron,[eds.]. *Proceedings of the Fire Continuum-Preparing for the future of wildland fire*; 2018 May 21-24; Missoula, MT. *Proceedings RMRS-P-78*. Fort Collins, CO: US Department of Agriculture, Forest Service, Rocky Mountain Research Station. p. 51-62., 78, 51–62, 2020.
- DeCarlo, P. F., Kimmel, J. R., Trimborn, A., Northway, M. J., Jayne, J. T., Aiken, A. C., Gonin, M., Fuhrer, K., Horvath, T., Docherty, K. S., Worsnop, D. R., and Jimenez, J. L.: Field-Deployable, High-Resolution, Time-of-Flight Aerosol Mass Spectrometer, *Analytical Chemistry*, 78, 8281–8289, <https://doi.org/10.1021/ac061249n>, PMID: 17165817, 2006.
- Dentener, F., Kinne, S., Bond, T., Boucher, O., Cofala, J., Generoso, S., Ginoux, P., Gong, S., Hoelzemann, J. J., Ito, A., Marelli, L., Penner, J. E., Putaud, J.-P., Textor, C., Schulz, M., van der Werf, G. R., and Wilson, J.: Emissions of primary aerosol and precursor gases in the years 2000 and 1750 prescribed data-sets for AeroCom, *Atmospheric Chemistry and Physics*, 6, 4321–4344, <https://doi.org/10.5194/acp-6-4321-2006>, 2006.

- Deshler, T., Hervig, M. E., Hofmann, D. J., Rosen, J. M., and Liley, J. B.: Thirty years of in situ stratospheric aerosol size distribution measurements from Laramie, Wyoming (41°N), using balloon-borne instruments, *Journal of Geophysical Research: Atmospheres*, 108, <https://doi.org/https://doi.org/10.1029/2002JD002514>, 2003.
- Dietmüller, S., Jöckel, P., Tost, H., Kunze, M., Gellhorn, C., Brinkop, S., Frömming, C., Ponater, M., Steil, B., Lauer, A., and Hendricks, J.: A new radiation infrastructure for the Modular Earth Submodel System (MESSy, based on version 2.51), *Geoscientific Model Development*, 9, 2209–2222, <https://doi.org/10.5194/gmd-9-2209-2016>, 2016.
- Doumbia, T., Granier, C., Elguindi, N., Bouarar, I., Darras, S., Brasseur, G., Gaubert, B., Liu, Y., Shi, X., Stavrakou, T., Tilmes, S., Lacey, F., Deroubaix, A., and Wang, T.: Changes in global air pollutant emissions during the COVID-19 pandemic: a dataset for atmospheric modeling, *Earth System Science Data*, 13, 4191–4206, <https://doi.org/10.5194/essd-13-4191-2021>, 2021.
- Dragoneas, A., Molleker, S., Appel, O., Hünig, A., Böttger, T., Hermann, M., Drewnick, F., Schneider, J., Weigel, R., and Borrmann, S.: The realization of autonomous, aircraft-based, real-time aerosol mass spectrometry in the upper troposphere and lower stratosphere, *Atmospheric Measurement Techniques*, 15, 5719–5742, <https://doi.org/10.5194/amt-15-5719-2022>, 2022.
- Drewnick, F., Hings, S. S., DeCarlo, P., Jayne, J. T., Gonin, M., Fuhrer, K., Weimer, S., Jimenez, J. L., Demerjian, K. L., Borrmann, S., and and, D. R. W.: A New Time-of-Flight Aerosol Mass Spectrometer (TOF-AMS)—Instrument Description and First Field Deployment, *Aerosol Science and Technology*, 39, 637–658, <https://doi.org/10.1080/02786820500182040>, 2005.
- Dunne, E. M., Gordon, H., Kürten, A., Almeida, J., Duplissy, J., Williamson, C., Ortega, I. K., Pringle, K. J., Adamov, A., Baltensperger, U., Barmet, P., Benduhn, F., Bianchi, F., Breitenlechner, M., Clarke, A., Curtius, J., Dommen, J., Donahue, N. M., Ehrhart, S., Flagan, R. C., Franchin, A., Guida, R., Hakala, J., Hansel, A., Heinritzi, M., Jokinen, T., Kangasluoma, J., Kirkby, J., Kulmala, M., Kupc, A., Lawler, M. J., Lehtipalo, K., Makhmutov, V., Mann, G., Mathot, S., Merikanto, J., Miettinen, P., Nenes, A., Onnela, A., Rap, A., Reddington, C. L. S., Riccobono, F., Richards, N. A. D., Rissanen, M. P., Rondo, L., Sarnela, N., Schobesberger, S., Sengupta, K., Simon, M., Sipilä, M., Smith, J. N., Stozkhov, Y., Tomé, A., Tröstl, J., Wagner, P. E., Wimmer, D., Winkler, P. M., Worsnop, D. R., and Carslaw, K. S.: Global atmospheric particle formation from CERN CLOUD measurements, *Science*, 354, 1119–1124, <https://doi.org/10.1126/science.aaf2649>, 2016.
- EANET: The Acid Deposition Monitoring Network in East Asia (EANET), <https://www.eanet.asia>, last access: 19 March 2025, 2025.
- Ebert, M., Weigel, R., Weinbruch, S., Schneider, L., Kandler, K., Lauterbach, S., Köllner, F., Plöger, F., Günther, G., Vogel, B., and Borrmann, S.: Characterization of refractory aerosol particles collected in the tropical upper troposphere–lower stratosphere (UTLS) within the Asian tropopause aerosol layer (ATAL), *Atmospheric Chemistry and Physics*, 24, 4771–4788, <https://doi.org/10.5194/acp-24-4771-2024>, 2024.
- Ehrhart, S., Dunne, E. M., Manninen, H. E., Nieminen, T., Lelieveld, J., and Pozzer, A.: Two new submodels for the Modular Earth Submodel System (MESSy): New Aerosol Nucleation (NAN) and small ions (IONS) version 1.0, *Geoscientific Model Development*, 11, 4987–5001, <https://doi.org/10.5194/gmd-11-4987-2018>, 2018.
- Elkins, J., Hints, E., and Moore, F.: ATom: Measurements from the UAS Chromatograph for Atmospheric Trace Species (UCATS), ORNL Distributed Active Archive Center [data set], <https://doi.org/10.3334/ORNLDAAAC/1750>, 2020.
- Fountoukis, C. and Nenes, A.: ISORROPIA II: a computationally efficient thermodynamic equilibrium model for $K^+ - Ca^{2+} - Mg^{2+} - NH_4^+ - Na^+ - SO_4^{2-} - NO_3^- - Cl^- - H_2O$ aerosols, *Atmospheric Chemistry and Physics*, 7, 4639–4659, <https://doi.org/10.5194/acp-7-4639-2007>, 2007.
- Froyd, K. D., Yu, P., Schill, G. P., Brock, C. A., Kupc, A., Williamson, C. J., Jensen, E. J., Ray, E., Rosenlof, K. H., Bian, H., et al.: Dominant role of mineral dust in cirrus cloud formation revealed by global-scale measurements, *Nature Geoscience*, 15, 177–183, <https://doi.org/10.1038/s41561-022-00901-w>, 2022.

- Gao, R. S., Schwarz, J. P., Kelly, K. K., Fahey, D. W., Watts, L. A., Thompson, T. L., Spackman, J. R., Slowik, J. G., Cross, E. S., Han, J.-H., Davidovits, P., Onasch, T. B., and and, D. R. W.: A Novel Method for Estimating Light-Scattering Properties of Soot Aerosols Using a Modified Single-Particle Soot Photometer, *Aerosol Science and Technology*, 41, 125–135, <https://doi.org/10.1080/02786820601118398>, 2007.
- 770 Gettelman, A., Hoor, P., Pan, L. L., Randel, W. J., Hegglin, M. I., and Birner, T.: THE EXTRATROPICAL UPPER TROPOSPHERE AND LOWER STRATOSPHERE, *Reviews of Geophysics*, 49, <https://doi.org/https://doi.org/10.1029/2011RG000355>, 2011.
- Giauque, W. F., Hornung, E. W., Kunzler, J. E., and Rubin, T. R.: The Thermodynamic Properties of Aqueous Sulfuric Acid Solutions and Hydrates from 15 to 300°K.1, *Journal of the American Chemical Society*, 82, 62–70, <https://doi.org/10.1021/ja01486a014>, 1960.
- Giorgetta, M. A., Manzini, E., Roeckner, E., Esch, M., and Bengtsson, L.: Climatology and Forcing of the Quasi-Biennial Oscillation in the MAECHAM5 Model, *Journal of Climate*, 19, 3882 – 3901, <https://doi.org/10.1175/JCLI3830.1>, 2006.
- 775 Gordon, H., Kirkby, J., Baltensperger, U., Bianchi, F., Breitenlechner, M., Curtius, J., Dias, A., Dommen, J., Donahue, N. M., Dunne, E. M., Duplissy, J., Ehrhart, S., Flagan, R. C., Frege, C., Fuchs, C., Hansel, A., Hoyle, C. R., Kulmala, M., Kürten, A., Lehtipalo, K., Makhmutov, V., Molteni, U., Rissanen, M. P., Stozhkov, Y., Tröstl, J., Tsagkogeorgas, G., Wagner, R., Williamson, C., Wimmer, D., Winkler, P. M., Yan, C., and Carslaw, K. S.: Causes and importance of new particle formation in the present-day and preindustrial atmospheres, *Journal of Geophysical Research: Atmospheres*, 122, 8739–8760, <https://doi.org/https://doi.org/10.1002/2017JD026844>, 2017.
- 780 Granier, C., Darras, S., Denier van der Gon, H., Doubalova, J., Elguindi, N., Galle, B., Gauss, M., Guevara, M., Jalkanen, J.-P., Kuenen, J., Lioussé, C., Quack, B., Simpson, D., and Sindelarova, K.: The Copernicus Atmosphere Monitoring Service global and regional emissions (April 2019 version), Copernicus Atmosphere Monitoring Service (CAMS) report, <https://doi.org/10.24380/d0bn-kx16>, 2019.
- Guelle, W., Schulz, M., Balkanski, Y., and Dentener, F.: Influence of the source formulation on modeling the atmospheric global distribution of sea salt aerosol, *Journal of Geophysical Research: Atmospheres*, 106, 27 509–27 524, <https://doi.org/10.1029/2001JD900249>, 2001.
- 785 Guenther, A. B., Jiang, X., Heald, C. L., Sakulyanontvittaya, T., Duhl, T., Emmons, L. K., and Wang, X.: The Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1): an extended and updated framework for modeling biogenic emissions, *Geoscientific Model Development*, 5, 1471–1492, <https://doi.org/10.5194/gmd-5-1471-2012>, 2012.
- He, X.-C., Tham, Y. J., Dada, L., Wang, M., Finkenzeller, H., Stolzenburg, D., Iyer, S., Simon, M., Kürten, A., Shen, J., Rörup, B., Rissanen, M., Schobesberger, S., Baalbaki, R., Wang, D. S., Koenig, T. K., Jokinen, T., Sarnela, N., Beck, L. J., Almeida, J., Amanatidis, S., Amorim, A., Ataei, F., Baccarini, A., Bertozzi, B., Bianchi, F., Brilke, S., Caudillo, L., Chen, D., Chiu, R., Chu, B., Dias, A., Ding, A., Dommen, J., Duplissy, J., Haddad, I. E., Carracedo, L. G., Granzin, M., Hansel, A., Heinritzi, M., Hofbauer, V., Junninen, H., Kangasluoma, J., Kemppainen, D., Kim, C., Kong, W., Krechmer, J. E., Kvashin, A., Laitinen, T., Lamkaddam, H., Lee, C. P., Lehtipalo, K., Leiminger, M., Li, Z., Makhmutov, V., Manninen, H. E., Marie, G., Marten, R., Mathot, S., Mauldin, R. L., Mentler, B., Möhler, O., Müller, T., Nie, W., Onnela, A., Petäjä, T., Pfeifer, J., Philippov, M., Ranjithkumar, A., Saiz-Lopez, A., Salma, I., Scholz, W., Schuchmann, S., Schulze, B., Steiner, G., Stozhkov, Y., Tauber, C., Tomé, A., Thakur, R. C., Väisänen, O., Vazquez-Pufleau, M., Wagner, A. C., Wang, Y., Weber, S. K., Winkler, P. M., Wu, Y., Xiao, M., Yan, C., Ye, Q., Ylisirniö, A., Zauner-Wieczorek, M., Zha, Q., Zhou, P., Flagan, R. C., Curtius, J., Baltensperger, U., Kulmala, M., Kerminen, V.-M., Kurtén, T., Donahue, N. M., Volkamer, R., Kirkby, J., Worsnop, D. R., and Sipilä, M.: Role of iodine oxoacids in atmospheric aerosol nucleation, *Science*, 371, 589–595, <https://doi.org/10.1126/science.abe0298>, 2021.
- 795 800 Healy, R. M., Wang, J. M., Sofowote, U., Su, Y., Debosz, J., Noble, M., Munoz, A., Jeong, C.-H., Hilker, N., Evans, G. J., and Doerksen, G.: Black carbon in the Lower Fraser Valley, British Columbia: Impact of 2017 wildfires on local air quality and aerosol optical properties, *Atmospheric Environment*, 217, 116 976, <https://doi.org/https://doi.org/10.1016/j.atmosenv.2019.116976>, 2019.

- Hersbach, H., Bell, B., Berrisford, P., Hirahara, S., Horányi, A., Muñoz-Sabater, J., Nicolas, J., Peubey, C., Radu, R., Schepers, D., Simmons, A., Soci, C., Abdalla, S., Abellan, X., Balsamo, G., Bechtold, P., Biavati, G., Bidlot, J., Bonavita, M., De Chiara, G., Dahlgren, P., Dee, D., Diamantakis, M., Dragani, R., Flemming, J., Forbes, R., Fuentes, M., Geer, A., Haimberger, L., Healy, S., Hogan, R. J., Hólm, E., Janisková, M., Keeley, S., Laloyaux, P., Lopez, P., Lupu, C., Radnoti, G., de Rosnay, P., Rozum, I., Vamborg, F., Villaume, S., and Thépaut, J.-N.: The ERA5 global reanalysis, *Quarterly Journal of the Royal Meteorological Society*, 146, 1999–2049, <https://doi.org/10.1002/qj.3803>, 2020.
- Hertzog, A. and Plougonven, R.: Strateole-2: High-resolution observations of the tropical tropopause layer with long-duration balloons, in: Egu general assembly conference abstracts, pp. EGU21–7109, <https://doi.org/10.5194/egusphere-egu21-7109>, 2021.
- Höpfner, M., Ungermann, J., Borrmann, S., Wagner, R., Spang, R., Riese, M., Stiller, G., Appel, O., Batenburg, A. M., Bucci, S., et al.: Ammonium nitrate particles formed in upper troposphere from ground ammonia sources during Asian monsoons, *Nature Geoscience*, 12, 608–612, <https://doi.org/10.1038/s41561-019-0385-8>, 2019.
- Hsu, N., Tsay, S.-C., King, M., and Herman, J.: Aerosol properties over bright-reflecting source regions, *IEEE Transactions on Geoscience and Remote Sensing*, 42, 557–569, <https://doi.org/10.1109/TGRS.2004.824067>, 2004.
- Hünig, A., Appel, O., Dragoneas, A., Molleker, S., Clemen, H.-C., Helleis, F., Klimach, T., Köllner, F., Böttger, T., Drewnick, F., Schneider, J., and Borrmann, S.: Design, characterization, and first field deployment of a novel aircraft-based aerosol mass spectrometer combining the laser ablation and flash vaporization techniques, *Atmospheric Measurement Techniques*, 15, 2889–2921, <https://doi.org/10.5194/amt-15-2889-2022>, 2022.
- Jeuken, A. B. M., Siegmund, P. C., Heijboer, L. C., Feichter, J., and Bengtsson, L.: On the potential of assimilating meteorological analyses in a global climate model for the purpose of model validation, *Journal of Geophysical Research: Atmospheres*, 101, 16 939–16 950, <https://doi.org/10.1029/96JD01218>, 1996.
- Jimenez, J., Campuzano-Jost, P., Day, D., Nault, B., Price, D., and Schroder, J.: ATom: L2 Measurements from CU High-Resolution Aerosol Mass Spectrometer (HR-AMS), ORNL Distributed Active Archive Center [data set], <https://doi.org/10.3334/ORNLDAAAC/1716>, 2019.
- Jöckel, P., Tost, H., Pozzer, A., Brühl, C., Buchholz, J., Ganzeveld, L., Hoor, P., Kerkweg, A., Lawrence, M. G., Sander, R., Steil, B., Stiller, G., Tanarhte, M., Taraborrelli, D., van Aardenne, J., and Lelieveld, J.: The atmospheric chemistry general circulation model ECHAM5/MESSy1: consistent simulation of ozone from the surface to the mesosphere, *Atmospheric Chemistry and Physics*, 6, 5067–5104, <https://doi.org/10.5194/acp-6-5067-2006>, 2006.
- Jöckel, P., Kerkweg, A., Pozzer, A., Sander, R., Tost, H., Riede, H., Baumgaertner, A., Gromov, S., and Kern, B.: Development cycle 2 of the Modular Earth Submodel System (MESSy2), *Geoscientific Model Development*, 3, 717–752, <https://doi.org/10.5194/gmd-3-717-2010>, 2010.
- Joppe, P., Schneider, J., Kaiser, K., Fischer, H., Hoor, P., Kunkel, D., Lachnitt, H.-C., Marsing, A., Röder, L., Schlager, H., Tomsche, L., Voigt, C., Zahn, A., and Borrmann, S.: The influence of extratropical cross-tropopause mixing on the correlation between ozone and sulfate aerosol in the lowermost stratosphere, *Atmospheric Chemistry and Physics*, 24, 7499–7522, <https://doi.org/10.5194/acp-24-7499-2024>, 2024.
- Junge, C. E., Chagnon, C. W., and Manson, J. E.: Stratospheric aerosols, *Journal of Meteorology*, 18, 81–108, 1961.
- Katich, J. M., Samset, B. H., Bui, T. P., Dollner, M., Froyd, K. D., Campuzano-Jost, P., Nault, B. A., Schroder, J. C., Weinzierl, B., and Schwarz, J. P.: Strong Contrast in Remote Black Carbon Aerosol Loadings Between the Atlantic and Pacific Basins, *Journal of Geophysical Research: Atmospheres*, 123, 13,386–13,395, <https://doi.org/https://doi.org/10.1029/2018JD029206>, 2018.

- 840 Kerkweg, A., Sander, R., Tost, H., and Jöckel, P.: Technical note: Implementation of prescribed (OFFLEM), calculated (ONLEM), and pseudo-emissions (TNUDGE) of chemical species in the Modular Earth Submodel System (MESSy), *Atmospheric Chemistry and Physics*, 6, 3603–3609, <https://doi.org/10.5194/acp-6-3603-2006>, 2006.
- Kirkby, J., Duplissy, J., Sengupta, K., Frege, C., Gordon, H., Williamson, C., Heinritzi, M., Simon, M., Yan, C., Almeida, J., et al.: Ion-induced nucleation of pure biogenic particles, *Nature*, 533, 521–526, <https://doi.org/10.1038/nature17953>, 2016.
- 845 Klingmüller, K., Metzger, S., Abdelkader, M., Karydis, V. A., Stenchikov, G. L., Pozzer, A., and Lelieveld, J.: Revised mineral dust emissions in the atmospheric chemistry–climate model EMAC (MESSy 2.52 DU_Astithal KKDU2017 patch), *Geoscientific Model Development*, 11, 989–1008, <https://doi.org/10.5194/gmd-11-989-2018>, 2018.
- Klobas, J. E., Wilmouth, D. M., Weisenstein, D. K., Anderson, J. G., and Salawitch, R. J.: Ozone depletion following future volcanic eruptions, *Geophysical Research Letters*, 44, 7490–7499, 2017.
- 850 Kloss, C., Sellitto, P., Legras, B., Vernier, J.-P., Jégou, F., Venkat Ratnam, M., Suneel Kumar, B., Lakshmi Madhavan, B., and Berthet, G.: Impact of the 2018 Ambae Eruption on the Global Stratospheric Aerosol Layer and Climate, *Journal of Geophysical Research: Atmospheres*, 125, e2020JD032410, <https://doi.org/https://doi.org/10.1029/2020JD032410>, 2020.
- Kohl, M., Lelieveld, J., Chowdhury, S., Ehrhart, S., Sharma, D., Cheng, Y., Tripathi, S. N., Sebastian, M., Pandithurai, G., Wang, H., and Pozzer, A.: Numerical simulation and evaluation of global ultrafine particle concentrations at the Earth’s surface, *Atmospheric Chemistry and Physics*, 23, 13 191–13 215, <https://doi.org/10.5194/acp-23-13191-2023>, 2023.
- 855 Kohl, M., Brühl, C., Schallack, J., Tost, H., Jöckel, P., Jost, A., Beirle, S., Höpfner, M., and Pozzer, A.: New submodel for emissions from Explosive Volcanic ERuptions (EVER v1.1) within the Modular Earth Submodel System (MESSy, version 2.55.1), *EGUsphere*, 2024, 1–35, <https://doi.org/10.5194/egusphere-2024-2200>, 2024.
- Kovilakam, M., Thomason, L. W., Ernest, N., Rieger, L., Bourassa, A., and Millán, L.: The Global Space-based Stratospheric Aerosol Climatology (version 2.0): 1979–2018, *Earth System Science Data*, 12, 2607–2634, <https://doi.org/10.5194/essd-12-2607-2020>, 2020.
- 860 Kovilakam, M., Thomason, L., and Knepp, T.: SAGE III/ISS aerosol/cloud categorization and its impact on GloSSAC, *Atmospheric Measurement Techniques*, 16, 2709–2731, <https://doi.org/10.5194/amt-16-2709-2023>, 2023.
- Kremser, S., Thomason, L. W., von Hobe, M., Hermann, M., Deshler, T., Timmreck, C., Toohey, M., Stenke, A., Schwarz, J. P., Weigel, R., Fueglistaler, S., Prata, F. J., Vernier, J.-P., Schlager, H., Barnes, J. E., Antuña-Marrero, J.-C., Fairlie, D., Palm, M., Mahieu, E., Notholt, J., Rex, M., Bingen, C., Vanhellemont, F., Bourassa, A., Plane, J. M. C., Klocke, D., Carn, S. A., Clarisse, L., Trickl, T., Neely, R., James, A. D., Rieger, L., Wilson, J. C., and Meland, B.: Stratospheric aerosol—Observations, processes, and impact on climate, *Reviews of Geophysics*, 54, 278–335, <https://doi.org/10.1002/2015RG000511>, 2016.
- 865 Kupc, A., Williamson, C., Wagner, N. L., Richardson, M., and Brock, C. A.: Modification, calibration, and performance of the Ultra-High Sensitivity Aerosol Spectrometer for particle size distribution and volatility measurements during the Atmospheric Tomography Mission (ATom) airborne campaign, *Atmospheric Measurement Techniques*, 11, 369–383, <https://doi.org/10.5194/amt-11-369-2018>, 2018.
- Lelieveld, J., Evans, J. S., Fnais, M., Giannadaki, D., and Pozzer, A.: The contribution of outdoor air pollution sources to premature mortality on a global scale, *Nature*, 525, 367–371, <https://doi.org/10.1038/nature15371>, 2015.
- Lohmann, U. and Feichter, J.: Global indirect aerosol effects: a review, *Atmospheric Chemistry and Physics*, 5, 715–737, <https://doi.org/10.5194/acp-5-715-2005>, 2005.
- 875 Lund, M. T., Samset, B. H., Skeie, R. B., Watson-Parris, D., Katich, J. M., Schwarz, J. P., and Weinzierl, B.: Short Black Carbon lifetime inferred from a global set of aircraft observations, *Npj Climate and Atmospheric Science*, 1, 31, <https://doi.org/10.1038/s41612-018-0040-x>, 2018.

- Luo, B.: stratospheric aerosol data for use in CCMi models, tp://iacftp.ethz.ch/pub_read/luo/ccmi/ [data set], last access: 11 April 2025, 2013.
- Ma, J., Brühl, C., He, Q., Steil, B., Karydis, V. A., Klingmüller, K., Tost, H., Chen, B., Jin, Y., Liu, N., Xu, X., Yan, P., Zhou, X., Abdelrahman, K., Pozzer, A., and Lelieveld, J.: Modeling the aerosol chemical composition of the tropopause over the Tibetan Plateau during the Asian summer monsoon, *Atmospheric Chemistry and Physics*, 19, 11 587–11 612, <https://doi.org/10.5194/acp-19-11587-2019>, 2019.
- 880 Maclean, A. M., Li, Y., Crescenzo, G. V., Smith, N. R., Karydis, V. A., Tsimpidi, A. P., Butenhoff, C. L., Faiola, C. L., Lelieveld, J., Nizkorodov, S. A., Shiraiwa, M., and Bertram, A. K.: Global Distribution of the Phase State and Mixing Times within Secondary Organic Aerosol Particles in the Troposphere Based on Room-Temperature Viscosity Measurements, *ACS Earth and Space Chemistry*, 5, 3458–3473, <https://doi.org/10.1021/acsearthspacechem.1c00296>, 2021.
- 885 Maheshwarkar, P., Cuesta, J., Formenti, P., and Lemmouchi, F.: Three-dimensional distribution of aerosols of multiple types at daily scale using TROPOMI spaceborne observations, *Science of The Total Environment*, 955, 177 037, <https://doi.org/10.1016/j.scitotenv.2024.177037>, 2024.
- McDuffie, E. E., Smith, S. J., O'Rourke, P., Tibrewal, K., Venkataraman, C., Marais, E. A., Zheng, B., Crippa, M., Brauer, M., and Martin, R. V.: A global anthropogenic emission inventory of atmospheric pollutants from sector- and fuel-specific sources (1970–2017): an application of the Community Emissions Data System (CEDS), *Earth System Science Data*, 12, 3413–3442, <https://doi.org/10.5194/essd-12-3413-2020>, 2020.
- McKain, K. and Sweeney, C.: ATom: CO₂, CH₄, and CO Measurements from Picarro, 2016–2018, ORNL Distributed Active Archive Center [data set], <https://doi.org/10.3334/ORNLDAAC/1732>, 2021.
- 895 Mishra, A. K., Koren, I., and Rudich, Y.: Effect of aerosol vertical distribution on aerosol-radiation interaction: A theoretical prospect, *Heliyon*, 1, <https://doi.org/10.1016/j.heliyon.2015.e00036>, publisher: Elsevier, 2015.
- Murphy, D. M., Thomson, D. S., and Mahoney, M. J.: In Situ Measurements of Organics, Meteoritic Material, Mercury, and Other Elements in Aerosols at 5 to 19 Kilometers, *Science*, 282, 1664–1669, <https://doi.org/10.1126/science.282.5394.1664>, 1998.
- Murphy, D. M., Cziczo, D. J., Hudson, P. K., and Thomson, D. S.: Carbonaceous material in aerosol particles in the lower stratosphere and tropopause region, *Journal of Geophysical Research: Atmospheres*, 112, <https://doi.org/10.1029/2006JD007297>, 2007.
- 900 Murphy, D. M., Abou-Ghanem, M., Cziczo, D. J., Froyd, K. D., Jacquot, J., Lawler, M. J., Maloney, C., Plane, J. M. C., Ross, M. N., Schill, G. P., and Shen, X.: Metals from spacecraft reentry in stratospheric aerosol particles, *Proceedings of the National Academy of Sciences*, 120, e2313374 120, <https://doi.org/10.1073/pnas.2313374120>, 2023.
- Murray, C. J., Aravkin, A. Y., Zheng, P., Abbafati, C., Abbas, K. M., Abbasi-Kangevari, M., Abd-Allah, F., Abdelalim, A., Abdollahi, M., Abdollahpour, I., et al.: Global burden of 87 risk factors in 204 countries and territories, 1990–2019: a systematic analysis for the Global Burden of Disease Study 2019, *The lancet*, 396, 1223–1249, [https://doi.org/10.1016/S0140-6736\(20\)30752-2](https://doi.org/10.1016/S0140-6736(20)30752-2), 2020.
- 905 Nault, B. A., Jo, D. S., McDonald, B. C., Campuzano-Jost, P., Day, D. A., Hu, W., Schroder, J. C., Allan, J., Blake, D. R., Canagaratna, M. R., Coe, H., Coggon, M. M., DeCarlo, P. F., Diskin, G. S., Dunmore, R., Flocke, F., Fried, A., Gilman, J. B., Gkatzelis, G., Hamilton, J. F., Hanisco, T. F., Hayes, P. L., Henze, D. K., Hodzic, A., Hopkins, J., Hu, M., Huey, L. G., Jobson, B. T., Kuster, W. C., Lewis, A., Li, M., Liao, J., Nawaz, M. O., Pollack, I. B., Peischl, J., Rappenglück, B., Reeves, C. E., Richter, D., Roberts, J. M., Ryerson, T. B., Shao, M., Sommers, J. M., Walega, J., Warneke, C., Weibring, P., Wolfe, G. M., Young, D. E., Yuan, B., Zhang, Q., de Gouw, J. A., and Jimenez, J. L.: Secondary organic aerosols from anthropogenic volatile organic compounds contribute substantially to air pollution mortality, *Atmospheric Chemistry and Physics*, 21, 11 201–11 224, <https://doi.org/10.5194/acp-21-11201-2021>, 2021.
- Platnick, S.: MODIS Atmosphere L3 Daily Product, NASA MODIS Adaptive Processing System, Goddard Space Flight Center, USA [data set], https://doi.org/10.5067/MODIS/MOD08_D3.061, 2015.
- 915

- Pöschl, U., von Kuhlmann, R., Poisson, N., and Crutzen, P. J.: Development and intercomparison of condensed isoprene oxidation mechanisms for global atmospheric modeling, *Journal of Atmospheric Chemistry*, 37, 29–52, <https://doi.org/10.1023/A:1006391009798>, 2000.
- Pozzer, A., de Meij, A., Pringle, K. J., Tost, H., Doering, U. M., van Aardenne, J., and Lelieveld, J.: Distributions and regional budgets of aerosols and their precursors simulated with the EMAC chemistry-climate model, *Atmospheric Chemistry and Physics*, 12, 961–987, <https://doi.org/10.5194/acp-12-961-2012>, 2012.
- Pozzer, A., Reifenberg, S. F., Kumar, V., Franco, B., Kohl, M., Taraborrelli, D., Gromov, S., Ehrhart, S., Jöckel, P., Sander, R., Fall, V., Rosanka, S., Karydis, V., Akritidis, D., Emmerichs, T., Crippa, M., Guizzardi, D., Kaiser, J. W., Clarisse, L., Kiendler-Scharr, A., Tost, H., and Tsimpidi, A.: Simulation of organics in the atmosphere: evaluation of EMACv2.54 with the Mainz Organic Mechanism (MOM) coupled to the ORACLE (v1.0) submodel, *Geoscientific Model Development*, 15, 2673–2710, <https://doi.org/10.5194/gmd-15-2673-2022>, 2022.
- Pringle, K. J., Tost, H., Metzger, S., Steil, B., Giannadaki, D., Nenes, A., Fountoukis, C., Stier, P., Vignati, E., and Lelieveld, J.: Description and evaluation of GMXe: a new aerosol submodel for global simulations (v1), *Geosci. Model Dev.*, 3, 391–412, <https://doi.org/10.5194/gmd-3-391-2010>, 2010.
- Putaud, J.-P., Raes, F., Van Dingenen, R., Brüggemann, E., Facchini, M.-C., Decesari, S., Fuzzi, S., Gehrig, R., Hüglin, C., Laj, P., Lorbeer, G., Maenhaut, W., Mihalopoulos, N., Müller, K., Querol, X., Rodriguez, S., Schneider, J., Spindler, G., ten Brink, H., Tørseth, K., and Wiedensohler, A.: A European aerosol phenomenology—2: chemical characteristics of particulate matter at kerbside, urban, rural and background sites in Europe, *Atmospheric Environment*, 38, 2579–2595, <https://doi.org/https://doi.org/10.1016/j.atmosenv.2004.01.041>, 2004.
- Pöschl, U.: Atmospheric Aerosols: Composition, Transformation, Climate and Health Effects, *Angewandte Chemie International Edition*, 44, 7520–7540, <https://doi.org/10.1002/anie.200501122>, 2005.
- Reddington, C. L., Carslaw, K. S., Stier, P., Schutgens, N., Coe, H., Liu, D., Allan, J., Browse, J., Pringle, K. J., Lee, L. A., Yoshioka, M., Johnson, J. S., Regayre, L. A., Spracklen, D. V., Mann, G. W., Clarke, A., Hermann, M., Henning, S., Wex, H., Kristensen, T. B., Leaitch, W. R., Pöschl, U., Rose, D., Andreae, M. O., Schmale, J., Kondo, Y., Oshima, N., Schwarz, J. P., Nenes, A., Anderson, B., Roberts, G. C., Snider, J. R., Leck, C., Quinn, P. K., Chi, X., Ding, A., Jimenez, J. L., and Zhang, Q.: The Global Aerosol Synthesis and Science Project (GASSP): Measurements and Modeling to Reduce Uncertainty, *Bulletin of the American Meteorological Society*, 98, 1857 – 1877, <https://doi.org/10.1175/BAMS-D-15-00317.1>, 2017.
- Rieger, L. A., Zawada, D. J., Bourassa, A. E., and Degenstein, D. A.: A Multiwavelength Retrieval Approach for Improved OSIRIS Aerosol Extinction Retrievals, *Journal of Geophysical Research: Atmospheres*, 124, 7286–7307, <https://doi.org/10.1029/2018JD029897>, 2019.
- Roeckner, E., Bäuml, G., Bonaventura, L., Brokopf, R., Esch, M., Giorgetta, M., Hagemann, S., Kirchner, I., Kornbluh, L., Manzini, E., et al.: The atmospheric general circulation model ECHAM 5. PART I: Model description, Tech. rep., Max-Planck-Institut für Meteorologie, <https://doi.org/10.17617/2.995269>, 2003.
- Ryerson, T., Thompson, C., Peischl, J., and Bourgeois, I.: ATom: L2 In Situ Measurements from NOAA Nitrogen Oxides and Ozone (NOyO3) Instrument, ORNL Distributed Active Archive Center [data set], <https://doi.org/10.3334/ORNLDAAAC/1734>, 2019.
- Sander, R., Baumgaertner, A., Cabrera-Perez, D., Frank, F., Gromov, S., Grooß, J.-U., Harder, H., Huijnen, V., Jöckel, P., Karydis, V. A., Niemeyer, K. E., Pozzer, A., Riede, H., Schultz, M. G., Taraborrelli, D., and Tauer, S.: The community atmospheric chemistry box model CAABA/MECCA-4.0, *Geoscientific Model Development*, 12, 1365–1385, <https://doi.org/10.5194/gmd-12-1365-2019>, 2019.

- Schallock, J., Brühl, C., Bingen, C., Höpfner, M., Rieger, L., and Lelieveld, J.: Reconstructing volcanic radiative forcing since 1990, using a comprehensive emission inventory and spatially resolved sulfur injections from satellite data in a chemistry-climate model, *Atmospheric Chemistry and Physics*, 23, 1169–1207, <https://doi.org/10.5194/acp-23-1169-2023>, 2023.
- 955 Schill, G. P., Froyd, K. D., Bian, H., Kupe, A., Williamson, C., Brock, C. A., Ray, E., Hornbrook, R. S., Hills, A. J., Apel, E. C., Chin, M., Colarco, P. R., and Murphy, D. M.: Widespread biomass burning smoke throughout the remote troposphere, *Nature Geoscience*, 13, 422–427, <https://doi.org/10.1038/s41561-020-0586-1>, publisher: Nature Publishing Group, 2020.
- Schmidt, A., Mills, M. J., Ghan, S., Gregory, J. M., Allan, R. P., Andrews, T., Bardeen, C. G., Conley, A., Forster, P. M., Gettelman, A., Portmann, R. W., Solomon, S., and Toon, O. B.: Volcanic Radiative Forcing From 1979 to 2015, *Journal of Geophysical Research: Atmospheres*, 123, 12 491–12 508, <https://doi.org/10.1029/2018JD028776>, 2018.
- 960 Schneider, J., Weigel, R., Klimach, T., Dragoneas, A., Appel, O., Hünig, A., Molleker, S., Köllner, F., Clemen, H.-C., Eppers, O., Hoppe, P., Hoor, P., Mahnke, C., Krämer, M., Rolf, C., Grooß, J.-U., Zahn, A., Obersteiner, F., Ravegnani, F., Ulanovsky, A., Schlager, H., Scheibe, M., Diskin, G. S., DiGangi, J. P., Nowak, J. B., Zöger, M., and Borrmann, S.: Aircraft-based observation of meteoric material in lower-stratospheric aerosol particles between 15 and 68° N, *Atmospheric Chemistry and Physics*, 21, 989–1013, [https://doi.org/10.5194/acp-21-](https://doi.org/10.5194/acp-21-989-2021)
- 965 989-2021, 2021.
- Schulz, C., Schneider, J., Amorim Holanda, B., Appel, O., Costa, A., de Sá, S. S., Dreiling, V., Fütterer, D., Jurkat-Witschas, T., Klimach, T., Knote, C., Krämer, M., Martin, S. T., Mertes, S., Pöhlker, M. L., Sauer, D., Voigt, C., Walser, A., Weinzierl, B., Ziereis, H., Zöger, M., Andreae, M. O., Artaxo, P., Machado, L. A. T., Pöschl, U., Wendisch, M., and Borrmann, S.: Aircraft-based observations of isoprene-epoxydiol-derived secondary organic aerosol (IEPOX-SOA) in the tropical upper troposphere over the Amazon region, *Atmospheric Chemistry and Physics*, 18, 14 979–15 001, <https://doi.org/10.5194/acp-18-14979-2018>, 2018.
- 970 Schwarz, J. and Katich, J.: ATom: L2 In Situ Measurements from Single Particle Soot Photometer (SP2), ORNL Distributed Active Archive Center [data set], <https://doi.org/10.3334/ORNLDAAAC/1672>, 2019.
- Schwarz, J. P., Spackman, J. R., Gao, R. S., Perring, A. E., Cross, E., Onasch, T. B., Ahern, A., Wrobel, W., Davidovits, P., Olfert, J., Dubey, M. K., Mazzoleni, C., and and, D. W. F.: The Detection Efficiency of the Single Particle Soot Photometer, *Aerosol Science and Technology*, 44, 612–628, <https://doi.org/10.1080/02786826.2010.481298>, 2010.
- 975 Shen, J., Russell, D. M., DeVivo, J., Kunkler, F., Baalbaki, R., Mentler, B., Scholz, W., Yu, W., Caudillo-Plath, L., Sommer, E., et al.: New particle formation from isoprene under upper-tropospheric conditions, *Nature*, 636, 115–123, [https://doi.org/10.1038/s41586-024-08196-](https://doi.org/10.1038/s41586-024-08196-0)
- 0, 2024a.
- Shen, S., Li, C., van Donkelaar, A., Jacobs, N., Wang, C., and Martin, R. V.: Enhancing Global Estimation of Fine Particulate Matter Concentrations by Including Geophysical a Priori Information in Deep Learning, *ACS ES&T Air*, 1, 332–345, <https://doi.org/10.1021/acsestair.3c00054>, 2024b.
- 980 Szopa, S., Naik, V., Adhikary, B., Artaxo, P., Bernsten, T., Collins, W., Fuzzi, S., Gallardo, L., Kiendler-Scharr, A., Klimont, Z., Liao, H., Unger, N., and Zanis, P.: Short-Lived Climate Forcers, p. 817–922, Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, <https://doi.org/10.1017/9781009157896.008>, 2021.
- 985 Tabazadeh, A., Toon, O. B., Clegg, S. L., and Hamill, P.: A new parameterization of H₂SO₄/H₂O aerosol composition: Atmospheric implications, *Geophysical Research Letters*, 24, 1931–1934, <https://doi.org/https://doi.org/10.1029/97GL01879>, 1997.
- Tanré, D., Geleyn, J., and Slingo, J.: First results of the introduction of an advanced aerosol-radiation interaction in the ECMWF low resolution global model, *Aerosols and their climatic effects*, 133, 177, 1984.

- Thomason, L. W., Ernest, N., Millán, L., Rieger, L., Bourassa, A., Vernier, J.-P., Manney, G., Luo, B., Arfeuille, F., and Peter, T.: A global
990 space-based stratospheric aerosol climatology: 1979–2016, *Earth System Science Data*, 10, 469–492, <https://doi.org/10.5194/essd-10-469-2018>, 2018.
- Thompson, C. R., Wofsy, S. C., Prather, M. J., Newman, P. A., Hanisco, T. F., Ryerson, T. B., Fahey, D. W., Apel, E. C., Brock, C. A.,
Brune, W. H., Froyd, K., Katich, J. M., Nicely, J. M., Peischl, J., Ray, E., Veres, P. R., Wang, S., Allen, H. M., Asher, E., Bian, H., Blake,
D., Bourgeois, I., Budney, J., Bui, T. P., Butler, A., Campuzano-Jost, P., Chang, C., Chin, M., Commane, R., Correa, G., Crounse, J. D.,
995 Daube, B., Dibb, J. E., DiGangi, J. P., Diskin, G. S., Dollner, M., Elkins, J. W., Fiore, A. M., Flynn, C. M., Guo, H., Hall, S. R., Hannun,
R. A., Hills, A., Hints, E. J., Hodzic, A., Hornbrook, R. S., Huey, L. G., Jimenez, J. L., Keeling, R. F., Kim, M. J., Kupc, A., Lacey,
F., Lait, L. R., Lamarque, J.-F., Liu, J., McKain, K., Meinardi, S., Miller, D. O., Montzka, S. A., Moore, F. L., Morgan, E. J., Murphy,
D. M., Murray, L. T., Nault, B. A., Neuman, J. A., Nguyen, L., Gonzalez, Y., Rollins, A., Rosenlof, K., Sargent, M., Schill, G., Schwarz,
J. P., Clair, J. M. S., Steenrod, S. D., Stephens, B. B., Strahan, S. E., Strode, S. A., Sweeney, C., Thames, A. B., Ullmann, K., Wagner,
1000 N., Weber, R., Weinzierl, B., Wennberg, P. O., Williamson, C. J., Wolfe, G. M., and Zeng, L.: The NASA Atmospheric Tomography
(ATom) Mission: Imaging the Chemistry of the Global Atmosphere, *Bulletin of the American Meteorological Society*, 103, E761 – E790,
<https://doi.org/10.1175/BAMS-D-20-0315.1>, 2022.
- Tost, H., Jöckel, P., Kerkweg, A., Sander, R., and Lelieveld, J.: Technical note: A new comprehensive SCAVenging submodel for global
atmospheric chemistry modelling, *Atmospheric Chemistry and Physics*, 6, 565–574, <https://doi.org/10.5194/acp-6-565-2006>, 2006.
- 1005 Tsimpidi, A. P., Karydis, V. A., Pozzer, A., Pandis, S. N., and Lelieveld, J.: ORACLE (v1.0): module to simulate the organic aerosol
composition and evolution in the atmosphere, *Geoscientific Model Development*, 7, 3153–3172, <https://doi.org/10.5194/gmd-7-3153-2014>, 2014.
- Tsimpidi, A. P., Karydis, V. A., Pandis, S. N., and Lelieveld, J.: Global-scale combustion sources of organic aerosols: sensitivity to formation
and removal mechanisms, *Atmospheric Chemistry and Physics*, 17, 7345–7364, <https://doi.org/10.5194/acp-17-7345-2017>, 2017.
- 1010 Tsimpidi, A. P., Scholz, S. M. C., Milousis, A., Mihalopoulos, N., and Karydis, V. A.: Aerosol Composition Trends during
2000–2020: In depth insights from model predictions and multiple worldwide observation datasets, *EGUsphere*, 2024, 1–66,
<https://doi.org/10.5194/egusphere-2024-3590>, 2024.
- van Donkelaar, A., Martin, R. V., Brauer, M., Hsu, N. C., Kahn, R. A., Levy, R. C., Lyapustin, A., Sayer, A. M., and Winker, D. M.: Global
Estimates of Fine Particulate Matter using a Combined Geophysical-Statistical Method with Information from Satellites, Models, and
1015 Monitors, *Environmental Science & Technology*, 50, 3762–3772, <https://doi.org/10.1021/acs.est.5b05833>, PMID: 26953851, 2016.
- Vernier, J. P., Pommereau, J. P., Garnier, A., Pelon, J., Larsen, N., Nielsen, J., Christensen, T., Cairo, F., Thomason, L. W., Leblanc, T., and
McDermid, I. S.: Tropical stratospheric aerosol layer from CALIPSO lidar observations, *Journal of Geophysical Research: Atmospheres*,
114, <https://doi.org/https://doi.org/10.1029/2009JD011946>, 2009.
- Vernier, J.-P., Thomason, L. W., and Kar, J.: CALIPSO detection of an Asian tropopause aerosol layer, *Geophysical Research Letters*, 38,
1020 <https://doi.org/https://doi.org/10.1029/2010GL046614>, 2011.
- Voigt, C., Lelieveld, J., Schlager, H., Schneider, J., Curtius, J., Meerkötter, R., Sauer, D., Bugliaro, L., Bohn, B., Crowley, J. N., Erbertseder,
T., Groß, S., Hahn, V., Li, Q., Mertens, M., Pöhlker, M. L., Pozzer, A., Schumann, U., Tomsche, L., Williams, J., Zahn, A., Andreae, M.,
Borrmann, S., Brüner, T., Dörich, R., Dörnbrack, A., Edtbauer, A., Ernle, L., Fischer, H., Giez, A., Granzin, M., Grewe, V., Harder, H.,
Heinritzi, M., Holanda, B. A., Jöckel, P., Kaiser, K., Krüger, O. O., Lucke, J., Marsing, A., Martin, A., Matthes, S., Pöhlker, C., Pöschl, U.,
1025 Reifenberg, S., Ringsdorf, A., Scheibe, M., Tadic, I., Zauner-Wieczorek, M., Henke, R., and Rapp, M.: Cleaner Skies during the COVID-
19 Lockdown, *Bulletin of the American Meteorological Society*, 103, E1796 – E1827, <https://doi.org/10.1175/BAMS-D-21-0012.1>, 2022.

- Wang, M., Xiao, M., Bertozzi, B., Marie, G., Rörup, B., Schulze, B., Bardakov, R., He, X.-C., Shen, J., Scholz, W., Marten, R., Dada, L., Baalbaki, R., Lopez, B., Lamkaddam, H., Manninen, H. E., Amorim, A., Ataei, F., Bogert, P., Brasseur, Z., Caudillo, L., De Menezes, L.-P., Duplissy, J., Ekman, A. M. L., Finkenzeller, H., Carracedo, L. G., Granzin, M., Guida, R., Heinritzi, M., Hofbauer, V., Höhler, K., Korhonen, K., Krechmer, J. E., Kürten, A., Lehtipalo, K., Mahfouz, N. G. A., Makhmutov, V., Massabò, D., Mathot, S., Mauldin, R. L., Mentler, B., Müller, T., Onnela, A., Petäjä, T., Philippov, M., Piedehierro, A. A., Pozzer, A., Ranjithkumar, A., Schervish, M., Schobesberger, S., Simon, M., Stozhkov, Y., Tomé, A., Umo, N. S., Vogel, F., Wagner, R., Wang, D. S., Weber, S. K., Welti, A., Wu, Y., Zauner-Wieczorek, M., Sipilä, M., Winkler, P. M., Hansel, A., Baltensperger, U., Kulmala, M., Flagan, R. C., Curtius, J., Riipinen, I., Gordon, H., Lelieveld, J., El-Haddad, I., Volkamer, R., Worsnop, D. R., Christoudias, T., Kirkby, J., Möhler, O., and Donahue, N. M.: Synergistic HNO₃-H₂SO₄-NH₃ upper tropospheric particle formation, *Nature*, 605, 483–489, <https://doi.org/10.1038/s41586-022-04605-4>, publisher: Nature Publishing Group, 2022.
- Watson-Parris, D., Schutgens, N., Reddington, C., Pringle, K. J., Liu, D., Allan, J. D., Coe, H., Carslaw, K. S., and Stier, P.: In situ constraints on the vertical distribution of global aerosol, *Atmospheric Chemistry and Physics*, 19, 11 765–11 790, <https://doi.org/10.5194/acp-19-11765-2019>, 2019.
- Weigel, R., Mahnke, C., Baumgartner, M., Dragoneas, A., Vogel, B., Ploeger, F., Viciani, S., D’Amato, F., Bucci, S., Legras, B., Luo, B., and Borrmann, S.: In situ observation of new particle formation (NPF) in the tropical tropopause layer of the 2017 Asian monsoon anticyclone – Part 1: Summary of StratoClim results, *Atmospheric Chemistry and Physics*, 21, 11 689–11 722, <https://doi.org/10.5194/acp-21-11689-2021>, 2021.
- Williamson, C. J., Kupc, A., Axisa, D., Bilsback, K. R., Bui, T., Campuzano-Jost, P., Dollner, M., Froyd, K. D., Hodshire, A. L., Jimenez, J. L., Kodros, J. K., Luo, G., Murphy, D. M., Nault, B. A., Ray, E. A., Weinzierl, B., Wilson, J. C., Yu, F., Yu, P., Pierce, J. R., and Brock, C. A.: A large source of cloud condensation nuclei from new particle formation in the tropics, *Nature*, 574, 399–403, <https://doi.org/10.1038/s41586-019-1638-9>, publisher: Nature Publishing Group, 2019.
- Winker, D. M., Vaughan, M. A., Omar, A., Hu, Y., Powell, K. A., Liu, Z., Hunt, W. H., and Young, S. A.: Overview of the CALIPSO Mission and CALIOP Data Processing Algorithms, *Journal of Atmospheric and Oceanic Technology*, 26, 2310 – 2323, <https://doi.org/10.1175/2009JTECHA1281.1>, 2009.
- Winker, D. M., Tackett, J. L., Getzewich, B. J., Liu, Z., Vaughan, M. A., and Rogers, R. R.: The global 3-D distribution of tropospheric aerosols as characterized by CALIOP, *Atmospheric Chemistry and Physics*, 13, 3345–3361, <https://doi.org/10.5194/acp-13-3345-2013>, publisher: Copernicus GmbH, 2013.
- Xenofontos, C., Kohl, M., Ruhl, S., Almeida, J., Beckmann, H. M., Caudillo-Plath, L., Ehrhart, S., Höhler, K., Kaniyodical Sebastian, M., Kong, W., et al.: The impact of ammonia on particle formation in the Asian Tropopause Aerosol Layer, *npj Climate and Atmospheric Science*, 7, 215, <https://doi.org/10.1038/s41612-024-00758-3>, 2024.
- Zhang, Q., Jimenez, J. L., Canagaratna, M. R., Allan, J. D., Coe, H., Ulbrich, I., Alfarra, M. R., Takami, A., Middlebrook, A. M., Sun, Y. L., Dzepina, K., Dunlea, E., Docherty, K., DeCarlo, P. F., Salcedo, D., Onasch, T., Jayne, J. T., Miyoshi, T., Shimono, A., Hatakeyama, S., Takegawa, N., Kondo, Y., Schneider, J., Drewnick, F., Borrmann, S., Weimer, S., Demerjian, K., Williams, P., Bower, K., Bahreini, R., Cottrell, L., Griffin, R. J., Rautiainen, J., Sun, J. Y., Zhang, Y. M., and Worsnop, D. R.: Ubiquity and dominance of oxygenated species in organic aerosols in anthropogenically-influenced Northern Hemisphere midlatitudes, *Geophysical Research Letters*, 34, <https://doi.org/https://doi.org/10.1029/2007GL029979>, 2007.

- 1065 Zhang, R., Xie, H.-B., Ma, F., Yin, R., Chen, J., and He, X.-C.: Quaternary Nucleation of Iodine and Sulfur Oxoacids in the Marine Atmosphere: Unexpected Role of Methanesulfonic Acid, *Journal of Geophysical Research: Atmospheres*, 130, e2024JD042220, <https://doi.org/https://doi.org/10.1029/2024JD042220>, 2025.
- Zhu, H., Martin, R. V., van Donkelaar, A., Hammer, M. S., Li, C., Meng, J., Oxford, C. R., Liu, X., Li, Y., Zhang, D., Singh, I., and Lyapustin, A.: Importance of aerosol composition and aerosol vertical profiles in global spatial variation in the relationship between $PM_{2.5}$ and aerosol optical depth, *Atmospheric Chemistry and Physics*, 24, 11 565–11 584, <https://doi.org/10.5194/acp-24-11565-2024>, 2024.

5.1 Supplementary Information

1 Supplementary Figures

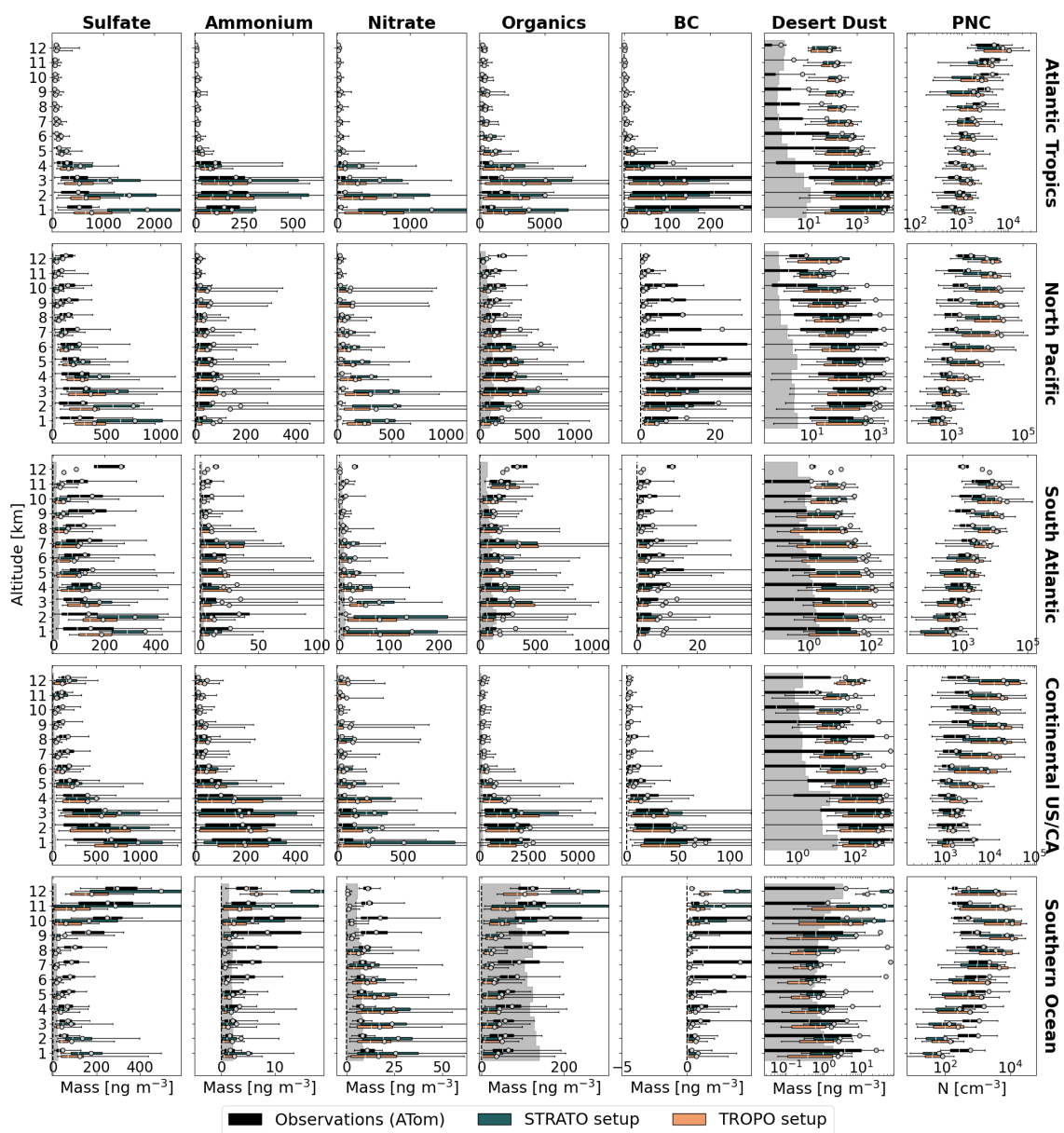


Figure S1. As Fig. 4 from the manuscript, but with simulated values from the STRATO and the TROPO setup.

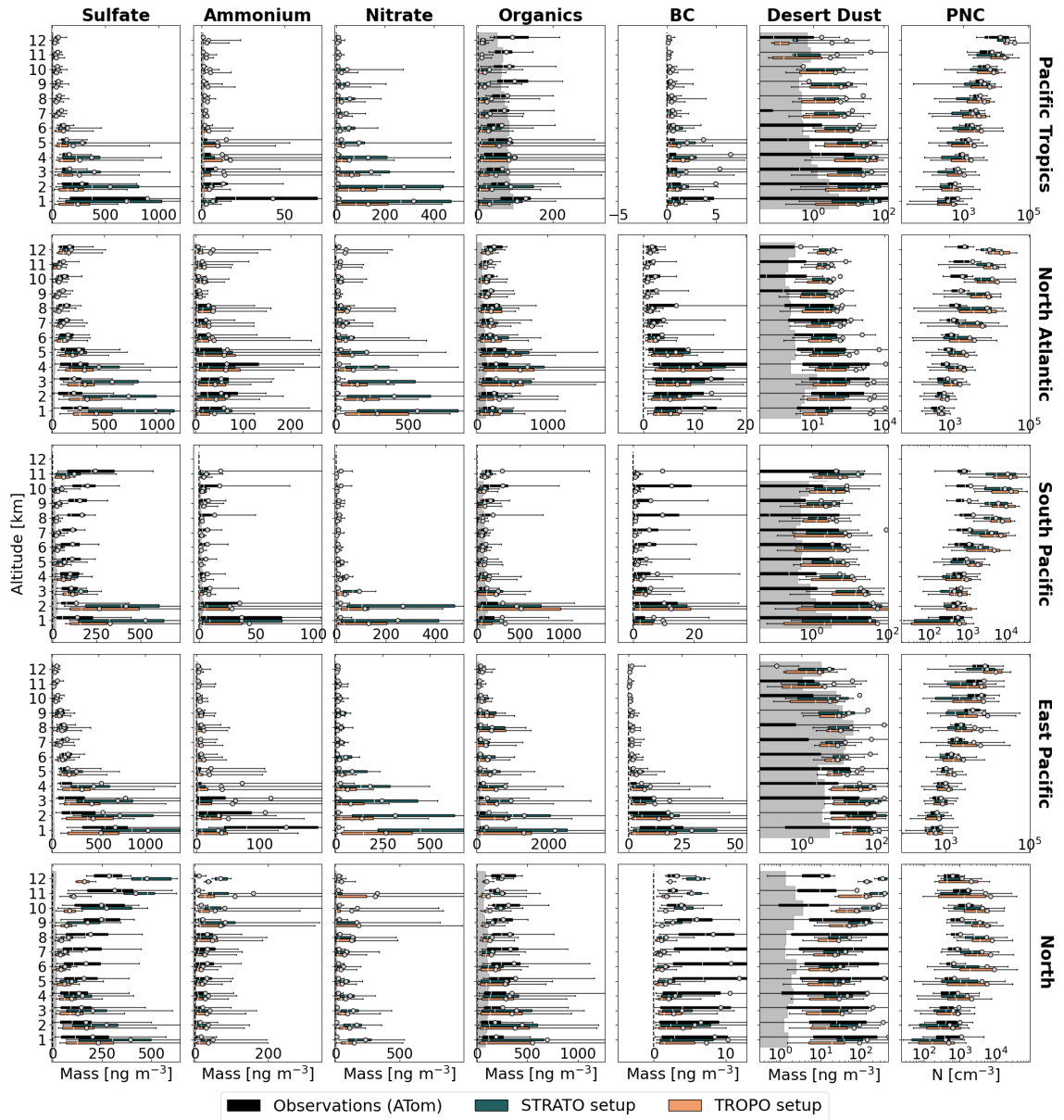


Figure S2. As Fig. 4 from the manuscript, but with simulated values from the STRATO and the TROPO setup.

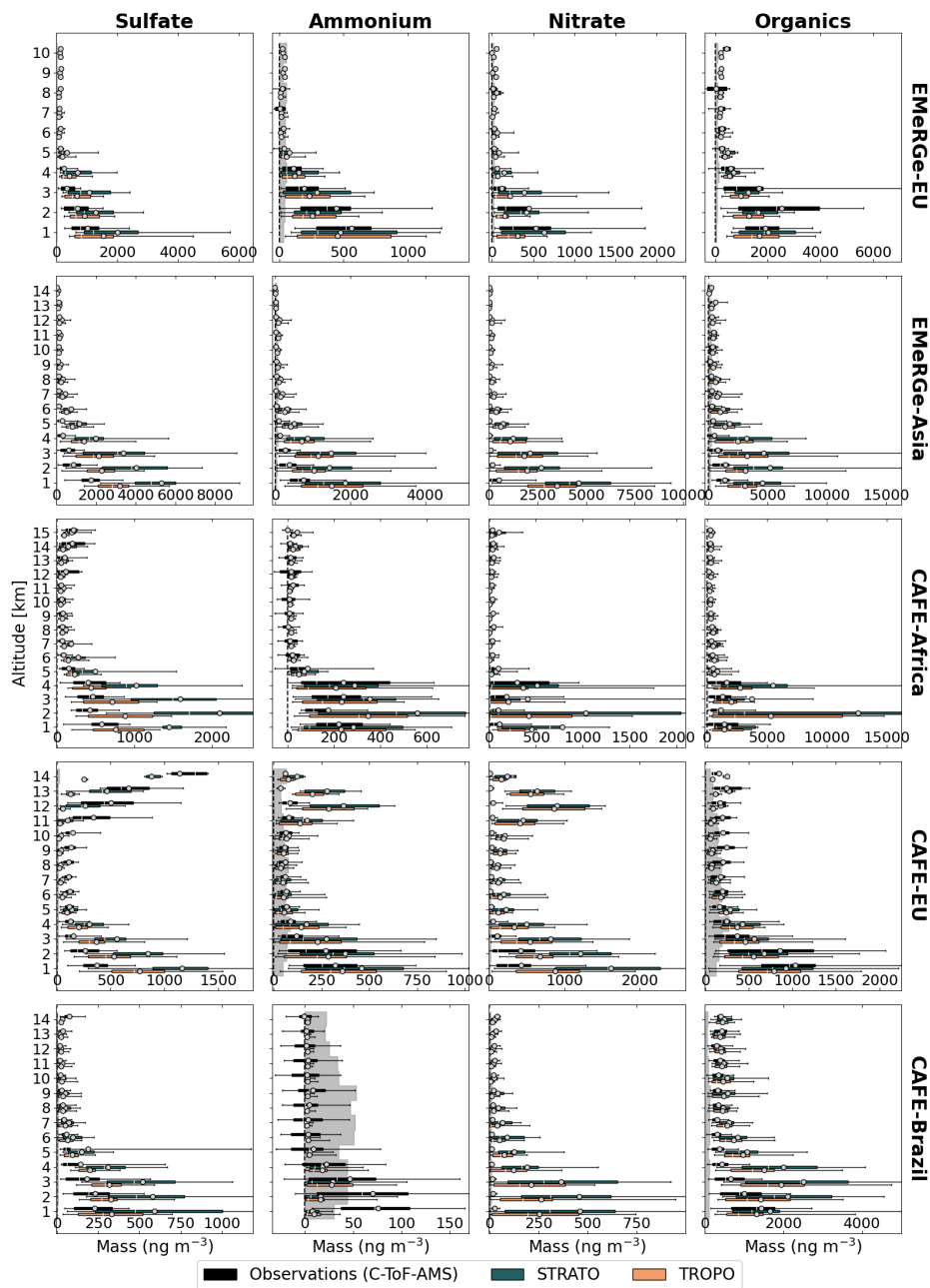


Figure S3. As Fig. 5 from the manuscript, but with simulated values from the STRATO and the TROPO setup.

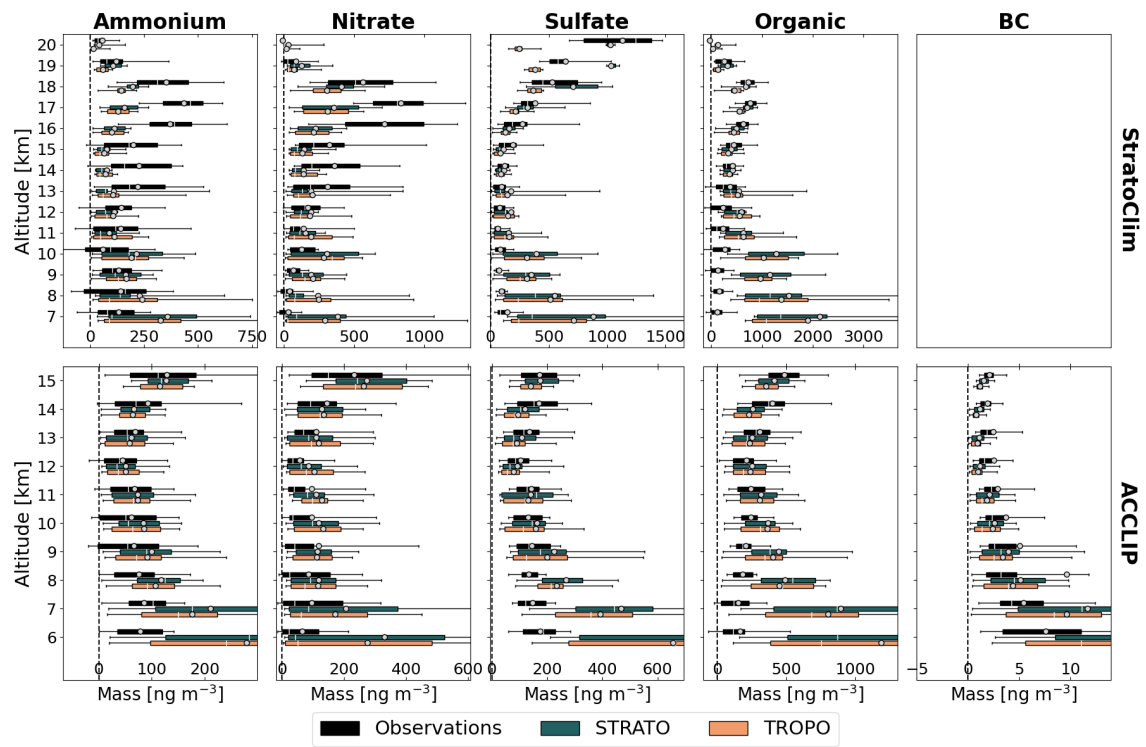


Figure S4. As Fig. 6 from the manuscript, but with simulated values from the STRATO and the TROPO setup

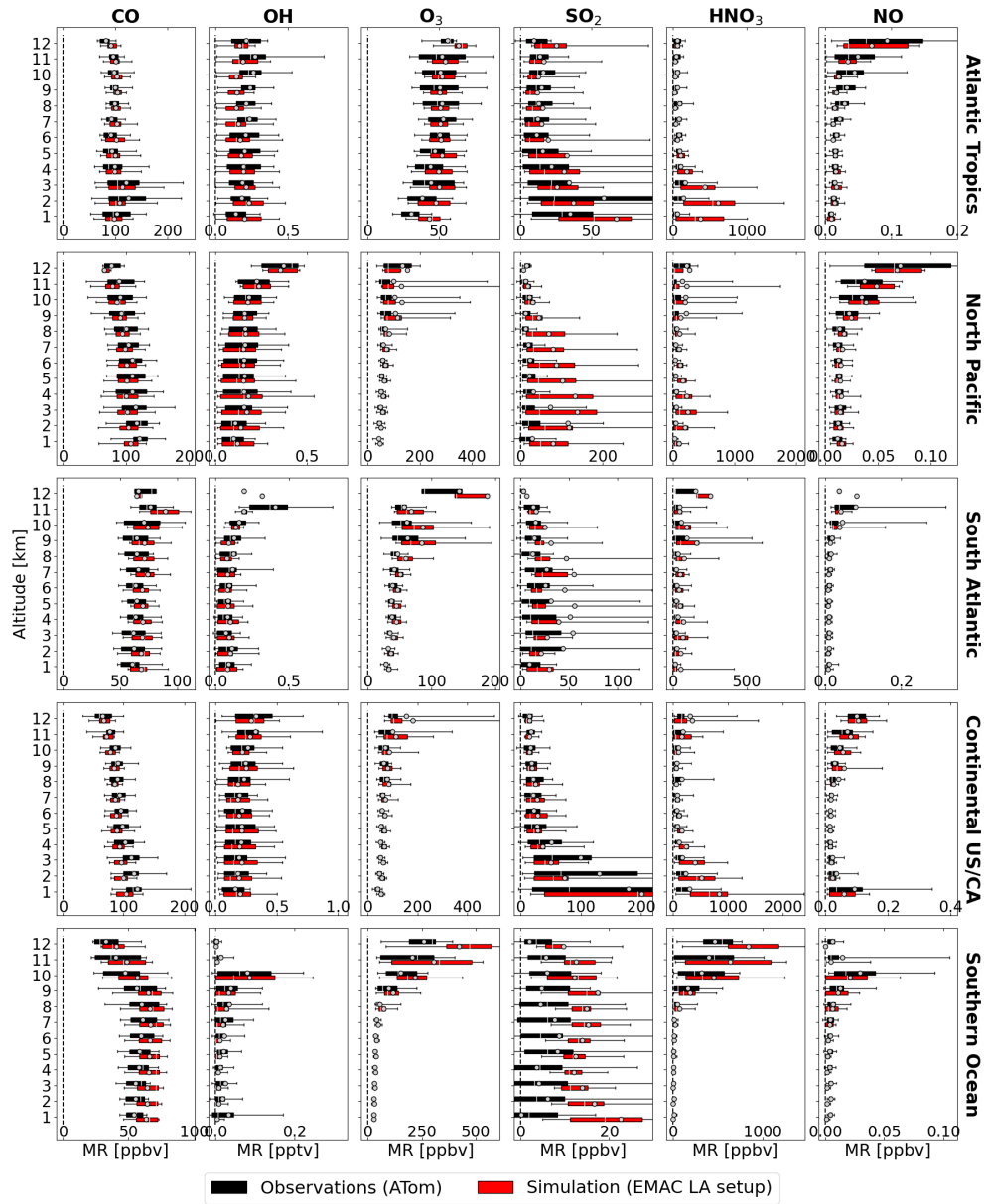


Figure S5. Comparison of simulated (red) and observed (blue) mixing ratios (MR) of carbon monoxide (CO), the hydroxyl radical (OH), ozone (O₃), sulfur dioxide (SO₂), nitric acid (HNO₃), and nitric oxide (NO) from the ATom mission across selected regions, as defined in Fig. C1 from the manuscript. Box-whisker plots follow the format of Fig. 4 from the manuscript.

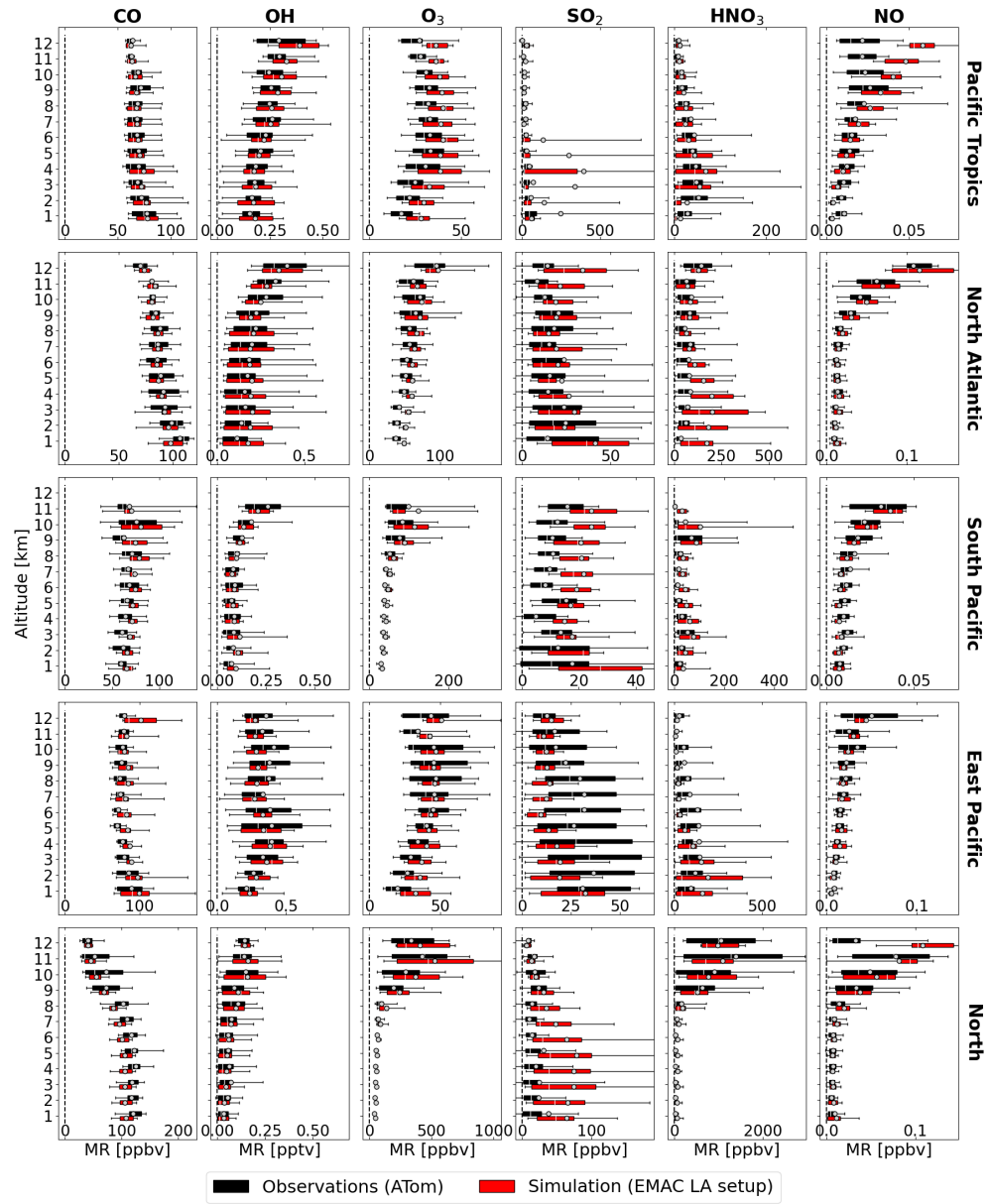


Figure S6. As Fig. S5, but for the remaining regions

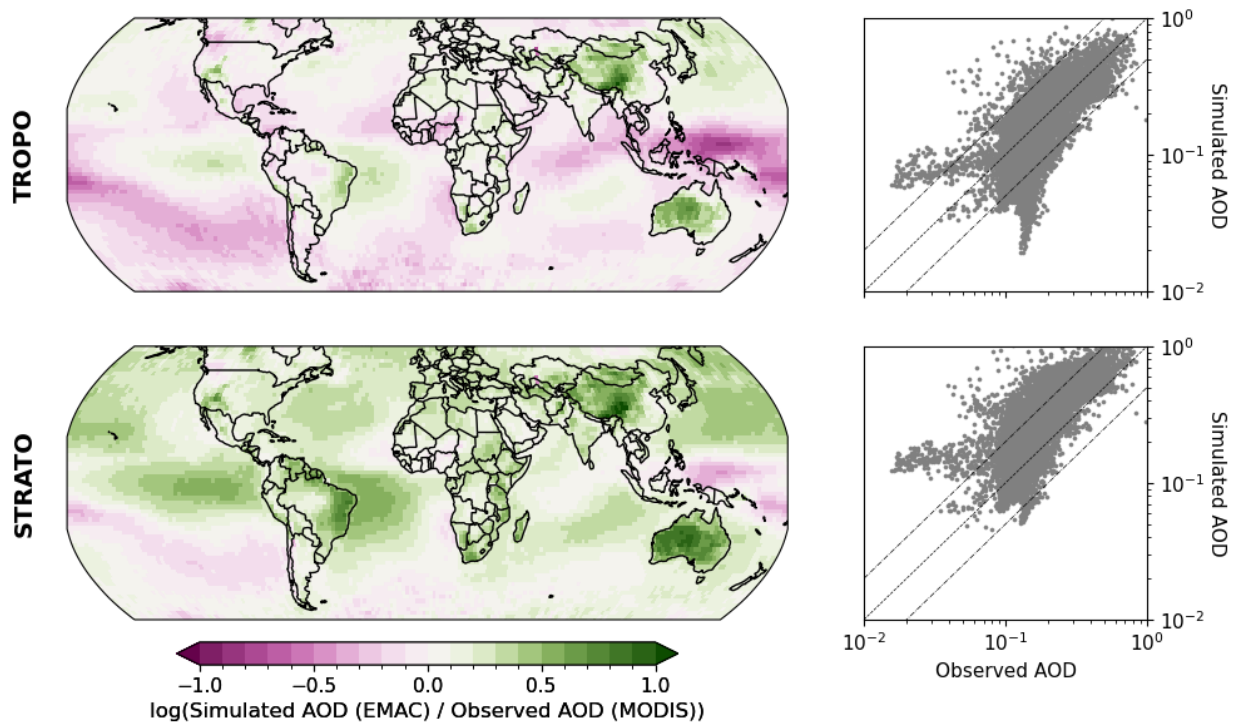


Figure S7. As Fig. 3 from the manuscript, but with simulated values from the STRATO and the TROPO setup.

6 CHAPTER

Process studies & Extended Outlook

The preceding chapters focused on the development of a model setup for the simulation of atmospheric aerosols from the surface to the stratosphere, its evaluation, and resulting global aerosol distributions. In addition to global aerosol budgets, the model setup provides a basis for detailed studies of multiphase chemical processes in the atmosphere. This Chapter presents an overview of various studies based on the model developments presented above.

6.1 Asian Summer Monsoon influence on the UTLS

The AMA influence on the UTLS composition is an emerging research topic since the discovery of the ATAL in 2011 [135] (see Sect. 2.5.4 for more details). Three aircraft campaigns have been conducted in recent years to study the AMA and the corresponding aerosol processes. First, the StratoClim mission (July/August 2017) directly sampled the ATAL and its composition in the centre of the AMA. Second, the ACCLIP mission (August 2022) investigated transport pathways of aerosols originating from the ATAL in the eastward extent of the AMA. Third, the PHILEAS mission (August/September 2023) studied the outflow of the AMA to the Extratropical Lower Stratosphere (ExLS). The developed model setup from Chapter 5 was used for two studies, looking into the formation and the fate of ATAL aerosol.

6.1.1 The role of ammonia in the ATAL formation

The work summarized in this Section was published in Nature Partner Journal (npj) — Climate and Atmospheric Science (Xenofontos, Kohl et al., 2024 [157]). I provided the model setup (see Chapter 5) and assisted in the performance of the numerical simulations.

Observations during the StratoClim campaign in 2017, sampling the ATAL region, suggested that the majority of ATAL aerosol is purely secondary (up to 70%), originating from NPF [136, 132], as primary particles are washed out during convection. The ATAL is mostly composed of ammonium nitrate and organics [136]. Ammonia, as a gaseous precursor of ammonium, is emitted in great abundance in South and East Asia, mostly originating from agricultural activity. Moreover, recent experiments from the CLOUD collaboration at CERN found, that ammonia can enhance NPF significantly, especially in the presence of nitric acid [158].

We conducted model simulations to investigate the impact of ammonia on NPF and secondary aerosol formation in general in the ATAL. After demonstrating reasonable agreement with observations, sensitivity studies were performed, separately excluding ammonia emissions and

various nucleation pathways. The results indicate great importance of ammonia emissions to the formation of the ATAL. Excluding ammonia emissions leads to a reduction of NPF and ammonium nitrate concentrations by an order of magnitude. Thus, ammonia emissions do not only sustain ammonium concentrations in the ATAL, but are crucial for the formation of the ATAL itself.

The additionally formed particles from South and East Asian ammonia emissions increase CCN concentrations up to 80% locally compared to a scenario without any ammonia emissions, with potential impacts on indirect radiative forcing by influencing the cloud albedo. Furthermore, the AOD at 550 nm increases by up to 70% locally due to the formation of ammonium nitrate limited by ammonia availability, exerting a direct radiative forcing.

6.1.2 Transport of aerosol originating from the AMA into the ExLS

The work presented in this Section is part of a manuscript currently in review for Nature Communications Earth & Environment (Köllner, Kohl et al., 2025). All model simulations were performed and analysed by me.

Introduction and Methods Once the ATAL is formed, particles can either be removed by sedimentation and wet scavenging, or they can be distributed across the UTLS region. Previous studies showed that after initial convection, air masses are slowly transported upwards in anticyclonic spiralling motion at a rate of about 1 K d^{-1} [159, 160] between 360 and 440 K in potential temperature. During this transport, particles can grow via coagulation and condensation. Further transport is speculated to be realised via the large-scale Brewer Dobson circulation or quasi-horizontal isentropic transport to the ExLS, also called eddy-shedding [160]. It has been shown, that the quasi-horizontal isentropic transport is more effective than the Brewer Dobson transport for gas phase species from the AMA [159, 161].

The PHILEAS mission in 2023 aimed to investigate the outflow of the AMA to higher latitudes. The ERICA-AMS instrument [162] on board of the HALO research aircraft detected enhanced concentrations of ammonium, nitrate and organics in ExLS air masses (above 370 K potential temperature) associated with high methane and dichloromethane mixing ratios, originating from AMA air (Fig. 6.1a). In addition, the GLORIA instrument [163, 164] detected solid ammonium nitrate particles in the same air masses (Fig. 6.1b).

I performed EMAC simulations to study transport pathways from the ATAL into and the persistence of AMA aerosol in the ExLS, with model setup and evaluation as outlined in Chapter 5. To distinguish between aerosol originating from Asian sources trapped in the AMA and other sources, we performed a series of sensitivity simulations. In addition to the reference simulation with baseline emissions from the CEDS database [165], we removed ammonia (NH_3), nitric oxide (NO) and sulfur dioxide (SO_2) emissions from South and East Asia altogether, and each species separately, to study the effect of the emissions on the stratosphere in general and the ExLS in particular. Figure 6.2 exemplarily shows the reduced ammonia emissions in the sensitivity compared to the reference simulation.

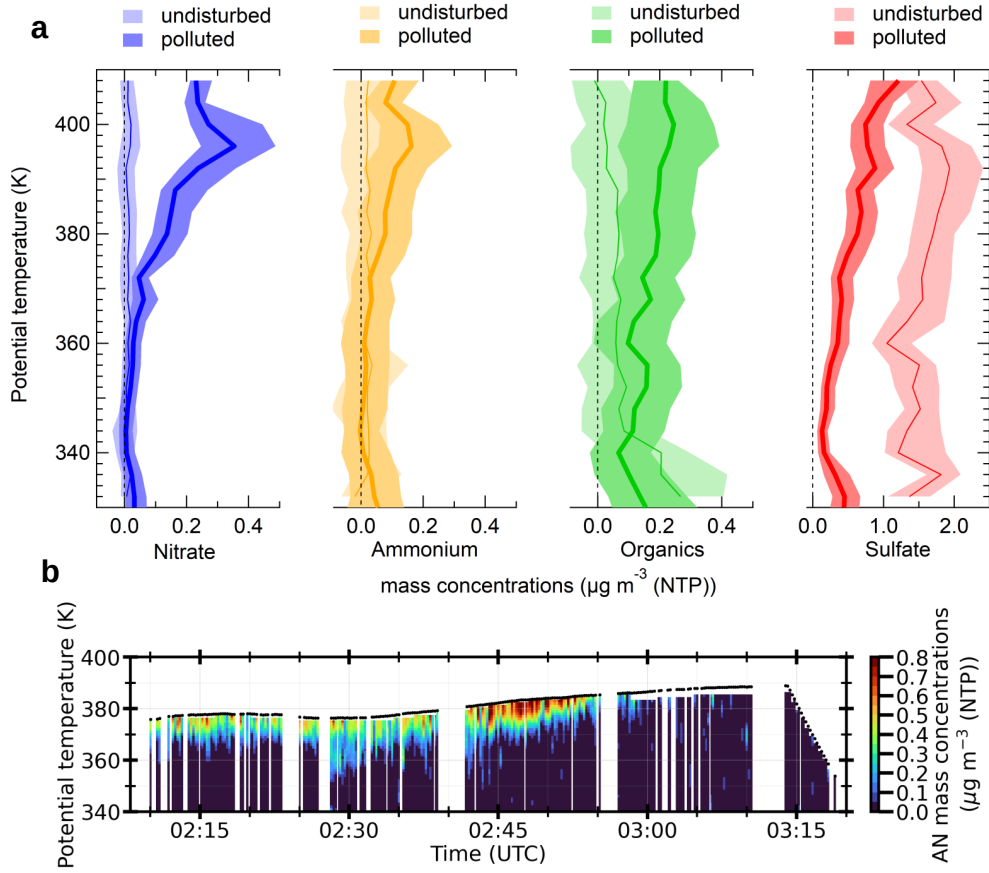


Figure 6.1 **a)** Profiles of aerosol mass concentrations in the ExLS, measured by the ERICA-AMS during the PHILEAS campaign. The lines show median values, and the shaded area the 25/75 percentile of the measurements at CH₄-poor (CH₄ < 1860 ppb; light-shaded area) and at CH₄-rich (CH₄ > 1920 ppb; dark-shaded area). **b)** Limb-imaging observations from the GLORIA instrument of particulate ammonium nitrate measured during the PHILEAS mission at September 01. The figure was prepared by Franziska Köllner, data was collected by the ERICA-AMS and GLORIA instruments.

Model Results First, we studied the transport pathways of air masses containing aerosol from the ATAL. Figure 6.3d-f displays zonally (75° E to 135° W, including the ATAL and the eastward outflow region probed during the PHILEAS mission) and monthly averaged profiles of ammonium concentrations in the Northern Hemisphere above 300 K from the reference simulation. In August and September 2023 (Fig. 6.3d), during the PHILEAS mission, a clear maximum in ammonium concentrations centred at 400 K and 30° N is simulated, representing the ATAL. Towards higher latitudes, ammonium concentration isolines along constant potential temperatures imply isentropic transport of ammonium-rich air masses to the ExLS. By November (Fig. 6.3e), the ammonium layer descends onto the 8 PVU (Potential Vorticity Unit) line, marking the upper boundary of the tropopause region, driven by large-scale subsidence from the Brewer-Dobson circulation. Ammonium concentrations are depleted over the turn of the year, and a background ammonium layer forms, visible in February 2024 (Fig. 6.3f), about 2-3 months before the onset of the next monsoon season.

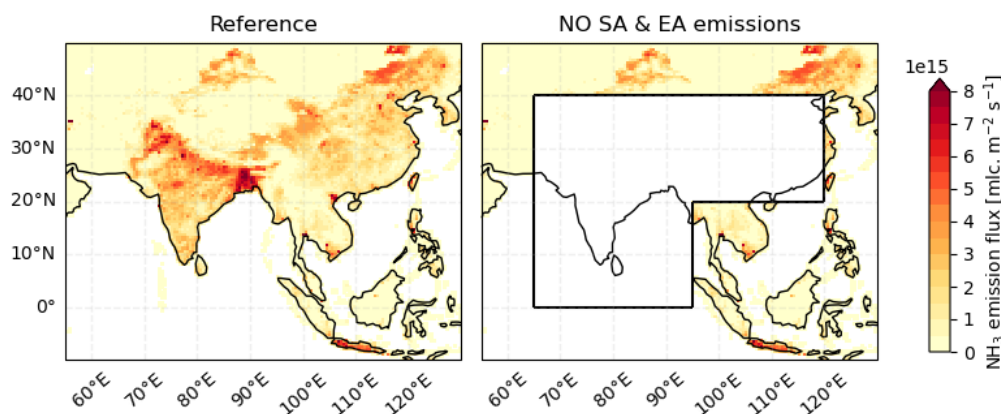


Figure 6.2 Ammonia (NH_3) emission flux from anthropogenic surface sources in August. The left panel shows baseline emissions in the reference simulation from the CEDS database, the right panel shows the regions, where NH_3 emissions were excluded in the sensitivity studies.

This analysis confirms the assumption of quasi-horizontal transport of ATAL aerosol into the ExLS, and highlights the persistence of elevated ammonium concentrations over several months.

Second, we studied the role of anthropogenic emissions for UTLS ammonium nitrate concentrations, and the respective seasonality and persistence. Figure 6.3a,c display zonally (global) averaged ammonium and nitrate concentrations between 370 and 390 K in potential temperature. The monsoon season usually starts in late Northern Hemispheric spring, forming the ATAL around May with elevated ammonium and nitrate concentrations. The AMA-sourced ammonium is then transported towards higher latitudes and diluted. By comparing the reference with the sensitivity simulation, removing anthropogenic ammonia, nitric acid and sulfuric acid emissions from South and East Asia, we obtain the contribution of these emissions to UTLS ammonium concentrations (6.3b). Indeed, elevated ammonium concentrations can almost exclusively be attributed to South and East Asian emissions. The same holds true for nitrate concentrations (not shown). Moreover, an additional sensitivity simulation, removing only anthropogenic ammonia emissions, highlighted that South and East Asian ammonia emissions are almost solely responsible for the enhanced ammonium nitrate concentrations in the UTLS. We concluded that ammonium nitrate formation and persistence is limited by ammonia availability, while nitric acid is sufficiently available to form ammonium nitrate.

While enhanced ammonium concentrations persist until the onset of the next monsoon season, nitrate concentrations appear to deplete faster (comp. Fig. 6.3a and c). Nitrate concentrations show high variability over the years, and elevated nitrate concentrations are only occasionally simulated at higher latitudes. This phenomenon might be attributed to the thermodynamics of the ammonia-nitric acid-sulfuric acid system, simulated by ISORROPIA-II [73], a thermodynamic equilibrium model. Once ammonium nitrate particles are transported to higher latitudes, they are mixed with stratospheric air masses containing elevated sulfur concentrations, transitioning from ammonia-rich to ammonia-poor conditions relative to

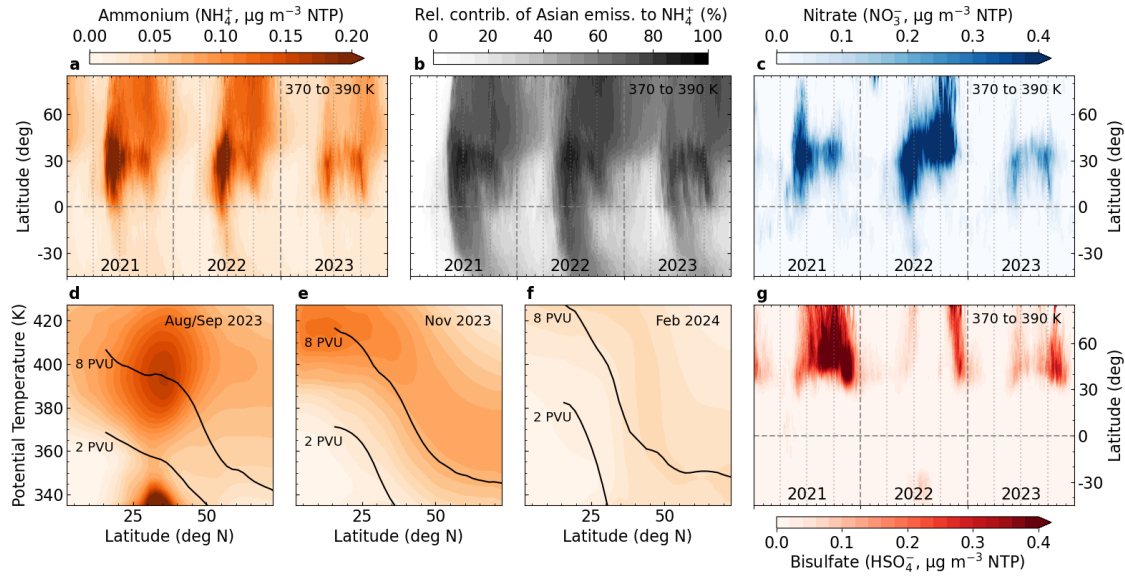


Figure 6.3 Simulated UTLS aerosol concentrations using the EMAC model. **a-c)** Zonally (75°E to 135°W) and monthly averaged profiles of ammonium concentrations at different stages of the evolution of ASM aerosol in the UTLS: During the PHILEAS mission from Mid-August to Mid-September 2023 (a), in November 2023 (b), and in February 2024 (c). Potential temperature is used as a vertical coordinate, and the black lines represent the 2 PVU and 8 PVU isolines. **d,f,g)** Zonally (global) and vertically (between 370 and 390 K in potential temperature) averaged concentrations of ammonium (d), nitrate (f) and bisulfate (g) from EMAC simulations as a function of latitude and time for the period of 2021-2023. **e)** Difference in ammonium concentrations between the reference simulation and a simulation without anthropogenic emissions from South and East Asia, revealing the ASM contribution to the UTLS ammonium concentrations.

sulfur abundance. Subsequently, the formation of ammonium (bi-)sulfate is favoured over ammonium nitrate, and the nitrate potentially evaporates as nitric acid. In the transition phase, ammonium bisulfate is formed, displayed in Fig. 6.3g. In order to establish a new equilibrium, ammonium nitrate is rapidly replaced by ammonium sulfate, if the ATAL air encounters sulfur-rich stratospheric air within a grid box. However, this process is likely to be overestimated by ISORROPIA-II in the simulations. In reality, the replacement of nitrate by sulfate may be slower, as coagulation is slow, and gas phase sulfuric acid mixing ratios are low in absence of large volcanic eruptions in the stratosphere in general.

Fig. 6.4 supports these findings, showing the time development of zonally averaged relevant aerosol molecular concentrations at ATAL attributed latitudes (30°N ; left column) and in the outflow region (60°N ; right column), and further illustrates the differences between the three simulated years. The abundance of ammonium and nitrate in the ExLS mainly depends on two factors:

- First, the strength of the Asian monsoon is controlled by the El Niño Southern Oscillation (ENSO) [166, 167], with stronger convection in La Niña years where moist Pacific air is transported to South and East Asia and weaker convection in El Niño years. Thus, the La Niña years 2021 and 2022 show a stronger ATAL with higher

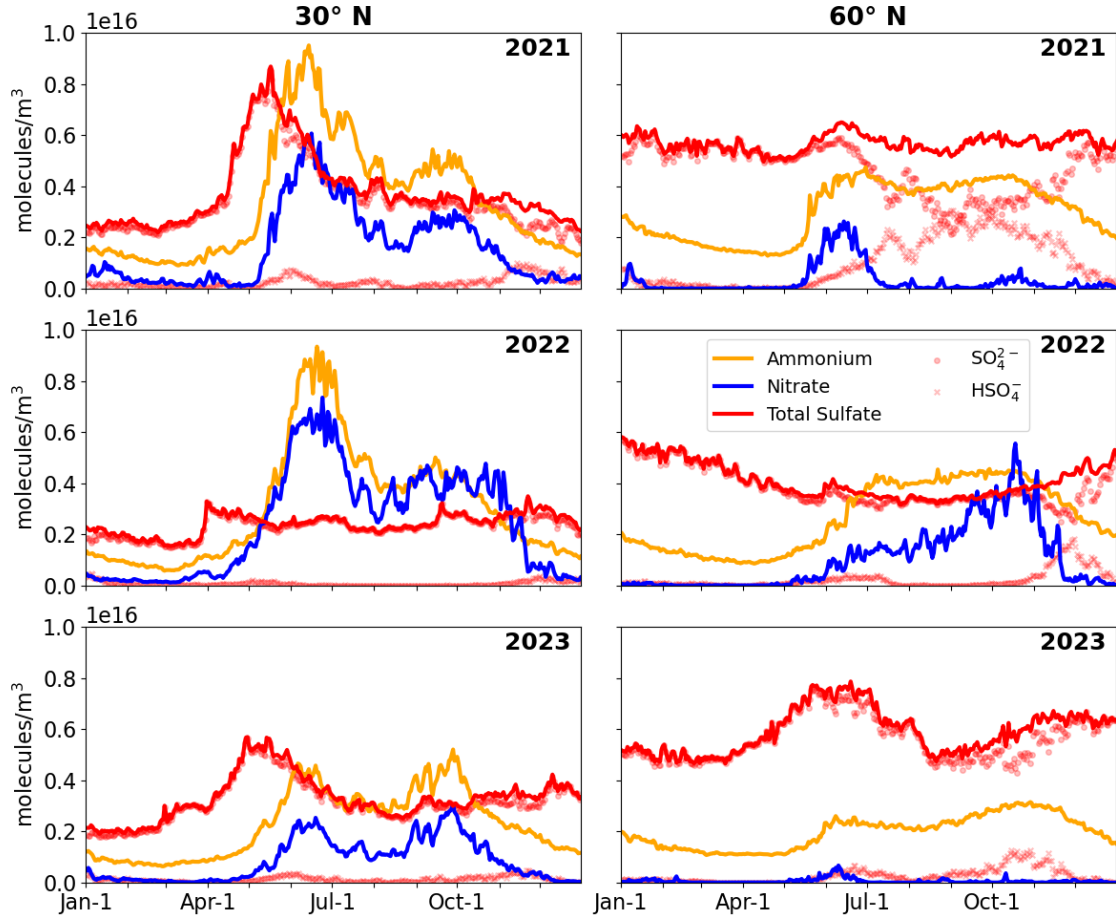


Figure 6.4 EMAC simulated timeline of zonally averaged inorganic aerosol concentrations between 370 and 390 K at 30° N (left; ATAL region) and 60° N (right; showing the ASM outflow) latitude. Total sulfate concentrations are displayed (red), consisting of the contribution of bisulfate (HSO_4^- ; red crosses) and the remaining sulfate (SO_4^{2-} ; red circles). Bisulfate forms in the transition between ammonia-rich to ammonia-poor conditions, according to the model results. The years 2023 and 2021 were influenced by increased volcanic activity. In 2023, the ATAL was weaker than in the previous years.

ammonium nitrate concentrations in the source region (30° N) relative to the El Niño year 2023. Subsequently, more ammonium nitrate is transported to the ExLS.

- Second, the availability of sulfur species in the stratosphere controls the persistence of particulate nitrate in the stratosphere, as outlined above. The main cause for variability in stratospheric sulfur burdens are volcanic eruptions. 2021 was characterised by high volcanic activity, showing enhanced molecular sulfate concentrations in the ExLS throughout the year, exceeding ammonium concentrations. As a result, ammonium (bi-)sulfate quickly forms, as fresh sulfuric acid condenses on the ATAL particles, and particulate nitrate is almost entirely depleted in August. 2023 combines high sulfur aerosol loadings with a weak ATAL, leading to very low nitrate concentrations overall. 2022 in contrast showed relatively lower stratospheric sulfur aerosol burdens, with

enhanced nitrate concentrations in the ExLS until mid of November.

6.2 Amazon basin

The amazon basin is one of the most important ecosystems on planet Earth, representing about 40% of global tropical rainforest area [168, 169]. However, the Amazon rainforest is quickly changing, driven by deforestation, agricultural expansion and climate variability [170]. In December 2022 and January 2023, the CAFE-Brazil campaign was conducted to explore the chemical and microphysical processes over the Amazon rainforest. Additionally, to understand the physical and chemical processes of atmospheric composition within and above the canopy, long-term continuous measurements of various parameters are being performed using a comprehensive suite of instruments at the ATTO tower [171, 172, 173]. The EMAC model setup presented in Chapter 5 was used to further investigate these processes and support observations.

6.2.1 Sensitivity of Amazonian chemistry and climate to biogenic VOC emissions

The work presented in this Section is part of a manuscript accepted for Nature Communications (Tripathi et al., 2025). All model simulations were performed and analysed by me, with the help of Ryan Vella and Andrea Pozzer

Introduction and Methods Terrestrial vegetation emits biogenic volatile organic compounds (BVOCs) in response to their physiological (i.e., temperature, light, and stress) and ecological process. Globally, the Amazon rainforest accounts for 40% of BVOC emissions, which can affect local, regional, and global atmospheric chemistry and climate [91]. Isoprene (C_5H_8) and monoterpenes ($C_{10}H_{16}$) are the most dominant primary BVOCs emitted from the Amazon rainforest [91]. They both undergo oxidation in the atmosphere, producing respective oxidation products (OP). These OPs contribute to SOA formation in the atmosphere, and were shown to form new particles [79, 78, 174, 175].

Measurements of mixing ratios of various BVOCs have been performed during the CAFE-Brazil campaign and at the ATTO tower to understand their sources, chemistry and transport. CAFE-Brazil measurements show enhanced mixing ratios of isoprene, monoterpene and isoprene OP in the UT (see blue boxes in Fig. 6.5) between 10 and 13 km. These enhancements are attributed to rapid convective transport of locally emitted BVOCs to the UT. The transport is illustrated in more detail in Fig. 6.6, displaying diurnal cycles of isoprene, isoprene OP and monoterpenes as measured from ATTO in the BL (320 m), and in the FT (3–9 km) and the UT (9–14 km) during the CAFE-Brazil campaign.

The highest BL isoprene and monoterpene mixing ratios are observed in the noon and afternoon hours, coinciding with the highest solar flux and temperature, required for BVOC emissions. The lifetime of isoprene against reaction with OH is short at day [173], resulting in a rapid build-up of isoprene OP in the BL. Due to the limited availability of OH at night, isoprene and isoprene OP can accumulate in the residual layer and are lofted to the FT at nighttime. Early morning convection then rapidly transports the BVOCs to the UT, leading to the highest isoprene mixing ratios between 4 am and 7 am, where they can quickly oxidize

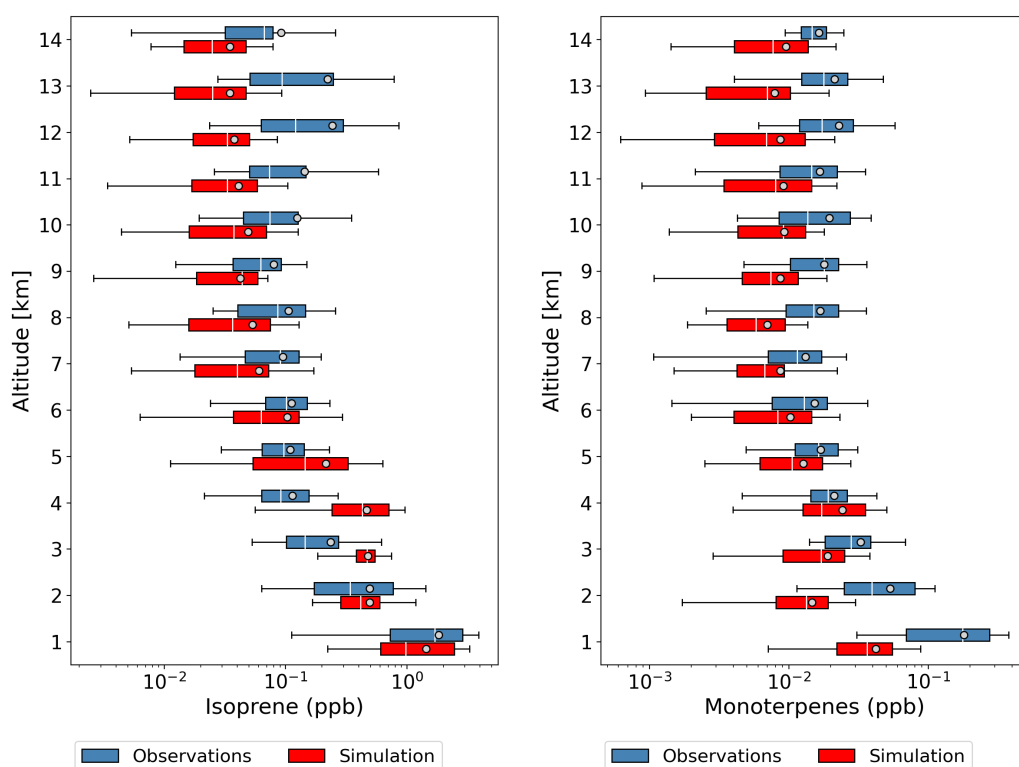


Figure 6.5 Comparison of the vertical profile of the observed and simulated isoprene and monoterpene mixing ratios during the CAFE-Brazil campaign (December 2022 to January 2023). The box-whisker plot represents the mean (circle), median, lower quartile, upper quartile and the 5th and 95th percentiles for 1 km bins. In the free and upper troposphere, the simulated mixing ratios mostly fall below the detection limit. To facilitate comparison with the observations, the simulated values were adjusted (“smeared”) using instrument noise.

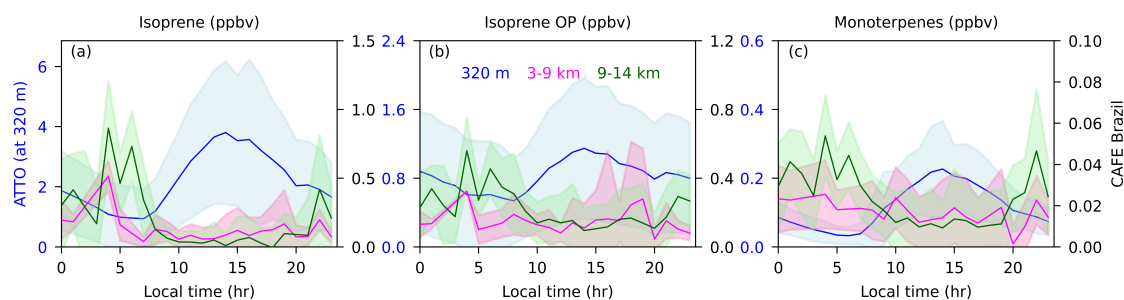


Figure 6.6 Diel cycles of isoprene, its OP and monoterpenes for three different altitude regimes of 320 m (left side y-axis) at ATTO, 3-9 km, and 9-14 km (right side y-axis) during the CAFE-Brazil campaign. The figure was prepared by Nidhi Tripathi, data was collected during the CAFE-Brazil mission and at the ATTO tower.

again after sunrise, when OH is produced.

BL isoprene and monoterpene mixing ratios over deforested regions were measured to be

reduced by about a factor of 4 compared to forested regions during the CAFE-Brazil campaign. Conversely, BVOC emissions might increase with climate change, by up to 50% in a doubled CO_2 scenario [176]. To investigate the sensitivity of atmospheric chemistry and climate dynamics to respective changes in BVOC emissions, several simulations were performed with decreased (50% and 75%) and increased (50%) BVOC emissions, by uniformly scaling model emissions from MEGAN [91] over the Amazon basin.

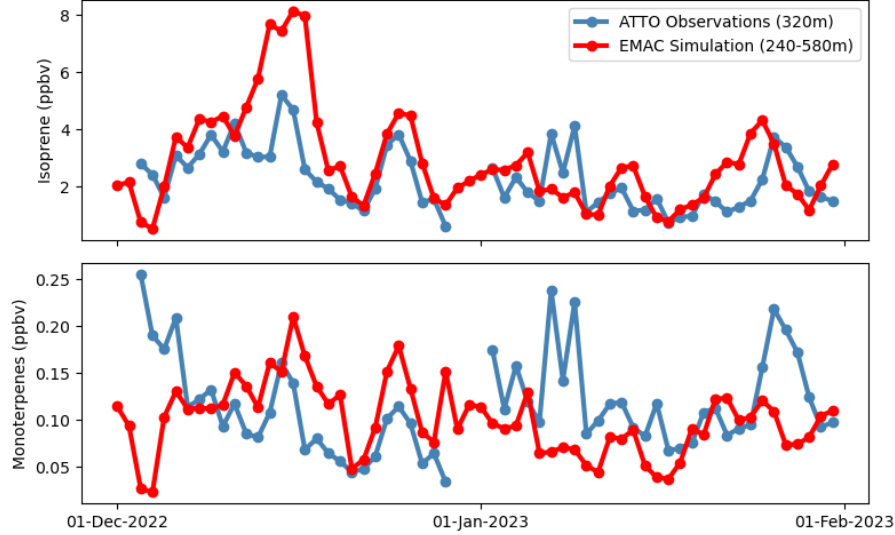


Figure 6.7 Observations of isoprene and monoterpene mixing ratios at ATTO tower (at 320 m) compared to simulated values in the grid box covering 240 to 580 m.

The simulations were performed with the setup outlined in Chapter 5. The reference simulation with baseline emissions was evaluated against ATTO (Fig. 6.7) and CAFE-Brazil (Fig. 6.5) observations. BVOC mixing ratios in the BL agree well with ATTO observations, validating the model BVOC emissions. The elevated isoprene and monoterpene mixing ratios in the UT cannot be reproduced by sampling the model output at the time and location of the flights. This model underestimation may be attributed to the flight planning, actively sampling convective outflow. Conversely, local convective events cannot be explicitly resolved in the model simulation, and only an average upward mass flux per grid box is calculated. Furthermore, observed convective events do not necessarily coincide with simulated ones.

Model results While the observed elevated BVOC mixing ratios in the UT over the Amazon basin cannot be reproduced on average over the CAFE-Brazil flight tracks, the model is capable of reproducing the diurnal cycle and the order of magnitude of maximum isoprene mixing ratios. The left panel of Fig. 6.8 displays the averaged diurnal cycle of maximum isoprene mixing ratios over the Amazon rainforest in December 2022 and January 2023 (period of CAFE-Brazil campaign). As observed, the highest BL isoprene mixing ratios are simulated in the afternoon, with delayed elevations in FT isoprene mixing ratios, resulting from shallow convection up to around 4 to 5 km. Nighttime deep convective events transport isoprene to the UT. The highest isoprene mixing ratios over the Amazon basin around 2 ppbv are simulated in the early morning hours before sunrise. Subsequently, isoprene oxidizes with OH and is depleted in the UT. These model results support the findings from the isoprene

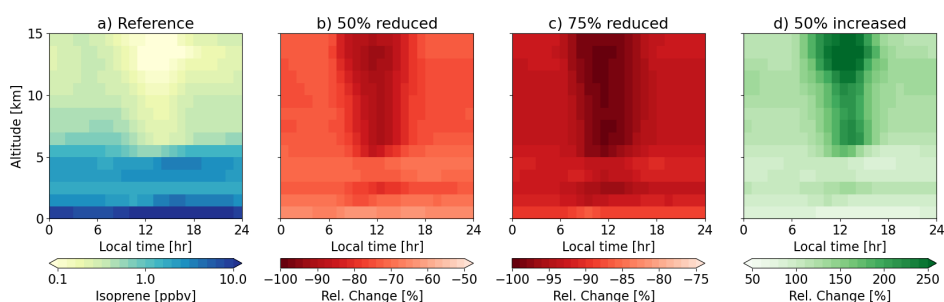


Figure 6.8 Simulated average diel cycle of maximum isoprene mixing ratios in December 2022 and January 2023 over the Amazon basin from the surface to the UT in the reference simulation (left). Highest boundary layer mixing ratios are realized in the afternoon, while the UT shows highest isoprene mixing ratios in the early morning hours. In addition, we provide changes to this scenario in the sensitivity simulations, showing a non-linear decrease, respectively increase in the UT, especially at day time, in response to decreased (increased) BVOC emissions.

observations during CAFE-Brazil.

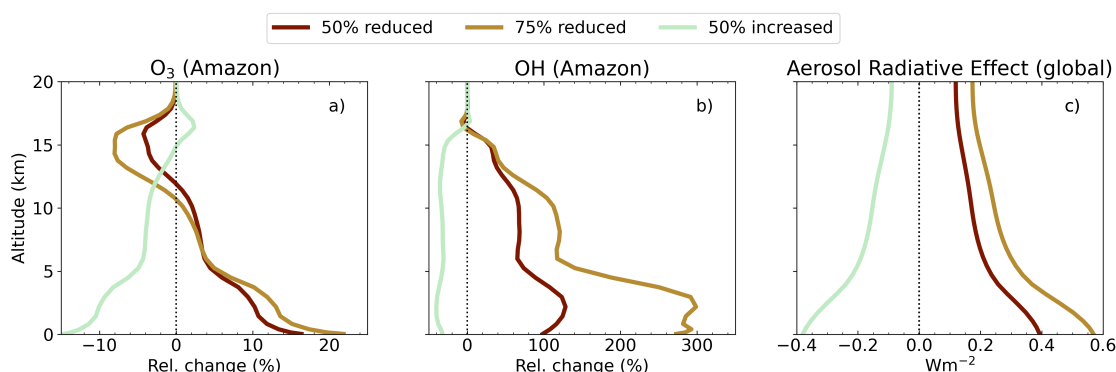


Figure 6.9 The vertical profiles of relative changes in a) O₃ and b) OH and of absolute changes in c) the global radiative effect due to aerosol between the base simulation and the simulation with 50 and 75% reduction and 50% increase in the BVOCs emission. The radiative effect is the imbalance of incoming and outgoing radiation and it is calculated at each vertical level

The sensitivity of isoprene, ozone and OH mixing ratios, and the Earth's radiative balance to changes in Amazonian BVOC emissions is displayed in Fig. 6.8b-d and 6.9. The main oxidant for BVOCs in the atmosphere is OH. Consequently, OH mixing ratios are very sensitive to changes in BVOC emissions. A reduction of BVOC emissions by 75% leads to an increase of OH mixing ratios by around a factor of 3 in the lowest 5 km of the atmosphere above the Amazon basin (Fig. 6.9b) as less OH is consumed in reactions with BVOCs. The strength of this effect decreases towards the upper troposphere due to competing processes, however still leading to an OH increase of around 60%. Similarly, OH mixing ratios decrease with increasing BVOC emissions, but to a lesser extent. This non-linearity in the OH response to OH mixing ratios may be attributed to the efficient buffering of OH mixing ratios by OH

recycling, that prevent the depletion of OH [177].

This OH effect in turn leads to a non-linear response of isoprene mixing ratios to BVOC emissions. A decrease of BVOC emissions, including isoprene, by 50% leads to a decrease in isoprene mixing ratios of at least 60–70% everywhere, and up to 90% in the UT at daytime. The higher OH availability additionally depletes the already lower isoprene levels even more. Thus, isoprene levels are not only proportionally influenced by BVOC emissions, but additionally encounter a positive feedback by the decreased (increased) isoprene lifetime for emission reductions (increases).

Ozone over the Amazon basin encounters differing effects depending on altitude and ozone production sensitivity (Fig. 6.9a). Ozone production can either be limited by NO_x or VOC availability [178]. Close to the surface, O_3 production is NO_x limited. Thus, changes in VOC emissions do not significantly influence O_3 production. However, O_3 can also oxidize BVOCs, and thereby acts as a sink. Consequently, O_3 encounters similar effects in the BL and FT as OH, albeit at reduced magnitude. In the UT, this effect is reversed. Due to the high availability of lightning NO_x , O_3 production is limited by VOC availability. As a result, reductions (increases) in BVOC emissions lead to reductions (increases) in O_3 mixing ratios in the UT.

As O_3 is a greenhouse gas, changes in ozone mixing ratios, lead to changes in Earth’s radiative balance. Reducing Amazonian BVOC invokes a local warming close to the Earth’s surface, and a slight global cooling effect in the tropical UT around the globe. However, the radiative effect of changes in BVOC emissions is strongly dominated by the aerosol radiative effect. Monoterpenes readily oxidize in the atmosphere and potentially form SOA. Thus, a decrease in monoterpene emissions leads to reduced AOD, lowering the Earth’s albedo, and thereby warming Earth’s surface and atmosphere. A reduction of Amazonian BVOC emission by 75% leads to a radiative effect of 0.6 W m^{-2} globally, roughly offsetting 60% of the global cooling through anthropogenic aerosol emissions (compare Chapter 2.6). Additionally, the simulations reveal an impact of the altered OH availability on methane lifetime, a strong greenhouse gas. A 75% reduction in BVOC emissions would reduce global methane lifetime by 0.3–0.4 years, resulting in a slight cooling, in agreement with findings from Scott et al. (2018) [179].

This study mostly focused on oxidation capacity and short-lived climate forcers in the atmosphere. However, changes in BVOC emissions would be accompanied by additional atmospheric effects, that could not be explored in the presented simulations. Deforestation or afforestation of the Amazonian rainforest would strongly influence the carbon cycle with strong implications for the climate system, more detailed investigated by Weber et al. (2014) [180]. In addition, recent studies found, that isoprene OPs in the Amazonian UT lead to NPF, potentially transported to lower altitudes, and producing CCN, thereby influencing the hydrological cycle and again the climate system [175, 174]. This effect is not yet accounted for in our model, but ongoing research is presented in the following.

6.2.2 Aerosol cycle

The work presented in this Section summarizes ongoing studies in collaboration with Mira Pöhlker, Joachim Curtius, Christopher Pöhlker, Douglas Russell, Peter Lloyd, Rolf Sander,

Jos Lelieveld, Andrea Pozzer, and others on PNC over the Amazon rainforest. I perform and analyse all model simulations, and implemented the yield and the new NPF pathway.

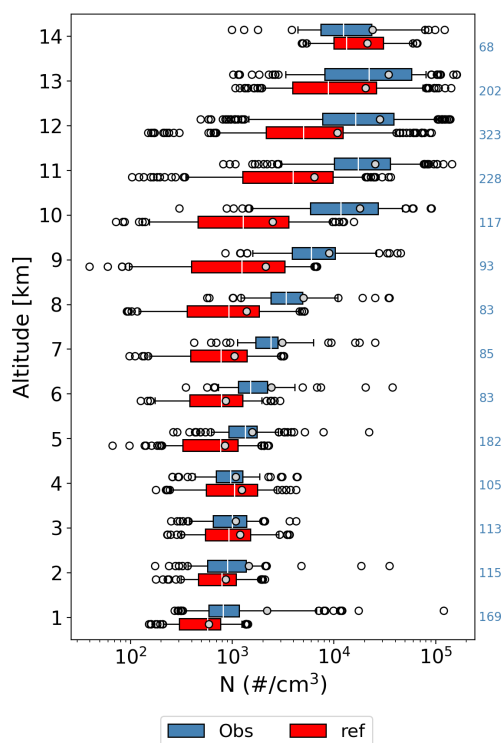


Figure 6.10 Comparison of the vertical profile of observed and simulated (reference simulation) PNC during the CAFE-Brazil campaign (December 2022 to January 2023). The box-whisker plot represents the mean (circle), median, lower quartile, upper quartile and the 5th and 95th percentiles for 1 km bins. Outliers are shown as individual points. Observational data were provided by Mira Pöhlker and Peter Lloyd.

PNC are in very good agreement within the lowest 4 km of the atmosphere, and again near the tropopause around 14 km. However, between approximately 5 and 13 km, the model strongly underestimates observed PNC. An analysis of the ozone-to-carbon monoxide ratio, used as an indicator to distinguish between stratospheric and tropospheric air masses, reveals that the underestimation predominantly occurs in tropospheric air (not shown). This points towards a likely missing source of NPF in the model, potentially linked to the proposed NPF pathway involving isoprene OP in the tropical UT.

In Sect. 6.2.1, it was shown that both the diurnal cycle and the magnitude of peak isoprene mixing ratios in the Amazonian UT can be reasonably reproduced by the model,

Introduction and Methods Recently, two studies proposed a new NPF pathway in the tropical UT, involving isoprene OP in combination with acids, based on field measurements [175] and on laboratory experiments at the CLOUD facility [174]. At the same time, evaluation of the newly developed model setup against observations from the ATom aircraft mission (see Chapter 5) revealed an underestimation of PNC in the UT near the Amazon basin, potentially caused by the suggested nucleation mechanism. Therefore, we aim to assess the local transport and global climate impacts of freshly formed particles from the proposed pathway by conducting EMAC simulations with the model configuration described in Chapter 5. Here, I present the status of the implementation and first results.

Fig. 6.10 shows a comparison of simulated and observed PNC during the CAFE-Brazil campaign. Observations (*Mira Pöhlker and Peter Lloyd, personal communication*) are provided by the Fast Airborne Size Distribution (FASD) instrument in combination with an Ultra-High Sensitivity Aerosol Spectrometer (UHSAS), providing aerosol size distributions from 2 nm to 1 μ m. More information on the instrument is provided by Curtius et al. [175]. Simulated and observed

although values are underestimated when directly compared to aircraft observations from the CAFE-Brazil mission, potentially due to sampling biases introduced by flight planning (see discussion in Sect. 6.2.1).

Shen et al. [174] identify isoprene-oxygenated organic molecules (IP-OOMs), containing at least four carbon and four oxygen atoms, as drivers of NPF in the upper troposphere. One proposed formation pathway of IP-OOMs involves a twofold OH oxidation and reaction with O_2 , followed by reactions with either HO_2 or NO_2 , leading to the production of first- and second-generation OPs. As the chemical mechanisms currently implemented in EMAC only account for the first-generation products, i. e. reaction with OH, O_2 , and NO_2 or HO_2 , I am working on the integration of the more detailed isoprene oxidation mechanism developed by Wennberg et al. [181] in collaboration with Rolf Sander and Douglas Russell.

Shen et al. [174] additionally provide an estimate for the yield of second-generation OPs, derived from the reaction of ISOPOOH (first-generation isoprene OP formed via reactions with OH, O_2 , and HO_2) with OH. Their results suggest a yield range between 10% and 90%. As a first study, I performed two sensitivity simulations, applying globally constant yields of 20% and 75%, respectively, for the production of second-generation isoprene OPs. These simulations are compared to a reference simulation in which NPF from isoprene oxidation was not considered.

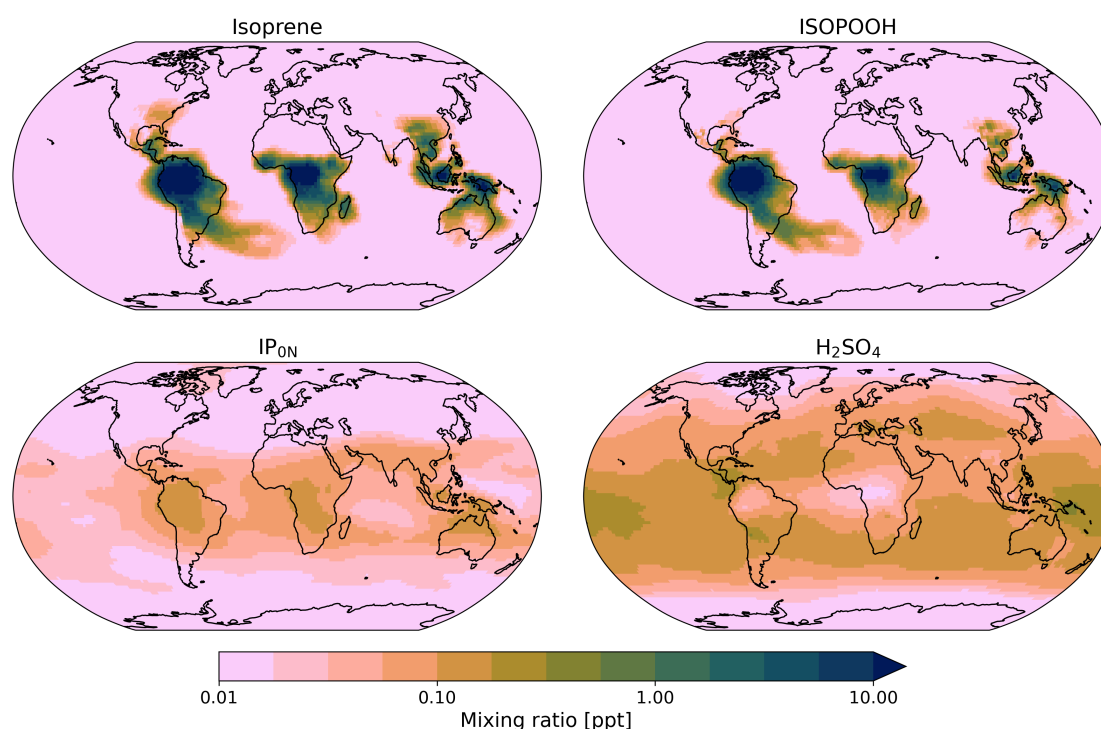


Figure 6.11 Annually averaged (2022) simulated (20% yield of IP_{0N}) UT (10–13 km) mixing ratios isoprene, the first-generation OP ISOPOOH, the second-generation OP IP_{0N} (assuming a 20% yield; see text for more details), and sulfuric acid (H_2SO_4).

A preliminary parameterization of the IP-OOM–acid nucleation rate is adopted from Shen

et al. [174], based on laboratory measurements at two temperatures (-30°C and -50°C), and implemented within the NAN submodel:

$$J_{1.7} = 10^{a \times \log_{10}(x) + b} \quad (6.1)$$

with $x = (\text{H}_2\text{SO}_4 + \text{HIO}_x) \cdot \text{IP}_{0\text{N}}$, where H_2SO_4 represents sulfuric acid concentrations and $\text{IP}_{0\text{N}}$ denotes second-generation isoprene OPs without nitrate addition (see [174, 175]). Iodic acid (HIO_x) is not considered in the current implementation, as it is not represented in the present EMAC configuration. The temperature-dependent parameters are $a = 1.505$ and $b = -19.948$ at -30°C , and $a = 1.718$ and $b = -22.198$ at -50°C . Nucleation is only considered at temperatures below -30°C , using a linear interpolation of the nucleation rates between -30°C and -50°C , and applying the -50°C parameterization for temperatures colder than -50°C .

Model results Figure 6.11 shows the annually and vertically averaged mixing ratios of key species in the upper troposphere (10–13 km) for the simulation with a 20% yield of $\text{IP}_{0\text{N}}$. Elevated isoprene mixing ratios are confined to regions above tropical rainforests, reflecting the strong biogenic source. The first-generation OP, ISOPOOH , exhibits a similar spatial distribution, albeit with reduced magnitude, as expected from its formation pathway. The concentration of $\text{IP}_{0\text{N}}$, the second-generation OP, is further diminished and exhibits a broader zonal spread. This distribution may result from a modelled re-evaporation or inefficient partitioning of $\text{IP}_{0\text{N}}$ in regions with low total organic concentrations, a behaviour that may not accurately reflect atmospheric conditions. Interestingly, sulfuric acid concentrations tend to show minima in regions with the highest isoprene levels, indicating either enhanced condensation sink effects or limited availability of the oxidant OH (see Sect. 6.2.1).

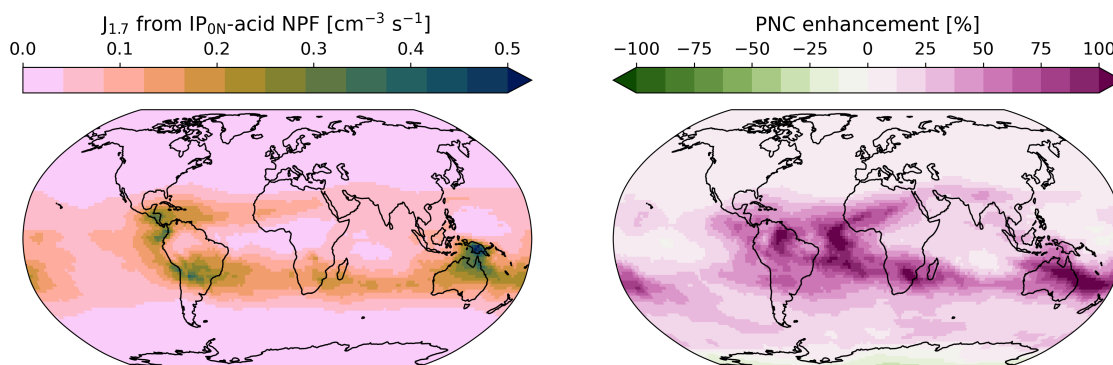


Figure 6.12 Annually averaged (2022) simulated (20% yield of $\text{IP}_{0\text{N}}$) UT (10–13 km) NPF rate ($J_{1.7}$) from the IP-OOM-acid pathway [174, 175] (left) and subsequent PNC enhancement compared to a reference simulation without the NPF pathway (right).

Figure 6.12 presents the resulting NPF rate ($J_{1.7}$) from IP-OOM–sulfuric acid nucleation, derived from the combined mixing ratios of sulfuric acid and $\text{IP}_{0\text{N}}$. Simulated nucleation rates in the tropical UT are of similar magnitude to other known NPF pathways (e.g., $\text{H}_2\text{SO}_4\text{-NH}_3\text{-HNO}_3$; not shown). The right panel displays the resulting enhancement in PNC, with local increases exceeding 100%. These results indicate a substantial influence of this

new nucleation pathway on aerosol number in the tropical UT.

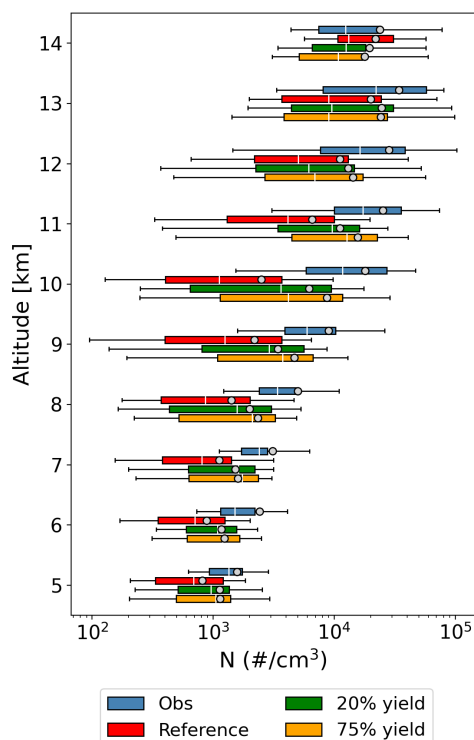


Figure 6.13 Comparison of the 20% and 75 yield simulations to the reference simulation and the observations. Box-whisker plot as in Fig. 6.10

The simulated PNC along the CAFE-Brazil flight tracks for the simulations with 20% and 75% yields are compared to the reference simulation in Fig. 6.13. The inclusion of the new NPF pathway partially reduces the discrepancy in the FT and UT, though PNC remains underestimated even at a 75% yield. Simultaneously, the introduction of this pathway leads to a slight overestimation of UT PNC (previously underestimated; see Chapter 5) in the tropical Atlantic and eastern Pacific, comparable to the other regions (not shown). This suggests that the apparent underestimation during CAFE-Brazil is likely linked to flight planning, which focused on sampling convective outflow, and thus may not be representative of average tropical UT conditions.

This study represents a first attempt, based on several assumptions and simplifications, and thus provides only limited insight into the global implications. The lack of detailed chemistry was addressed using a simplified yield approach, which does not distinguish

between different chemical species or atmospheric regimes. Moreover, only the oxidation pathway via ISOPOOH was considered, despite the existence of additional pathways that may contribute to the formation of NPF-relevant IP-OOMs [181]. The ongoing implementation of the more comprehensive isoprene oxidation mechanism by Wennberg et al. [181] will offer a more complete representation of IP-OOM formation in the tropical UT, and thereby allow for a more constrained and realistic estimation of the associated NPF rates.

Another limitation arises from the gas–aerosol partitioning of the IP-OOMs onto available aerosol surfaces. This process is primarily governed by two factors. First, the condensation of gas-phase species is kinetically limited by the available surface area and the mean free path of the molecules. However, this kinetic limitation is not yet accounted for in the current EMAC implementation of organic gas–aerosol partitioning. Second, the partitioning strongly depends on the volatility of the respective IP-OOMs. While the present implementation follows the volatility estimate of $\text{IP}_{0\text{N}}$ as reported by Curtius et al. [175], this estimate remains highly uncertain and varies across different IP-OOM species. Additional laboratory experiments and box model studies, in combination with the ongoing implementation of the extended isoprene oxidation chemistry, will provide more robust constraints on the volatility

and, consequently, on the partitioning behaviour of IP-OOMs.

Finally, the nucleation rate used in this study is preliminary and only includes limited temperature dependence. Recent experiments at the CLOUD facility have resolved isoprene oxidation at a broader range of temperatures, enabling a more comprehensive representation of temperature dependence.

6.3 Influence of non-stratospheric volcanic eruptions on sulfate concentrations in the lowermost stratosphere

The work presented in this Section summarizes an ongoing study in collaboration with Philipp Joppe, Johannes Schneider and Andrea Pozzer, based on discrepancies between simulated and observed sulfate concentrations during the CAFE-EU campaign, and findings from Joppe et al. [182], showing in-mixing of sulfate-rich air masses into the extratropical lowermost stratosphere in individual flights during the aircraft mission. I perform and analyse all model simulations for the studies.

Introduction and Methods The evaluation of the EMAC model in a simulation assessing the impact of the COVID-19 lockdown on tropospheric composition and aerosol radiative forcing [154] revealed an underestimation of simulated sulfate concentrations by more than one order of magnitude in the FT and UTLS, compared to observations from the CAFE-EU mission. The newly developed model setup for aerosol simulations from the surface to the stratosphere was evaluated in Chapter 5 using a range of aircraft campaign observations, including data from the CAFE-EU mission. While the new setup reduced the discrepancy, thanks to an improved representation of UTLS sulfate concentrations and the inclusion of SO₂ emissions from stratospheric volcanic eruptions using the EVER submodel (see Chapter 4), the model still underestimates observed sulfate concentrations in the UTLS during the CAFE-EU mission.

Joppe et al. [182] further investigated observed sulfate aerosol concentrations during the CAFE-EU campaign, finding elevated levels in the lowermost stratosphere over Europe, strongly influenced by tropospheric air. Based on tracer-tracer correlations, as well as measurements of SO₂, O₃, sulfate, and particle number concentrations, they concluded that mixing processes in the extratropical UTLS transported tropospheric SO₂ into the lowermost stratosphere, with simultaneous gas-to-particle conversion. The enhanced tropospheric SO₂ was speculated to result from the explosive volcanic eruption of the Shiveluch volcano on the Kamchatka Peninsula, Russia.

To assess the impact of the Shiveluch eruption, initially not reaching the stratosphere, on sulfate concentrations in Europe approximately 45 days after the eruption, and to study UTLS mixing processes, a series of EMAC simulations were performed using the setup described in Chapter 5:

- **noVOLC** without the inclusion of any explosive volcanic eruptions.
- **StratVOLC** including stratospheric volcanic eruptions according to the historic EVER setup, described in Chapter 4.

- **Shiv(1 Tg)** with the additional injection of 1 Tg SO₂ at the horizontal geographic location of the volcano, between 7 and 10 km, following a vertically Gaussian distribution centred at 8.5 km with a sigma of 3 km. Emissions were uniformly distributed over three days (April 8–11, 2020).

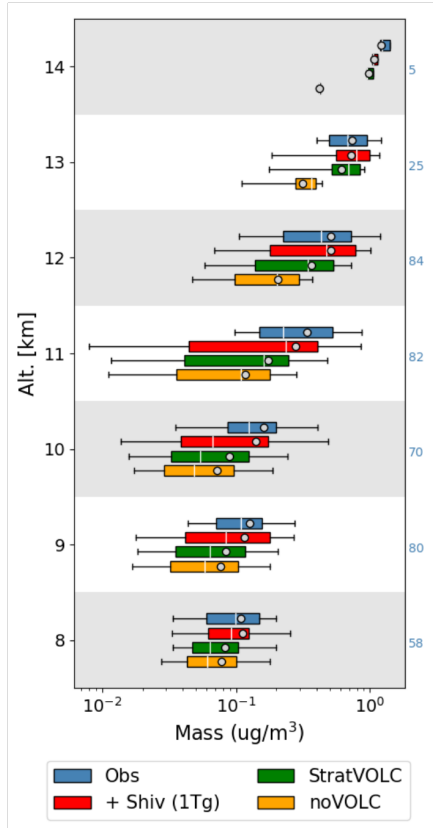


Figure 6.14 Comparison of observed sulfate concentrations (blue) with three EMAC simulations: one without any explosive volcanic eruptions (noVOLC; yellow), one including only stratospheric volcanic eruptions (StratVOLC; green), and one with an additional 1 Tg of SO₂ emissions from the Shiveluch eruption (+ Shiv (1 Tg); red).

concentrations in the Shiv(1 Tg) simulation compared to StratVOLC, between 316 and 251 hPa (tropopause levels at higher latitudes) on April 8 (the date of the eruption) and May 23, 2020 (date of first flight of the CAFE-EU campaign), thus illustrating only the effect of the Shiveluch eruption.

On April 8, shortly after the initial SO₂ injection, elevated SO₂ mixing ratios were confined to the vicinity of the volcano, with evidence of eastward transport. By May 23, approximately 1.5 months later, both SO₂ and sulfur aerosol were widely dispersed across the northern hemispheric extratropical UTLS, forming distinct filaments. This filament structure

Model results Figure 6.14 compares vertical profiles of observed and simulated sulfate concentrations along the CAFE-EU flight tracks for the three EMAC simulations. The simulation without any explosive volcanic eruptions (noVOLC) significantly underestimates aerosol concentrations in both the FT and UTLS. Including stratospheric volcanic eruptions leads (StratVOLC) leads to much better agreement above 10 km, highlighting the importance of the correct implementation of stratospheric volcanic eruptions in model simulations. In this case, the main contributors are the Raikoke (June/July 2019) and Taal (January 2020) eruptions. The additional inclusion of the Shiveluch eruption Shiv(1 Tg) further increases simulated FT sulfate concentration over Europe, leading to very close agreement with observations between 9 and 11 km.

The enhancement of stratospheric sulfur aerosol of tropospheric origin was observed only during individual flights, and not consistently throughout the CAFE-EU campaign. Therefore, we examined the horizontal distribution of the emitted SO₂ and the resulting sulfur aerosol, to better understand the transport. Figure 6.15 shows global maps of the enhancements in SO₂ and sulfur aerosol

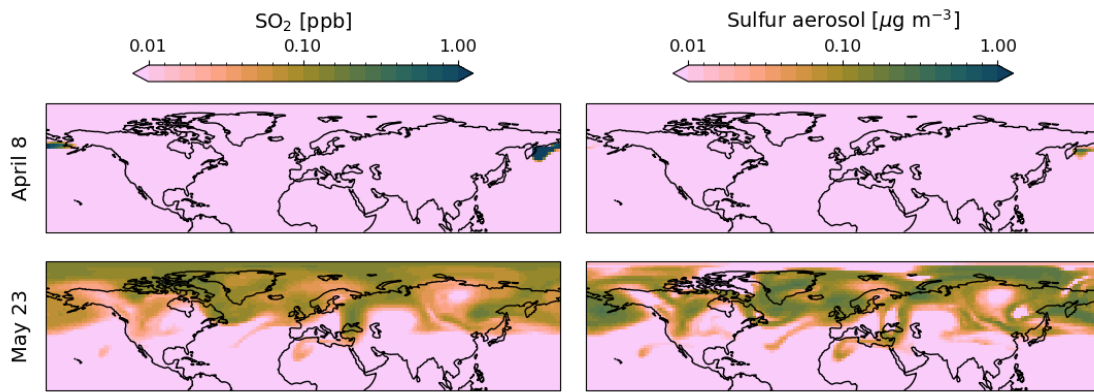


Figure 6.15 Global maps of the enhancement in SO_2 (left) and sulfur aerosol (right) concentrations from the Shiveluch eruption on April 8, 2020 (top; date of eruption) and May 23, 2020 (bottom; first CAFE-EU flight). Enhancements are calculated as the difference between the simulation including SO_2 emissions from Shiveluch and the StratVolc simulation.

likely explains why the enhancements were not observed in all CAFE-EU flights. While the model cannot fully resolve such structures over long transport timescales due to its limited horizontal resolution (approximately 200×200 km at the equator), the impact on the campaign-averaged profiles is well captured.

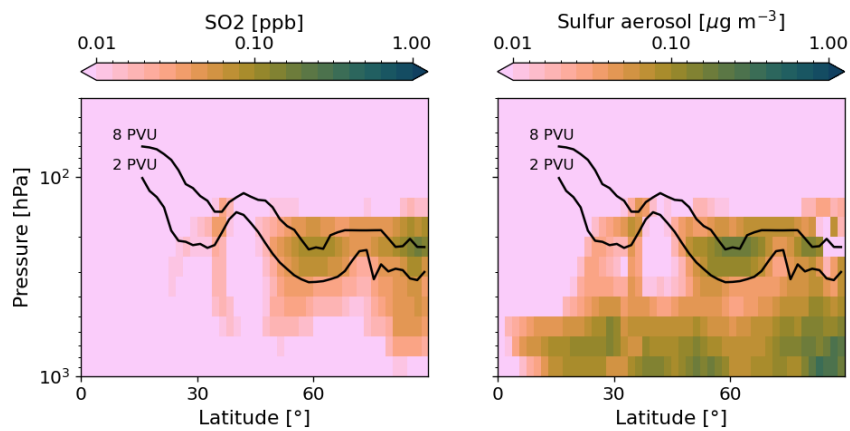


Figure 6.16 Latitudinal profiles of the enhancement in SO_2 mixing ratios (left) and sulfur aerosol concentrations (right) from the Shiveluch eruption on May 23 (first CAFE-EU flight) over the latitudinal extent of Europe, sampled during the CAFE-EU campaign (1°W to 14°E). Enhancements are calculated as the difference between the simulation including SO_2 emissions from Shiveluch and the StratVOLC simulation. The 2 PVU and 8 PVU lines indicate the extratropical transition layer (ExTL).

Figure 6.16 further illustrates the vertical extent of the enhancement in SO_2 and sulfur aerosol over Europe on May 23, 2020. The 2 PVU and 8 PVU lines mark the extratropical transition layer (ExTL). The enhancement is largely confined to this transition zone, where mixing between tropospheric and stratospheric air occurs, and does not extend much further

into the stratosphere. The observed increase in sulfur aerosol close to the Earth's surface is likely due to the re-evaporation of previously scavenged sulfur aerosol from higher altitudes.

The exact amount of SO₂ injected during the Shiveluch eruption remains uncertain, and the assumed emission of 1 Tg represents only an estimate. Therefore, while the results cannot provide a definitive quantification of the eruption's impact, they do support the conclusions of Joppe et al. [182], suggesting that even non-stratospheric volcanic eruptions can influence the lowermost stratosphere over extended periods through turbulent mixing and quasi-isentropic transport. Further studies will investigate the sensitivity of these results to variations in the injected mass and the vertical distribution of the injection altitude.

6.4 Further studies on ultrafine particles

The manuscript presented in Chapter 3 resulted in a first global UFP dataset, crucial for studies on health impacts of long-term exposure to UFP. Assessing the global health effects of UFP requires two steps. First, epidemiological studies are needed, assessing the outcomes of UFP exposure in sample populations. Previous studies have found associations between UFP exposure and pulmonary and systemic inflammation, heart rate variability, blood pressure, and increased mortality in general [183, 16]. A recent cohort study linked type 2 diabetes to long-term exposure to UFP using the dataset developed in Chapter 3 [184]. Second, exposure-response functions developed from these epidemiological studies can be applied on global UFP datasets to extrapolate the findings on a global scale. Section 6.4.2 presents a study on global health implications of exposure to UFP pollution, based on the dataset presented in Chapter 3 and a new high-resolution UFP dataset based on machine learning, presented in Sect. 6.4.1.

6.4.1 High-resolution global ultrafine particle concentrations

The work summarized in this Section is currently in review for Nature Scientific Data (Georgiades, Kohl et al., 2025) pending minor revisions. I provided observational datasets and advised the project with the expertise from global atmospheric UFP simulations.

The global UFP dataset developed in Chapter 3 at a horizontal resolution of 10 x 10 km is considered to be still relative coarse for exposure studies, as it cannot sufficiently resolve local UFP hotspots. In fact, it has been shown that UFP concentrations relax to background levels within 1 km, close to hotspots such as highways [102]. Global atmospheric chemistry models therefore currently do not have the capability to simulate UFP concentrations at such high resolution. To bridge this gap, we developed a high-resolution 1 x 1 km dataset of UFP concentrations using a machine learning approach. Specifically, we trained an XGBoost model to predict particle number concentrations (PNC), given the limited availability of direct UFP observations. UFP concentrations were then estimated from the predicted PNC values, using a global observation-based scaling factor.

The input feature set included indicators of human activity (population, build-up volume, degree of urbanisation, human settlement, and road network), air quality data (NO₂ concentration and PM_{2.5} concentration), emissions (black carbon, carbon dioxide and nitrogen

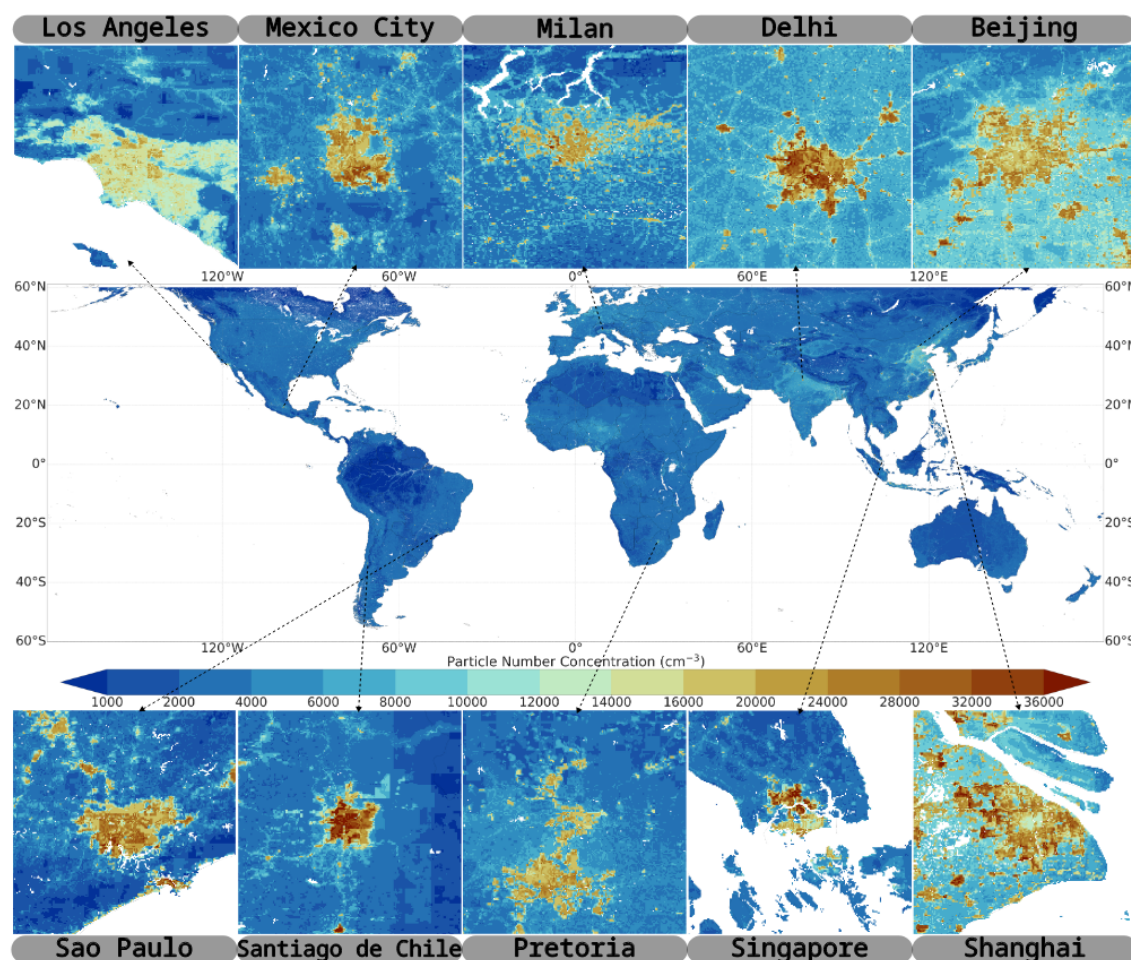


Figure 6.17 Global map of high-resolution PNC concentrations at the Earth's surface, with a zoom on urban regions across the globe. *The figure was prepared by Pantelis Georgiades, based on the newly created high-resolution PNC/UFP dataset*

oxides), and meteorological variables (temperature, BL height, and precipitation). All datasets were homogenized, and served as input for the XGBoost algorithm, along with a sample of the available PNC observations. Model results were validated using a 10-fold cross-validation, testing for spatial and temporal robustness. The resulting PNC concentrations, (from which UFP concentrations were estimated), are presented in Fig. 6.17.

6.4.2 Global health implications of exposure to ultrafine particle pollution

The work summarized in this Section is in review for Science (Lelieveld et al., 2025). I provided global UFP concentrations from an atmospheric chemistry model, and the basis for the sector contribution analysis. Additionally, I investigated the contribution of different species, and primary vs. secondary nature of UFP.

Several large-scale cohort studies, involving millions of individuals, have identified a significant association between exposure to UFP concentrations and both all-cause and disease-specific

mortality [185, 186, 187]. By averaging the reported hazard-ratios for all-cause mortality across these studies, and calculating the attributable fraction based on global population data, total and excess mortality rate were estimated globally. The underlying UFP dataset used in this analysis is based on the presented work in Sect. 6.4.1. We estimate that 1.98 (95% CI 1.11-3.07) million deaths annually can be attribute to UFP exposure, accounting for approximately 5% of global mortality from non-communicable diseases. This corresponds to a global mean excess mortality rate of 25.6 per 100,000 population per year (95% CI: 14.4–39.8).

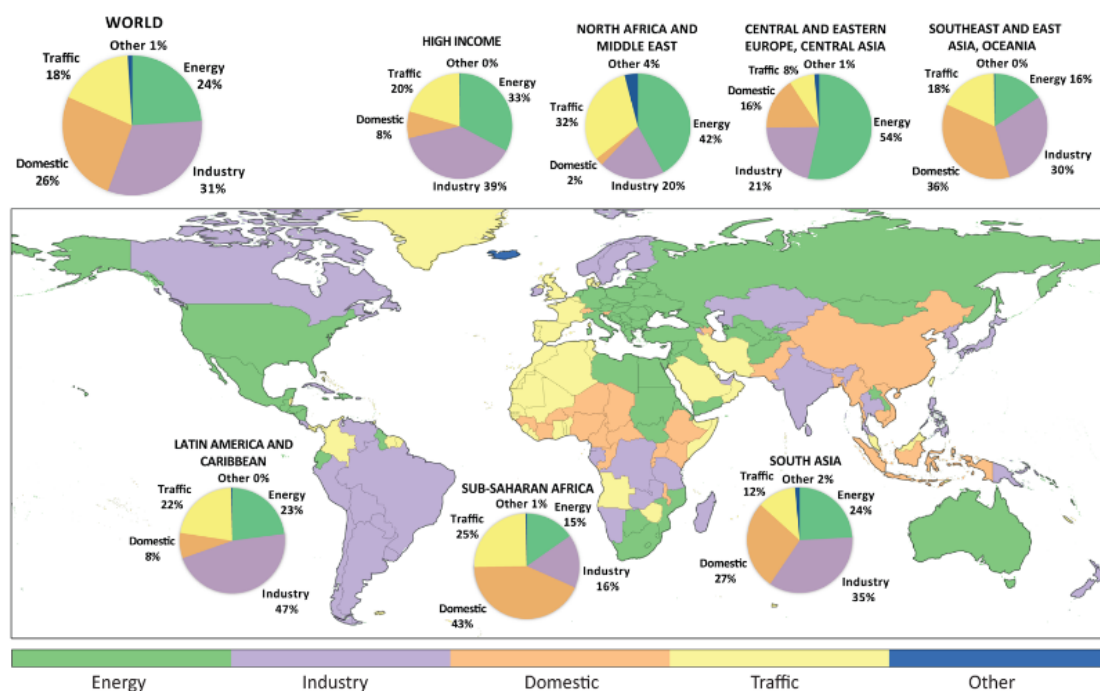


Figure 6.18 Main source sectors of UFP exposure per country (map), along with the importance of each sector for selected macro-regions (pie charts). *The figure was prepared by Jos Lelieveld based on sensitivity simulations with the UFP setup presented in Chapter 3, in which emissions sectors were singularly excluded.*

In addition, we performed sensitivity studies with the EMAC model, based on the simulation presented in Chapter 3, attributing UFP exposure to various source sectors. The main sources of UFP exposure are combustion processes involving fossil fuels. A detailed analysis on the contribution of the different source sectors is presented in Fig. 6.18.

7 CHAPTER

Conclusions

This thesis addressed the need of improved numerical simulations of atmospheric aerosols throughout all layers of the lower atmosphere at global scale, from the Earth's surface to the stratosphere, using the ECHAM5/MESSy2 Atmospheric Chemistry (EMAC) model.

In Chapter 3, a first global simulation of annually averaged UFP concentrations at the Earth's surface was performed, incorporating refined particle emission size distributions. These simulations were evaluated against ground-based observations, showing reasonable agreement in remote and rural regions. However, they failed to capture the observed UFP variability in grid boxes with high heterogeneity in population density. To address this, a novel observation-guided, emission-based downscaling approach was developed, enhancing the horizontal resolution of the UFP dataset from $1.875^\circ \times 1.875^\circ$ to $0.1^\circ \times 0.1^\circ$, and significantly improving agreement with observations (logarithmic correlation increased from 0.76 to 0.84 in non-remote regions; root mean squared logarithmic error decreased from 0.57 to 0.43). Simulated UFP concentrations span over three orders of magnitude, ranging from below 50 cm^{-3} over the remote ocean to more than $40,000 \text{ cm}^{-3}$ in polluted regions. The resulting dataset, which includes interannual variability and size-resolved UFP concentrations, has been made publicly available for use in health research and further research on UFP distributions.

In Sect. 6.4, machine learning techniques were applied to further enhance spatial resolution, producing a global UFP dataset at $1 \times 1 \text{ km}$ resolution. This high-resolution dataset was subsequently used to estimate the global health burden of UFP exposure, revealing an estimated 1.98 million excess deaths annually, corresponding to a mean excess mortality rate of 25.6 (95% CI: 14.4–39.8) deaths per 100,000 population per year. The underlying EMAC-simulated UFP dataset was also analysed to identify dominant sources of exposure, highlighting combustion processes involving fossil fuels as the main contributor for UFP concentration. Future work could focus on integrating global model outputs with machine learning approaches to further enhance the accuracy and robustness of high-resolution UFP concentration and source attribution datasets.

In Chapter 4, a new MESSy submodel for emissions from Explosive Volcanic ERuptions was presented, capable of handling particulate and gaseous emissions in flexible vertical shapes. Sensitivity studies revealed the importance of emitting a correct amount of SO_2 above the tropopause with appropriate altitude distribution, when modelling stratospheric volcanic eruptions, while horizontal position and emission timing are less important in the mid-term. Based on these insights and an existing volcanic emission inventory, a historic emission setup for stratospheric volcanic events was developed, and we advocate its inclusion

in future simulations focussing on the upper troposphere and the stratosphere. In Sect. 6.3, it was shown that including both stratospheric and large tropospheric eruptions can significantly improve agreement with observations of sulfate concentrations in the lowermost stratosphere. The new submodel also enables studies on the interaction of SO_2 and volcanic ash post-eruption, exploration of solar radiation modification scenarios, wildfire simulations, and analyses of atmospheric transport processes.

Chapter 5 builds on the preceding work, combining previous model configurations to develop one setup for aerosol simulations from the surface to the stratosphere, consistent with observations in most regions and layers of the lower atmosphere. This setup was used to produce a global aerosol distributions, chemical composition and particle number concentrations across the entire lower atmosphere. A key feature of the resulting distributions is a pronounced global minimum in aerosol mass concentrations between 400 and 200 hPa, allowing for frequent New Particle Formation in the free and upper troposphere, and separating the surface-influenced boundary layer and free troposphere from the stratosphere-influenced layers above. Stratospheric aerosol composition was found to consist predominantly (75–90%) of sulfur aerosol, while primary particles contribute only marginally, aligning better with recent observational evidence and in contrast to previous model studies. Secondary Organic Aerosol (SOA) significantly contributes to the aerosol burden in the lower tropical stratosphere, up to 90 to 70 hPa, forming a global band in the UTLS region.

Simulations based on the newly developed setup were used to investigate specific atmospheric processes, with a focus on the Asian monsoon system and the Amazon basin (Chapter 6). Results show that ammonia plays a key role in driving new particle formation and aerosol mass within the Asian Tropopause Aerosol Layer (ATAL), with the resulting particles persisting in the lowermost stratosphere for several months. The sensitivity of climate and atmospheric chemistry to perturbations in Amazonian biogenic volatile organic compound (BVOC) emissions was also explored, revealing a strong influence on local oxidation capacity, and a considerable impact on global climate.

Overall, this work addresses key research questions on global aerosol distributions from the surface to the stratosphere, and marks a step toward a more consistent and observation-based representation of aerosol burdens in the lower atmosphere. Building on the presented model setups by incorporating key aerosol feedbacks will support future studies on aerosol–climate–health interactions. Continued development, together with expanded observational datasets, can further improve the model’s ability to capture real-world aerosol distributions, advance our understanding of specific processes, and help reduce existing uncertainties.

Bibliography

- [1] S. Szopa, V. Naik, B. Adhikary, P. Artaxo, T. Berntsen, W. Collins, S. Fuzzi, L. Gallardo, A. Kiendler-Scharr, Z. Klimont, H. Liao, N. Unger, and P. Zanis. “Short-Lived Climate Forcers”. *Climate Change 2021: The Physical Science Basis. Contribution of Working Group I to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change*. Cambridge, United Kingdom and New York, NY, USA: Cambridge University Press, 2021, pp. 817–922. DOI: 10.1017/9781009157896.008.
- [2] C. J. Murray, A. Y. Aravkin, P. Zheng, C. Abbafati, K. M. Abbas, M. Abbasi-Kangevari, F. Abd-Allah, A. Abdelalim, M. Abdollahi, I. Abdollahpour, et al. “Global burden of 87 risk factors in 204 countries and territories, 1990–2019: a systematic analysis for the Global Burden of Disease Study 2019”. *The lancet* 396.10258 (2020), pp. 1223–1249. DOI: 10.1016/S0140-6736(20)30752-2.
- [3] C. A. P. III and D. W. Dockery. “Health Effects of Fine Particulate Air Pollution: Lines that Connect”. *Journal of the Air & Waste Management Association* 56.6 (2006), pp. 709–742. DOI: 10.1080/10473289.2006.10464485.
- [4] J. Lelieveld, J. S. Evans, M. Fnais, D. Giannadaki, and A. Pozzer. “The contribution of outdoor air pollution sources to premature mortality on a global scale”. *Nature* 525.7569 (2015), pp. 367–371. DOI: 10.1038/nature15371.
- [5] R. Rückerl, A. Schneider, S. Breitner, J. Cyrys, and A. Peters. “Health effects of particulate air pollution: a review of epidemiological evidence”. *Inhalation toxicology* 23.10 (2011), pp. 555–592. DOI: 10.3109/08958378.2011.593587.
- [6] V. Ramanathan, P. J. Crutzen, J. T. Kiehl, and D. Rosenfeld. “Aerosols, Climate, and the Hydrological Cycle”. *Science* 294.5549 (2001), pp. 2119–2124. DOI: 10.1126/science.1064034.
- [7] U. Lohmann and J. Feichter. “Global indirect aerosol effects: a review”. *Atmospheric Chemistry and Physics* 5.3 (2005), pp. 715–737. DOI: 10.5194/acp-5-715-2005.
- [8] P. Wu, N. Christidis, and P. Stott. “Anthropogenic impact on Earth’s hydrological cycle”. *Nature Climate Change* 3.9 (2013), pp. 807–810. DOI: 10.1038/nclimate1932.
- [9] O. Boucher, D. Randall, P. Artaxo, C. Bretherton, G. Feingold, P. Forster, V.-M. Kerminen, Y. Kondo, H. Liao, U. Lohmann, et al. “Clouds and aerosols”. *Climate change 2013: The physical science basis. Contribution of working group I to the fifth assessment report of the intergovernmental panel on climate change*. Cambridge, United Kingdom and New York, NY, USA: Cambridge University Press, 2013, pp. 571–657.

- [10] J. E. Klobas, D. M. Wilmouth, D. K. Weisenstein, J. G. Anderson, and R. J. Salawitch. “Ozone depletion following future volcanic eruptions”. *Geophysical Research Letters* 44.14 (2017), pp. 7490–7499.
- [11] X. Tie and G. Brasseur. “The response of stratospheric ozone to volcanic eruptions: Sensitivity to atmospheric chlorine loading”. *Geophysical Research Letters* 22.22 (1995), pp. 3035–3038. DOI: 10.1029/95GL03057.
- [12] D. Watson-Parris, N. Bellouin, L. T. Deaconu, N. A. J. Schutgens, M. Yoshioka, L. A. Regayre, K. J. Pringle, J. S. Johnson, C. J. Smith, K. S. Carslaw, and P. Stier. “Constraining Uncertainty in Aerosol Direct Forcing”. *Geophysical Research Letters* 47.9 (2020). e2020GL087141 2020GL087141, e2020GL087141. DOI: <https://doi.org/10.1029/2020GL087141>.
- [13] O. Boucher. “Atmospheric Aerosols”. *Atmospheric Aerosols: Properties and Climate Impacts*. Dordrecht: Springer Netherlands, 2015, pp. 9–24. ISBN: 978-94-017-9649-1. DOI: 10.1007/978-94-017-9649-1_2.
- [14] C. Tomasi and A. Lupi. “Primary and Secondary Sources of Atmospheric Aerosol”. *Atmospheric Aerosols*. John Wiley & Sons, Ltd, 2017. Chap. 1, pp. 1–86. ISBN: 9783527336449. DOI: <https://doi.org/10.1002/9783527336449.ch1>.
- [15] J. H. Seinfeld and S. N. Pandis. *Atmospheric chemistry and physics: from air pollution to climate change*. John Wiley & Sons, 2016. ISBN: 978-1-118-94740-1.
- [16] B. Ostro, J. Hu, D. Goldberg, P. Reynolds, A. Hertz, L. Bernstein, and M. J. Kleeman. “Associations of mortality with long-term exposures to fine and ultrafine particles, species and sources: results from the California Teachers Study Cohort”. *Environmental health perspectives* 123.6 (2015), pp. 549–556. DOI: 10.1289/ehp.1408565.
- [17] G. S. Downward, E. J. van Nunen, J. Kerckhoffs, P. Vineis, B. Brunekreef, J. M. Boer, K. P. Messier, A. Roy, W. M. M. Verschuren, Y. T. van der Schouw, et al. “Long-term exposure to ultrafine particles and incidence of cardiovascular and cerebrovascular disease in a prospective study of a Dutch cohort”. *Environmental Health Perspectives* 126.12 (2018), p. 127007. DOI: 10.1289/EHP3047.
- [18] D. E. Schraufnagel. “The health effects of ultrafine particles”. *Exp. Mol. Med.* 52 (2020), pp. 311–317. ISSN: 2092-6413. DOI: 10.1038/s12276-020-0403-3.
- [19] G. Hong and Y.-K. Jee. “Special issue on ultrafine particles: where are they from and how do they affect us?” *Exp. Mol. Med.* 52 (2020), pp. 309–310. ISSN: 2092-6413. DOI: 10.1038/s12276-020-0395-z.
- [20] S. Kremser, L. W. Thomason, M. von Hobe, M. Hermann, T. Deshler, C. Timmreck, M. Toohey, A. Stenke, J. P. Schwarz, R. Weigel, S. Fueglistaler, F. J. Prata, J.-P. Vernier, H. Schlager, J. E. Barnes, J.-C. Antuña-Marrero, D. Fairlie, M. Palm, E. Mahieu, J. Notholt, M. Rex, C. Bingen, F. Vanhellemont, A. Bourassa, J. M. C. Plane, D. Klocke, S. A. Carn, L. Clarisse, T. Trickl, R. Neely, A. D. James, L. Rieger, J. C. Wilson, and B. Meland. “Stratospheric aerosol—Observations, processes, and impact on climate”. *Reviews of Geophysics* 54.2 (), pp. 278–335. DOI: <https://doi.org/10.1002/2015RG000511>.

- [21] P. J. Crutzen. “The “Anthropocene””. *Earth System Science in the Anthropocene*. Ed. by E. Ehlers and T. Krafft. Berlin, Heidelberg: Springer Berlin Heidelberg, 2006, pp. 13–18. ISBN: 978-3-540-26590-0. DOI: 10.1007/3-540-26590-2_3.
- [22] F. K. Lutgens, E. J. Tarbuck, and D. Tusa. *The atmosphere*. Pearson, 2013. ISBN: 978-1-292-05421-6.
- [23] R. M. Goody and Y. L. Yung. *Atmospheric radiation: theoretical basis*. Oxford university press, 1995. ISBN: 978-0-195-05134-6.
- [24] J. M. Wallace and P. V. Hobbs. *Atmospheric science: an introductory survey*. Vol. 92. Elsevier, 2006. ISBN: 978-0-127-32951-2.
- [25] K.-N. Liou. *Radiation and cloud processes in the atmosphere: theory, observation, and modeling*. Oxford university press, 1992. ISBN: 0-19-504910-1.
- [26] H. Jacobowitz and R. J. Tighe. “The Earth Radiation Budget derived from the NIMBUS 7 ERB Experiment”. *Journal of Geophysical Research: Atmospheres* 89.D4 (1984), pp. 4997–5010. DOI: <https://doi.org/10.1029/JD089iD04p04997>.
- [27] J. R. Holton and G. J. Hakim. *An introduction to dynamic meteorology*. Vol. 88. Academic press, 2013. ISBN: 978-0-12-384866-6.
- [28] J. Garratt. “Review: the atmospheric boundary layer”. *Earth-Science Reviews* 37.1 (1994), pp. 89–134. ISSN: 0012-8252. DOI: [https://doi.org/10.1016/0012-8252\(94\)90026-4](https://doi.org/10.1016/0012-8252(94)90026-4).
- [29] H. Kraus. *Die Atmosphäre der Erde: Eine Einführung in die Meteorologie*. Springer-Verlag, 2007. Chap. 10. ISBN: 3-540-20656-6.
- [30] A. Gettelman, P. Hoor, L. L. Pan, W. J. Randel, M. I. Hegglin, and T. Birner. “The extratropical upper troposphere and lower stratosphere”. *Reviews of Geophysics* 49.3 (2011). DOI: <https://doi.org/10.1029/2011RG000355>.
- [31] G. P. Brasseur and S. Solomon. *Aeronomy of the middle atmosphere: Chemistry and physics of the stratosphere and mesosphere*. Vol. 32. Springer Science & Business Media, 2005. ISBN: 978-1-4020-3284-4.
- [32] S. Fueglistaler, A. E. Dessler, T. J. Dunkerton, I. Folkins, Q. Fu, and P. W. Mote. “Tropical tropopause layer”. *Reviews of Geophysics* 47.1 (2009). DOI: <https://doi.org/10.1029/2008RG000267>.
- [33] J. R. Holton, P. H. Haynes, M. E. McIntyre, A. R. Douglass, R. B. Rood, and L. Pfister. “Stratosphere-troposphere exchange”. *Reviews of Geophysics* 33.4 (1995), pp. 403–439. DOI: <https://doi.org/10.1029/95RG02097>.
- [34] F. J. Beron-Vera, M. G. Brown, M. J. Olascoaga, I. I. Rypina, H. Koçak, and I. A. Udovydchenkov. “Zonal Jets as Transport Barriers in Planetary Atmospheres”. *Journal of the Atmospheric Sciences* 65.10 (2008), pp. 3316–3326. DOI: 10.1175/2008JAS2579.1.

- [35] N. Butchart. “The Brewer-Dobson circulation”. *Reviews of Geophysics* 52.2 (2014), pp. 157–184. DOI: <https://doi.org/10.1002/2013RG000448>.
- [36] M. P. Baldwin, L. J. Gray, T. J. Dunkerton, K. Hamilton, P. H. Haynes, W. J. Randel, J. R. Holton, M. J. Alexander, I. Hirota, T. Horinouchi, D. B. A. Jones, J. S. Kinniersley, C. Marquardt, K. Sato, and M. Takahashi. “The quasi-biennial oscillation”. *Reviews of Geophysics* 39.2 (2001), pp. 179–229. DOI: <https://doi.org/10.1029/1999RG000073>.
- [37] D. M. Murphy, K. D. Froyd, J. P. Schwarz, and J. C. Wilson. “Observations of the chemical composition of stratospheric aerosol particles”. *Quarterly Journal of the Royal Meteorological Society* 140.681 (2014), pp. 1269–1278. DOI: <https://doi.org/10.1002/qj.2213>.
- [38] M. Kerker. *The scattering of light and other electromagnetic radiation: physical chemistry: a series of monographs*. Vol. 16. Academic press, 2013.
- [39] C. F. Bohren and D. R. Huffman. *Absorption and scattering of light by small particles*. John Wiley & Sons, 2008.
- [40] H. R. Pruppacher, J. D. Klett, and P. K. Wang. *Microphysics of clouds and precipitation*. 1998.
- [41] S. A. Twomey, M. Piepgrass, and T. L. Wolfe. “An assessment of the impact of pollution on global cloud albedo”. *Tellus B* 36B.5 (1984), pp. 356–366. DOI: <https://doi.org/10.1111/j.1600-0889.1984.tb00254.x>.
- [42] R. Pincus and M. B. Baker. “Effect of precipitation on the albedo susceptibility of clouds in the marine boundary layer”. *Nature* 372.6503 (1994), pp. 250–252. DOI: [10.1038/372250a0](https://doi.org/10.1038/372250a0).
- [43] B. A. Albrecht. “Aerosols, Cloud Microphysics, and Fractional Cloudiness”. *Science* 245.4923 (1989), pp. 1227–1230. DOI: [10.1126/science.245.4923.1227](https://doi.org/10.1126/science.245.4923.1227).
- [44] J. Kirkby, J. Curtius, J. Almeida, E. Dunne, J. Duplissy, S. Ehrhart, A. Franchin, S. Gagné, L. Ickes, A. Kürten, et al. “Role of sulphuric acid, ammonia and galactic cosmic rays in atmospheric aerosol nucleation”. *Nature* 476.7361 (2011), pp. 429–433. DOI: [10.1038/nature10343](https://doi.org/10.1038/nature10343).
- [45] J. Kirkby, A. Amorim, U. Baltensperger, K. S. Carslaw, T. Christoudias, J. Curtius, N. M. Donahue, I. E. Haddad, R. C. Flagan, H. Gordon, et al. “Atmospheric new particle formation from the CERN CLOUD experiment”. *Nature Geoscience* 16.11 (2023), pp. 948–957. DOI: [10.1038/s41561-023-01305-0](https://doi.org/10.1038/s41561-023-01305-0).
- [46] A. R. Ravishankara. “Heterogeneous and Multiphase Chemistry in the Troposphere”. *Science* 276.5315 (1997), pp. 1058–1065. DOI: [10.1126/science.276.5315.1058](https://doi.org/10.1126/science.276.5315.1058).
- [47] D. J. Jacob. “Heterogeneous chemistry and tropospheric ozone”. *Atmospheric Environment* 34.12 (2000), pp. 2131–2159. ISSN: 1352-2310. DOI: [https://doi.org/10.1016/S1352-2310\(99\)00462-8](https://doi.org/10.1016/S1352-2310(99)00462-8).

- [48] P. W. Atkins, J. De Paula, and J. Keeler. *Atkins' physical chemistry*. Oxford university press, 2023. ISBN: 978-0-19-884781-6.
- [49] R. M. Felder, R. W. Rousseau, and L. G. Bullard. *Elementary principles of chemical processes*. John Wiley & Sons, 2020. ISBN: 978-1-11-949863-6.
- [50] A. H. Goldstein and E. Galbally. “Known and Unexplored Organic Constituents in the Earth’s Atmosphere”. *Environmental Science & Technology* 41.5 (2007). PMID: 17396635, pp. 1514–1521. DOI: 10.1021/es072476p.
- [51] N. M. Donahue, S. A. Epstein, S. N. Pandis, and A. L. Robinson. “A two-dimensional volatility basis set: 1. organic-aerosol mixing thermodynamics”. *Atmospheric Chemistry and Physics* 11.7 (2011), pp. 3303–3318. DOI: 10.5194/acp-11-3303-2011.
- [52] D. Stolzenburg, M. Wang, M. Schervish, and N. M. Donahue. “Tutorial: Dynamic organic growth modeling with a volatility basis set”. *Journal of Aerosol Science* 166 (2022), p. 106063. ISSN: 0021-8502. DOI: <https://doi.org/10.1016/j.jaerosci.2022.106063>.
- [53] B. N. Murphy, N. M. Donahue, A. L. Robinson, and S. N. Pandis. “A naming convention for atmospheric organic aerosol”. *Atmospheric Chemistry and Physics* 14.11 (2014), pp. 5825–5839. DOI: 10.5194/acp-14-5825-2014. URL: <https://acp.copernicus.org/articles/14/5825/2014/>.
- [54] J. Aitken. “On the number of dust particles in the atmosphere”. *Nature* 37.957 (1888), pp. 428–430.
- [55] J. Aitken. “On Improvements in the Apparatus for Counting the Dust Particles in the Atmosphere”. *Proceedings of the Royal Society of Edinburgh* 16 (1890), pp. 135–172. DOI: 10.1017/S0370164600006222.
- [56] L. Rayleigh. “XXXIV. On the transmission of light through an atmosphere containing small particles in suspension, and on the origin of the blue of the sky”. *The London, Edinburgh, and Dublin Philosophical Magazine and Journal of Science* 47.287 (1899), pp. 375–384. DOI: 10.1080/14786449908621276.
- [57] J. Strutt. “XV. On the light from the sky, its polarization and colour”. *The London, Edinburgh, and Dublin Philosophical Magazine and Journal of Science* 41.271 (1871), pp. 107–120. DOI: 10.1080/14786447108640452.
- [58] V. Bjerknes. “Das Problem der Wettervorhersage, betrachtet vom Standpunkte der Mechanik und der Physik”. *Meteor. Z.* 21 (1904), pp. 1–7.
- [59] L. F. Richardson. *Weather prediction by numerical process*. University Press, 1922.
- [60] P. Lynch. “The origins of computer weather prediction and climate modeling”. *Journal of Computational Physics* 227.7 (2008). Predicting weather, climate and extreme events, pp. 3431–3444. ISSN: 0021-9991. DOI: <https://doi.org/10.1016/j.jcp.2007.02.034>.
- [61] P. Jöckel, H. Tost, A. Pozzer, C. Brühl, J. Buchholz, L. Ganzeveld, P. Hoor, A. Kerkweg, M. G. Lawrence, R. Sander, B. Steil, G. Stiller, M. Tanarhte, D. Taraborrelli,

- J. van Aardenne, and J. Lelieveld. “The atmospheric chemistry general circulation model ECHAM5/MESSy1: consistent simulation of ozone from the surface to the mesosphere”. *Atmospheric Chemistry and Physics* 6.12 (2006), pp. 5067–5104. DOI: 10.5194/acp-6-5067-2006.
- [62] P. Jöckel, A. Kerkweg, A. Pozzer, R. Sander, H. Tost, H. Riede, A. Baumgaertner, S. Gromov, and B. Kern. “Development cycle 2 of the Modular Earth Submodel System (MESSy2)”. *Geoscientific Model Development* 3.2 (2010), pp. 717–752. DOI: 10.5194/gmd-3-717-2010.
- [63] P. Jöckel, R. Sander, A. Kerkweg, H. Tost, and J. Lelieveld. “Technical Note: The Modular Earth Submodel System (MESSy) - a new approach towards Earth System Modeling”. *Atmospheric Chemistry and Physics* 5.2 (2005), pp. 433–444. DOI: 10.5194/acp-5-433-2005.
- [64] E. Roeckner, G. Bäuml, L. Bonaventura, R. Brokopf, M. Esch, M. Giorgetta, S. Hagemann, I. Kirchner, L. Kornbluh, E. Manzini, et al. *The atmospheric general circulation model ECHAM 5. PART I: Model description*. Tech. rep. Max-Planck-Institut für Meteorologie, 2003. DOI: 10.17617/2.995269.
- [65] P. Jöckel, A. Kerkweg, J. Buchholz-Dietsch, H. Tost, R. Sander, and A. Pozzer. “Technical Note: Coupling of chemical processes with the Modular Earth Submodel System (MESSy) submodel TRACER”. *Atmospheric Chemistry and Physics* 8.6 (2008), pp. 1677–1687. DOI: 10.5194/acp-8-1677-2008.
- [66] R. Eichinger and P. Jöckel. “The generic MESSy submodel TENDENCY (v1.0) for process-based analyses in Earth system models”. *Geoscientific Model Development* 7.4 (2014), pp. 1573–1582. DOI: 10.5194/gmd-7-1573-2014.
- [67] A. Kerkweg, C. Hofmann, P. Jöckel, M. Mertens, and G. Pante. “The on-line coupled atmospheric chemistry model system MECO(n) – Part 5: Expanding the Multi-Model-Driver (MMD v2.0) for 2-way data exchange including data interpolation via GRID (v1.0)”. *Geoscientific Model Development* 11.3 (2018), pp. 1059–1076. DOI: 10.5194/gmd-11-1059-2018.
- [68] R. Sander, A. Baumgaertner, D. Cabrera-Perez, F. Frank, S. Gromov, J.-U. Groö, H. Harder, V. Huijnen, P. Jöckel, V. A. Karydis, K. E. Niemeyer, A. Pozzer, H. Riede, M. G. Schultz, D. Taraborrelli, and S. Tauer. “The community atmospheric chemistry box model CAABA/MECCA-4.0”. *Geoscientific Model Development* 12.4 (2019), pp. 1365–1385. DOI: 10.5194/gmd-12-1365-2019.
- [69] R. Sander, P. Jöckel, O. Kirner, A. T. Kunert, J. Landgraf, and A. Pozzer. “The photolysis module JVAL-14, compatible with the MESSy standard, and the JVal PreProcessor (JVPP)”. *Geoscientific Model Development* 7.6 (2014), pp. 2653–2662. DOI: 10.5194/gmd-7-2653-2014.
- [70] A. Kerkweg, R. Sander, H. Tost, and P. Jöckel. “Technical note: Implementation of prescribed (OFFLEM), calculated (ONLEM), and pseudo-emissions (TNUDGE) of chemical species in the Modular Earth Submodel System (MESSy)”. *Atmospheric Chemistry and Physics* 6.11 (2006), pp. 3603–3609. DOI: 10.5194/acp-6-3603-2006.

- [71] K. J. Pringle, H. Tost, S. Metzger, B. Steil, D. Giannadaki, A. Nenes, C. Fountoukis, P. Stier, E. Vignati, and J. Lelieveld. “Description and evaluation of GMXe: a new aerosol submodel for global simulations (v1)”. *Geosci. Model Dev.* 3.2 (2010), pp. 391–412. DOI: 10.5194/gmd-3-391-2010.
- [72] E. Vignati, J. Wilson, and P. Stier. “M7: An efficient size-resolved aerosol microphysics module for large-scale aerosol transport models”. *Journal of Geophysical Research: Atmospheres* 109.D22 (2004). DOI: <https://doi.org/10.1029/2003JD004485>.
- [73] C. Fountoukis and A. Nenes. “ISORROPIA II: a computationally efficient thermodynamic equilibrium model for K^+ – Ca^{2+} – Mg^{2+} – NH_4^+ – Na^+ – SO_4^{2-} – NO_3^- – Cl^- – H_2O aerosols”. *Atmospheric Chemistry and Physics* 7.17 (2007), pp. 4639–4659. DOI: 10.5194/acp-7-4639-2007.
- [74] A. P. Tsimpidi, V. A. Karydis, A. Pozzer, S. N. Pandis, and J. Lelieveld. “ORACLE (v1.0): module to simulate the organic aerosol composition and evolution in the atmosphere”. *Geoscientific Model Development* 7.6 (2014), pp. 3153–3172. DOI: 10.5194/gmd-7-3153-2014.
- [75] H. Vehkamäki, M. Kulmala, I. Napari, K. E. J. Lehtinen, C. Timmreck, M. Noppel, and A. Laaksonen. “An improved parameterization for sulfuric acid–water nucleation rates for tropospheric and stratospheric conditions”. *Journal of Geophysical Research: Atmospheres* 107.D22 (2002), AAC 3-1-AAC 3–10. DOI: <https://doi.org/10.1029/2002JD002184>.
- [76] S. Ehrhart, E. M. Dunne, H. E. Manninen, T. Nieminen, J. Lelieveld, and A. Pozzer. “Two new submodels for the Modular Earth Submodel System (MESSy): New Aerosol Nucleation (NAN) and small ions (IONS) version 1.0”. *Geoscientific Model Development* 11.12 (2018), pp. 4987–5001. DOI: 10.5194/gmd-11-4987-2018.
- [77] E. M. Dunne, H. Gordon, A. Kürten, J. Almeida, J. Duplissy, C. Williamson, I. K. Ortega, K. J. Pringle, A. Adamov, U. Baltensperger, P. Barmet, F. Benduhn, F. Bianchi, M. Breitenlechner, A. Clarke, J. Curtius, J. Dommen, N. M. Donahue, S. Ehrhart, R. C. Flagan, A. Franchin, R. Guida, J. Hakala, A. Hansel, M. Heinritzi, T. Jokinen, J. Kangasluoma, J. Kirkby, M. Kulmala, A. Kupc, M. J. Lawler, K. Lehtipalo, V. Makhmutov, G. Mann, S. Mathot, J. Merikanto, P. Miettinen, A. Nenes, A. Onnela, A. Rap, C. L. S. Reddington, F. Riccobono, N. A. D. Richards, M. P. Rissanen, L. Rondo, N. Sarnela, S. Schobesberger, K. Sengupta, M. Simon, M. Sipilä, J. N. Smith, Y. Stozkhov, A. Tomé, J. Tröstl, P. E. Wagner, D. Wimmer, P. M. Winkler, D. R. Worsnop, and K. S. Carslaw. “Global atmospheric particle formation from CERN CLOUD measurements”. *Science* 354.6316 (2016), pp. 1119–1124. DOI: 10.1126/science.aaf2649.
- [78] J. Kirkby, J. Duplissy, K. Sengupta, C. Frege, H. Gordon, C. Williamson, M. Heinritzi, M. Simon, C. Yan, J. Almeida, et al. “Ion-induced nucleation of pure biogenic particles”. *Nature* 533.7604 (2016), pp. 521–526. DOI: 10.1038/nature17953.
- [79] F. Riccobono, S. Schobesberger, C. E. Scott, J. Dommen, I. K. Ortega, L. Rondo, J. Almeida, A. Amorim, F. Bianchi, M. Breitenlechner, A. David, A. Downard, E. M. Dunne, J. Duplissy, S. Ehrhart, R. C. Flagan, A. Franchin, A. Hansel, H. Junninen, M.

- Kajos, H. Keskinen, A. Kupc, A. Kürten, A. N. Kvashin, A. Laaksonen, K. Lehtipalo, V. Makhmutov, S. Mathot, T. Nieminen, A. Onnela, T. Petäjä, A. P. Praplan, F. D. Santos, S. Schallhart, J. H. Seinfeld, M. Sipilä, D. V. Spracklen, Y. Stozhkov, F. Stratmann, A. Tomé, G. Tsagkogeorgas, P. Vaattovaara, Y. Viisanen, A. Vrtala, P. E. Wagner, E. Weingartner, H. Wex, D. Wimmer, K. S. Carslaw, J. Curtius, N. M. Donahue, J. Kirkby, M. Kulmala, D. R. Worsnop, and U. Baltensperger. “Oxidation Products of Biogenic Emissions Contribute to Nucleation of Atmospheric Particles”. *Science* 344.6185 (2014), pp. 717–721. DOI: 10.1126/science.1243527.
- [80] H. Tost, P. Jöckel, and J. Lelieveld. “Influence of different convection parameterisations in a GCM”. *Atmospheric Chemistry and Physics* 6.12 (2006), pp. 5475–5493. DOI: 10.5194/acp-6-5475-2006.
- [81] A. Kerkweg, J. Buchholz, L. Ganzeveld, A. Pozzer, H. Tost, and P. Jöckel. “Technical Note: An implementation of the dry removal processes DRY DEPosition and SEDImentation in the Modular Earth Submodel System (MESSy)”. *Atmospheric Chemistry and Physics* 6.12 (2006), pp. 4617–4632. DOI: 10.5194/acp-6-4617-2006.
- [82] S. Dietmüller, P. Jöckel, H. Tost, M. Kunze, C. Gellhorn, S. Brinkop, C. Frömming, M. Ponater, B. Steil, A. Lauer, and J. Hendricks. “A new radiation infrastructure for the Modular Earth Submodel System (MESSy, based on version 2.51)”. *Geoscientific Model Development* 9.6 (2016), pp. 2209–2222. DOI: 10.5194/gmd-9-2209-2016.
- [83] A. Pozzer, P. Jöckel, R. Sander, J. Williams, L. Ganzeveld, and J. Lelieveld. “Technical Note: The MESSy-submodel AIRSEA calculating the air-sea exchange of chemical species”. *Atmospheric Chemistry and Physics* 6.12 (2006), pp. 5435–5444. DOI: 10.5194/acp-6-5435-2006.
- [84] J. W. Kaiser, A. Heil, M. O. Andreae, A. Benedetti, N. Chubarova, L. Jones, J.-J. Morcrette, M. Razinger, M. G. Schultz, M. Suttie, and G. R. van der Werf. “Biomass burning emissions estimated with a global fire assimilation system based on observed fire radiative power”. *Biogeosciences* 9.1 (2012), pp. 527–554. DOI: 10.5194/bg-9-527-2012.
- [85] M. O. Andreae, A. Afchine, R. Albrecht, B. A. Holanda, P. Artaxo, H. M. J. Barbosa, S. Borrmann, M. A. Cecchini, A. Costa, M. Dollner, D. Fütterer, E. Järvinen, T. Jurkat, T. Klimach, T. Konemann, C. Knote, M. Krämer, T. Krisna, L. A. T. Machado, S. Mertes, A. Minikin, C. Pöhlker, M. L. Pöhlker, U. Pöschl, D. Rosenfeld, D. Sauer, H. Schlager, M. Schnaiter, J. Schneider, C. Schulz, A. Spanu, V. B. Sperling, C. Voigt, A. Walser, J. Wang, B. Weinzierl, M. Wendisch, and H. Ziereis. “Aerosol characteristics and particle production in the upper troposphere over the Amazon Basin”. *Atmospheric Chemistry and Physics* 18.2 (2018), pp. 921–961. DOI: 10.5194/acp-18-921-2018.
- [86] M. Tiedtke. “A Comprehensive Mass Flux Scheme for Cumulus Parameterization in Large-Scale Models”. *Monthly Weather Review* 117.8 (1989), pp. 1779–1800. DOI: 10.1175/1520-0493(1989)117<1779:ACMFSF>2.0.CO;2.
- [87] H. Tost, M. G. Lawrence, C. Brühl, P. Jöckel, T. G. Team, and T. S.-O.-D. Team. “Uncertainties in atmospheric chemistry modelling due to convection parameterisa-

- tions and subsequent scavenging”. *Atmospheric Chemistry and Physics* 10.4 (2010), pp. 1931–1951. DOI: 10.5194/acp-10-1931-2010.
- [88] C. O. Hines. “Doppler-spread parameterization of gravity-wave momentum deposition in the middle atmosphere. Part 1: Basic formulation”. *Journal of Atmospheric and Solar-Terrestrial Physics* 59.4 (1997), pp. 371–386. ISSN: 1364-6826. DOI: [https://doi.org/10.1016/S1364-6826\(96\)00079-X](https://doi.org/10.1016/S1364-6826(96)00079-X).
- [89] C. O. Hines. “Doppler-spread parameterization of gravity-wave momentum deposition in the middle atmosphere. Part 2: Broad and quasi monochromatic spectra, and implementation”. *Journal of Atmospheric and Solar-Terrestrial Physics* 59.4 (1997), pp. 387–400. ISSN: 1364-6826. DOI: [https://doi.org/10.1016/S1364-6826\(96\)00080-6](https://doi.org/10.1016/S1364-6826(96)00080-6).
- [90] V. Grewe, D. Brunner, M. Dameris, J. Grenfell, R. Hein, D. Shindell, and J. Staehelin. “Origin and variability of upper tropospheric nitrogen oxides and ozone at northern mid-latitudes”. *Atmospheric Environment* 35.20 (2001), pp. 3421–3433. DOI: 10.1016/S1352-2310(01)00134-0.
- [91] A. B. Guenther, X. Jiang, C. L. Heald, T. Sakulyanontvittaya, T. Duhl, L. K. Emmons, and X. Wang. “The Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1): an extended and updated framework for modeling biogenic emissions”. *Geoscientific Model Development* 5.6 (2012), pp. 1471–1492. DOI: 10.5194/gmd-5-1471-2012.
- [92] A. van Donkelaar, R. V. Martin, M. Brauer, R. Kahn, R. Levy, C. Verduzco, and P. J. Villeneuve. “Global Estimates of Ambient Fine Particulate Matter Concentrations from Satellite-Based Aerosol Optical Depth: Development and Application”. *Environmental Health Perspectives* 118.6 (2010), pp. 847–855. DOI: 10.1289/ehp.0901623.
- [93] A. van Donkelaar, R. V. Martin, M. Brauer, N. C. Hsu, R. A. Kahn, R. C. Levy, A. Lyapustin, A. M. Sayer, and D. M. Winker. “Global Estimates of Fine Particulate Matter using a Combined Geophysical-Statistical Method with Information from Satellites, Models, and Monitors”. *Environmental Science & Technology* 50.7 (2016). PMID: 26953851, pp. 3762–3772. DOI: 10.1021/acs.est.5b05833.
- [94] M. Brauer, M. Amann, R. T. Burnett, A. Cohen, F. Dentener, M. Ezzati, S. B. Henderson, M. Krzyzanowski, R. V. Martin, R. Van Dingenen, A. van Donkelaar, and G. D. Thurston. “Exposure Assessment for Estimation of the Global Burden of Disease Attributable to Outdoor Air Pollution”. *Environmental Science & Technology* 46.2 (2012). PMID: 22148428, pp. 652–660. DOI: 10.1021/es2025752.
- [95] J.-P. Putaud, F. Raes, R. Van Dingenen, E. Brüggemann, M.-C. Facchini, S. Decesari, S. Fuzzi, R. Gehrig, C. Hüglin, P. Laj, G. Lorbeer, W. Maenhaut, N. Mihalopoulos, K. Müller, X. Querol, S. Rodriguez, J. Schneider, G. Spindler, H. ten Brink, K. Tørseth, and A. Wiedensohler. “A European aerosol phenomenology—2: chemical characteristics of particulate matter at kerbside, urban, rural and background sites in Europe”. *Atmospheric Environment* 38.16 (2004), pp. 2579–2595. ISSN: 1352-2310. DOI: <https://doi.org/10.1016/j.atmosenv.2004.01.041>.

- [96] Q. Zhang, J. L. Jimenez, M. R. Canagaratna, J. D. Allan, H. Coe, I. Ulbrich, M. R. Alfarra, A. Takami, A. M. Middlebrook, Y. L. Sun, K. Dzepina, E. Dunlea, K. Docherty, P. F. DeCarlo, D. Salcedo, T. Onasch, J. T. Jayne, T. Miyoshi, A. Shimono, S. Hatakeyama, N. Takegawa, Y. Kondo, J. Schneider, F. Drewnick, S. Borrmann, S. Weimer, K. Demerjian, P. Williams, K. Bower, R. Bahreini, L. Cottrell, R. J. Griffin, J. Rautiainen, J. Y. Sun, Y. M. Zhang, and D. R. Worsnop. “Ubiquity and dominance of oxygenated species in organic aerosols in anthropogenically-influenced Northern Hemisphere midlatitudes”. *Geophysical Research Letters* 34.13 (2007). DOI: <https://doi.org/10.1029/2007GL029979>.
- [97] B. A. Nault, D. S. Jo, B. C. McDonald, P. Campuzano-Jost, D. A. Day, W. Hu, J. C. Schroder, J. Allan, D. R. Blake, M. R. Canagaratna, H. Coe, M. M. Coggon, P. F. DeCarlo, G. S. Diskin, R. Dunmore, F. Flocke, A. Fried, J. B. Gilman, G. Gkatzelis, J. F. Hamilton, T. F. Hanisco, P. L. Hayes, D. K. Henze, A. Hodzic, J. Hopkins, M. Hu, L. G. Huey, B. T. Jobson, W. C. Kuster, A. Lewis, M. Li, J. Liao, M. O. Nawaz, I. B. Pollack, J. Peischl, B. Rappenglück, C. E. Reeves, D. Richter, J. M. Roberts, T. B. Ryerson, M. Shao, J. M. Sommers, J. Walega, C. Warneke, P. Weibring, G. M. Wolfe, D. E. Young, B. Yuan, Q. Zhang, J. A. de Gouw, and J. L. Jimenez. “Secondary organic aerosols from anthropogenic volatile organic compounds contribute substantially to air pollution mortality”. *Atmospheric Chemistry and Physics* 21.14 (2021), pp. 11201–11224. DOI: 10.5194/acp-21-11201-2021.
- [98] G. Chen, F. Canonaco, A. Tobler, W. Aas, A. Alastuey, J. Allan, S. Atabakhsh, M. Aurela, U. Baltensperger, A. Bougiatioti, J. F. De Brito, D. Ceburnis, B. Chazeau, H. Chebaicheb, K. R. Daellenbach, M. Ehn, I. El Haddad, K. Eleftheriadis, O. Favez, H. Flentje, A. Font, K. Fossun, E. Freney, M. Gini, D. C. Green, L. Heikkinen, H. Herrmann, A.-C. Kalogridis, H. Keernik, R. Lhotka, C. Lin, C. Lunder, M. Maasikmets, M. I. Manousakas, N. Marchand, C. Marin, L. Marmureanu, N. Mihalopoulos, G. Močnik, J. Nećki, C. O’Dowd, J. Ovadnevaite, T. Peter, J.-E. Petit, M. Pikridas, S. Matthew Platt, P. Pokorná, L. Poulain, M. Priestman, V. Riffault, M. Rinaldi, K. Róžański, J. Schwarz, J. Sciare, L. Simon, A. Skiba, J. G. Slowik, Y. Sosedova, I. Stavroulas, K. Styszko, E. Teinmaa, H. Timonen, A. Tremper, J. Vasilescu, M. Via, P. Vodička, A. Wiedensohler, O. Zografou, M. Cruz Minguillón, and A. S. Prévôt. “European aerosol phenomenology - 8: Harmonised source apportionment of organic aerosol using 22 Year-long ACSM/AMS datasets”. *Environment International* 166 (2022), p. 107325. ISSN: 0160-4120. DOI: <https://doi.org/10.1016/j.envint.2022.107325>.
- [99] J. F. Kok, A. A. Adebisi, S. Albani, Y. Balkanski, R. Checa-Garcia, M. Chin, P. R. Colarco, D. S. Hamilton, Y. Huang, A. Ito, M. Klose, D. M. Leung, L. Li, N. M. Mahowald, R. L. Miller, V. Obiso, C. Pérez García-Pando, A. Rocha-Lima, J. S. Wan, and C. A. Whicker. “Improved representation of the global dust cycle using observational constraints on dust properties and abundance”. *Atmospheric Chemistry and Physics* 21.10 (2021), pp. 8127–8167. DOI: 10.5194/acp-21-8127-2021.
- [100] H. Gordon, J. Kirkby, U. Baltensperger, F. Bianchi, M. Breitenlechner, J. Curtius, A. Dias, J. Dommen, N. M. Donahue, E. M. Dunne, J. Duplissy, S. Ehrhart, R. C. Flagan, C. Frege, C. Fuchs, A. Hansel, C. R. Hoyle, M. Kulmala, A. Kürten, K. Lehtipalo, V. Makhmutov, U. Molteni, M. P. Rissanen, Y. Stozhkov, J. Tröstl, G.

- Tsagkogeorgas, R. Wagner, C. Williamson, D. Wimmer, P. M. Winkler, C. Yan, and K. S. Carslaw. “Causes and importance of new particle formation in the present-day and preindustrial atmospheres”. *Journal of Geophysical Research: Atmospheres* 122.16 (2017), pp. 8739–8760. DOI: <https://doi.org/10.1002/2017JD026844>.
- [101] P. K. Saha, S. Hankey, J. D. Marshall, A. L. Robinson, and A. A. Presto. “High-Spatial-Resolution Estimates of Ultrafine Particle Concentrations across the Continental United States”. *Environmental Science & Technology* 55.15 (2021), pp. 10320–10331. DOI: [10.1021/acs.est.1c03237](https://doi.org/10.1021/acs.est.1c03237).
- [102] A. A. Karner, D. S. Eisinger, and D. A. Niemeier. “Near-Roadway Air Quality: Synthesizing the Findings from Real-World Data”. *Environmental Science & Technology* 44.14 (2010). PMID: 20560612, pp. 5334–5344. DOI: [10.1021/es100008x](https://doi.org/10.1021/es100008x).
- [103] M. Ketzel, L. M. Frohn, J. H. Christensen, J. Brandt, A. Massling, C. Andersen, U. Im, S. S. Jensen, J. Khan, O.-K. Nielsen, M. S. Plejdrup, A. Manders, H. D. van der Gon, P. Kumar, and O. Raaschou-Nielsen. “Modelling ultrafine particle number concentrations at address resolution in Denmark from 1979 to 2018 - Part 2: Local and street scale modelling and evaluation”. *Atmospheric Environment* 264 (2021), p. 118633. ISSN: 1352-2310. DOI: <https://doi.org/10.1016/j.atmosenv.2021.118633>.
- [104] L. M. Frohn, M. Ketzel, J. H. Christensen, J. Brandt, U. Im, A. Massling, C. Andersen, M. S. Plejdrup, O.-K. Nielsen, H. D. van der Gon, A. Manders-Groot, and O. Raaschou-Nielsen. “Modelling ultrafine particle number concentrations at address resolution in Denmark from 1979-2018 – Part 1: Regional and urban scale modelling and evaluation”. *Atmospheric Environment* 264 (2021), p. 118631. ISSN: 1352-2310. DOI: <https://doi.org/10.1016/j.atmosenv.2021.118631>.
- [105] P. Kumar, L. Morawska, W. Birmili, P. Paasonen, M. Hu, M. Kulmala, R. M. Harrison, L. Norford, and R. Britter. “Ultrafine particles in cities”. *Environment International* 66 (2014), pp. 1–10. ISSN: 0160-4120. DOI: <https://doi.org/10.1016/j.envint.2014.01.013>.
- [106] Q. Bourgeois, A. M. L. Ekman, J.-B. Renard, R. Krejci, A. Devasthale, F. A.-M. Bender, I. Riipinen, G. Berthet, and J. L. Tackett. “How much of the global aerosol optical depth is found in the boundary layer and free troposphere?” *Atmospheric Chemistry and Physics* 18.10 (2018), pp. 7709–7720. DOI: [10.5194/acp-18-7709-2018](https://doi.org/10.5194/acp-18-7709-2018).
- [107] J. Shao, F. Yi, and Z. Yin. “Aerosol layers in the free troposphere and their seasonal variations as observed in Wuhan, China”. *Atmospheric Environment* 224 (2020), p. 117323. ISSN: 1352-2310. DOI: <https://doi.org/10.1016/j.atmosenv.2020.117323>.
- [108] H. Venzac, K. Sellegri, P. Laj, P. Villani, P. Bonasoni, A. Marinoni, P. Cristofanelli, F. Calzolari, S. Fuzzi, S. Decesari, M.-C. Facchini, E. Vuillermoz, and G. P. Verza. “High frequency new particle formation in the Himalayas”. *Proceedings of the National Academy of Sciences* 105.41 (2008), pp. 15666–15671. DOI: [10.1073/pnas.0801355105](https://doi.org/10.1073/pnas.0801355105).

- [109] C. Rose, K. Sellegri, F. Velarde, I. Moreno, M. Ramonet, K. Weinhold, R. Krejci, P. Ginot, M. Andrade, A. Wiedensohler, and P. Laj. “Frequent nucleation events at the high altitude station of Chacaltaya (5240 m a.s.l.), Bolivia”. *Atmospheric Environment* 102 (2015), pp. 18–29. ISSN: 1352-2310. DOI: <https://doi.org/10.1016/j.atmosenv.2014.11.015>.
- [110] Q. Xiao, J. Zhang, Y. Wang, L. D. Ziemba, E. Crosbie, E. L. Winstead, C. E. Robinson, J. P. DiGangi, G. S. Diskin, J. S. Reid, K. S. Schmidt, A. Sorooshian, M. R. A. Hilario, S. Woods, P. Lawson, S. A. Stammes, and J. Wang. “New particle formation in the tropical free troposphere during CAMP²Ex: statistics and impact of emission sources, convective activity, and synoptic conditions”. *Atmospheric Chemistry and Physics* 23.17 (2023), pp. 9853–9871. DOI: 10.5194/acp-23-9853-2023.
- [111] F. Bianchi, J. Tröstl, H. Junninen, C. Frege, S. Henne, C. R. Hoyle, U. Molteni, E. Herrmann, A. Adamov, N. Bukowiecki, X. Chen, J. Duplissy, M. Gysel, M. Hutterli, J. Kangasluoma, J. Kontkanen, A. Kürten, H. E. Manninen, S. Münch, O. Peräkylä, T. Petäjä, L. Rondo, C. Williamson, E. Weingartner, J. Curtius, D. R. Worsnop, M. Kulmala, J. Dommen, and U. Baltensperger. “New particle formation in the free troposphere: A question of chemistry and timing”. *Science* 352.6289 (2016), pp. 1109–1112. DOI: 10.1126/science.aad5456.
- [112] M. de Reus, J. Ström, J. Curtius, L. Pirjola, E. Vignati, F. Arnold, H. C. Hansson, M. Kulmala, J. Lelieveld, and F. Raes. “Aerosol production and growth in the upper free troposphere”. *Journal of Geophysical Research: Atmospheres* 105.D20 (2000), pp. 24751–24762. DOI: <https://doi.org/10.1029/2000JD900382>.
- [113] K. D. Perry and P. V. Hobbs. “Further evidence for particle nucleation in clear air adjacent to marine cumulus clouds”. *Journal of Geophysical Research: Atmospheres* 99.D11 (1994), pp. 22803–22818. DOI: <https://doi.org/10.1029/94JD01926>.
- [114] A. D. Clarke, V. N. Kapustin, F. L. Eisele, R. J. Weber, and P. H. McMurry. “Particle production near marine clouds: Sulfuric acid and predictions from classical binary nucleation”. *Geophysical Research Letters* 26.16 (1999), pp. 2425–2428. DOI: <https://doi.org/10.1029/1999GL900438>.
- [115] J. Zhang, X. Gong, E. Crosbie, G. Diskin, K. Froyd, S. Hall, A. Kupc, R. Moore, J. Peischl, A. Rollins, J. Schwarz, M. Shook, C. Thompson, K. Ullmann, C. Williamson, A. Wisthaler, L. Xu, L. Ziemba, C. A. Brock, and J. Wang. “Stratospheric air intrusions promote global-scale new particle formation”. *Science* 385.6705 (2024), pp. 210–216. DOI: 10.1126/science.adn2961.
- [116] C. E. Junge, C. W. Chagnon, and J. E. Manson. “STRATOSPHERIC AEROSOLS”. *Journal of Atmospheric Sciences* 18.1 (1961), pp. 81–108. DOI: 10.1175/1520-0469(1961)018<0081:SA>2.0.CO;2.
- [117] C. E. Junge, C. W. Chagnon, and J. E. Manson. “A World-wide Stratospheric Aerosol Layer”. *Science* 133.3463 (1961), pp. 1478–1479. DOI: 10.1126/science.133.3463.1478.b.

- [118] C. E. Junge and J. E. Manson. “Stratospheric aerosol studies”. *Journal of Geophysical Research (1896-1977)* 66.7 (1961), pp. 2163–2182. DOI: <https://doi.org/10.1029/JZ066i007p02163>.
- [119] C. W. Chagnon and C. E. Junge. “THE VERTICAL DISTRIBUTION OF SUB-MICRON PARTICLES IN THE STRATOSPHERE”. *Journal of Atmospheric Sciences* 18.6 (1961), pp. 746–752. DOI: 10.1175/1520-0469(1961)018<0746:TVD0SM>2.0.CO;2.
- [120] P. J. Crutzen. “The possible importance of CSO for the sulfate layer of the stratosphere”. *Geophysical Research Letters* 3.2 (1976), pp. 73–76. DOI: <https://doi.org/10.1029/GL003i002p00073>.
- [121] C. Brühl, J. Lelieveld, P. J. Crutzen, and H. Tost. “The role of carbonyl sulphide as a source of stratospheric sulphate aerosol and its impact on climate”. *Atmospheric Chemistry and Physics* 12.3 (2012), pp. 1239–1253. DOI: 10.5194/acp-12-1239-2012.
- [122] J.-X. Sheng, D. K. Weisenstein, B.-P. Luo, E. Rozanov, A. Stenke, J. Anet, H. Bingemer, and T. Peter. “Global atmospheric sulfur budget under volcanically quiescent conditions: Aerosol-chemistry-climate model predictions and validation”. *Journal of Geophysical Research: Atmospheres* 120.1 (2015), pp. 256–276. DOI: <https://doi.org/10.1002/2014JD021985>.
- [123] J. Schallrock, C. Brühl, C. Bingen, M. Höpfner, L. Rieger, and J. Lelieveld. “Reconstructing volcanic radiative forcing since 1990, using a comprehensive emission inventory and spatially resolved sulfur injections from satellite data in a chemistry-climate model”. *Atmospheric Chemistry and Physics* 23.2 (2023), pp. 1169–1207. DOI: 10.5194/acp-23-1169-2023.
- [124] A. Schmidt, M. J. Mills, S. Ghan, J. M. Gregory, R. P. Allan, T. Andrews, C. G. Bardeen, A. Conley, P. M. Forster, A. Gettelman, R. W. Portmann, S. Solomon, and O. B. Toon. “Volcanic Radiative Forcing From 1979 to 2015”. *Journal of Geophysical Research: Atmospheres* 123.22 (2018), pp. 12491–12508. DOI: 10.1029/2018JD028776.
- [125] J. M. C. Plane. “Cosmic dust in the earth’s atmosphere”. *Chem. Soc. Rev.* 41 (19 2012), pp. 6507–6518. DOI: 10.1039/C2CS35132C.
- [126] M. E. Hervig, C. G. Bardeen, D. E. Siskind, M. J. Mills, and R. Stockwell. “Meteoric smoke and H₂SO₄ aerosols in the upper stratosphere and mesosphere”. *Geophysical Research Letters* 44.2 (2017), pp. 1150–1157. DOI: <https://doi.org/10.1002/2016GL072049>.
- [127] “Cosmic dust fluxes in the atmospheres of Earth, Mars, and Venus”. *Icarus* 335 (2020), p. 113395. ISSN: 0019-1035. DOI: <https://doi.org/10.1016/j.icarus.2019.113395>.
- [128] C. J. Williamson, A. Kupc, D. Axisa, K. R. Billsback, T. Bui, P. Campuzano-Jost, M. Dollner, K. D. Froyd, A. L. Hodshire, J. L. Jimenez, et al. “A large source of cloud condensation nuclei from new particle formation in the tropics”. *Nature* 574.7778 (2019), pp. 399–403. DOI: 10.1038/s41586-019-1638-9.

- [129] R. Weigel, S. Borrmann, J. Kazil, A. Minikin, A. Stohl, J. C. Wilson, J. M. Reeves, D. Kunkel, M. de Reus, W. Frey, E. R. Lovejoy, C. M. Volk, S. Viciani, F. D’Amato, C. Schiller, T. Peter, H. Schlager, F. Cairo, K. S. Law, G. N. Shur, G. V. Belyaev, and J. Curtius. “In situ observations of new particle formation in the tropical upper troposphere: the role of clouds and the nucleation mechanism”. *Atmospheric Chemistry and Physics* 11.18 (2011), pp. 9983–10010. DOI: [10.5194/acp-11-9983-2011](https://doi.org/10.5194/acp-11-9983-2011).
- [130] A. D. Clarke, F. Eisele, V. N. Kapustin, K. Moore, D. Tanner, L. Mauldin, M. Litchy, B. Lienert, M. A. Carroll, and G. Albercook. “Nucleation in the equatorial free troposphere: Favorable environments during PEM-Tropics”. *Journal of Geophysical Research: Atmospheres* 104.D5 (1999), pp. 5735–5744. DOI: <https://doi.org/10.1029/98JD02303>.
- [131] C. A. Brock, P. Hamill, J. C. Wilson, H. H. Jonsson, and K. R. Chan. “Particle Formation in the Upper Tropical Troposphere: A Source of Nuclei for the Stratospheric Aerosol”. *Science* 270.5242 (1995), pp. 1650–1653. DOI: [10.1126/science.270.5242.1650](https://doi.org/10.1126/science.270.5242.1650).
- [132] R. Weigel, C. Mahnke, M. Baumgartner, A. Dragoneas, B. Vogel, F. Ploeger, S. Viciani, F. D’Amato, S. Bucci, B. Legras, B. Luo, and S. Borrmann. “In situ observation of new particle formation (NPF) in the tropical tropopause layer of the 2017 Asian monsoon anticyclone – Part 1: Summary of StratoClim results”. *Atmospheric Chemistry and Physics* 21.15 (2021), pp. 11689–11722. DOI: [10.5194/acp-21-11689-2021](https://doi.org/10.5194/acp-21-11689-2021).
- [133] A. G. Turner and H. Annamalai. “Climate change and the South Asian summer monsoon”. *Nature Climate Change* 2.8 (2012), pp. 587–595. DOI: [10.1038/nclimate1495](https://doi.org/10.1038/nclimate1495).
- [134] D. Yihui and J. C. Chan. “The East Asian summer monsoon: an overview”. *Meteorology and Atmospheric Physics* 89.1 (2005), pp. 117–142. DOI: [10.1007/s00703-005-0125-z](https://doi.org/10.1007/s00703-005-0125-z).
- [135] J.-P. Vernier, L. W. Thomason, and J. Kar. “CALIPSO detection of an Asian tropopause aerosol layer”. *Geophysical Research Letters* 38.7 (2011). DOI: <https://doi.org/10.1029/2010GL046614>.
- [136] O. Appel, F. Köllner, A. Dragoneas, A. Hünig, S. Molleker, H. Schlager, C. Mahnke, R. Weigel, M. Port, C. Schulz, F. Drewnick, B. Vogel, F. Stroh, and S. Borrmann. “Chemical analysis of the Asian tropopause aerosol layer (ATAL) with emphasis on secondary aerosol particles using aircraft-based in situ aerosol mass spectrometry”. *Atmospheric Chemistry and Physics* 22.20 (2022), pp. 13607–13630. DOI: [10.5194/acp-22-13607-2022](https://doi.org/10.5194/acp-22-13607-2022).
- [137] M. Höpfner, J. Ungermann, S. Borrmann, R. Wagner, R. Spang, M. Riese, G. Stiller, O. Appel, A. M. Batenburg, S. Bucci, et al. “Ammonium nitrate particles formed in upper troposphere from ground ammonia sources during Asian monsoons”. *Nature Geoscience* 12.8 (2019), pp. 608–612. DOI: [10.1038/s41561-019-0385-8](https://doi.org/10.1038/s41561-019-0385-8).
- [138] M. O. Andreae, A. Afchine, R. Albrecht, B. A. Holanda, P. Artaxo, H. M. J. Barbosa, S. Borrmann, M. A. Cecchini, A. Costa, M. Dollner, D. Fütterer, E. Järvinen, T. Jurkat, T. Klimach, T. Konemann, C. Knote, M. Krämer, T. Krisna, L. A. T. Machado,

- S. Mertes, A. Minikin, C. Pöhlker, M. L. Pöhlker, U. Pöschl, D. Rosenfeld, D. Sauer, H. Schlager, M. Schnaiter, J. Schneider, C. Schulz, A. Spanu, V. B. Sperling, C. Voigt, A. Walser, J. Wang, B. Weinzierl, M. Wendisch, and H. Ziereis. “Aerosol characteristics and particle production in the upper troposphere over the Amazon Basin”. *Atmospheric Chemistry and Physics* 18.2 (2018), pp. 921–961. DOI: 10.5194/acp-18-921-2018.
- [139] Y. Liu, H. Su, S. Wang, C. Wei, W. Tao, M. L. Pöhlker, C. Pöhlker, B. A. Holanda, O. O. Krüger, T. Hoffmann, M. Wendisch, P. Artaxo, U. Pöschl, M. O. Andreae, and Y. Cheng. “Strong particle production and condensational growth in the upper troposphere sustained by biogenic VOCs from the canopy of the Amazon Basin”. *Atmospheric Chemistry and Physics* 23.1 (2023), pp. 251–272. DOI: 10.5194/acp-23-251-2023.
- [140] J. Lelieveld, K. Klingmüller, A. Pozzer, R. T. Burnett, A. Haines, and V. Ramanathan. “Effects of fossil fuel and total anthropogenic emission removal on public health and climate”. *Proceedings of the National Academy of Sciences* 116.15 (2019), pp. 7192–7197. DOI: 10.1073/pnas.1819989116.
- [141] D. Nuvolone, D. Petri, and F. Voller. “The effects of ozone on human health”. en. *Environmental Science and Pollution Research* 25.9 (Mar. 2018), pp. 8074–8088. ISSN: 1614-7499. DOI: 10.1007/s11356-017-9239-3.
- [142] D. Akritidis, S. Bacer, P. Zanis, A. K. Georgoulas, S. Chowdhury, L. W. Horowitz, V. Naik, F. M. O’Connor, J. Keeble, P. L. Sager, T. van Noije, P. Zhou, S. Turnock, J. J. West, J. Lelieveld, and A. Pozzer. “Strong increase in mortality attributable to ozone pollution under a climate change and demographic scenario”. *Environmental Research Letters* 19.2 (Feb. 2024). Publisher: IOP Publishing, p. 024041. DOI: 10.1088/1748-9326/ad2162.
- [143] K. R. Hakeem, M. Sabir, M. Ozturk, M. S. Akhtar, and F. H. Ibrahim. “Nitrate and Nitrogen Oxides: Sources, Health Effects and Their Remediation”. *Reviews of Environmental Contamination and Toxicology Volume 242*. Ed. by P. de Voogt. Cham: Springer International Publishing, 2017, pp. 183–217. ISBN: 978-3-319-51243-3. DOI: 10.1007/398_2016_11.
- [144] G. Thompson and T. Eidhammer. “A Study of Aerosol Impacts on Clouds and Precipitation Development in a Large Winter Cyclone”. *Journal of the Atmospheric Sciences* 71.10 (2014), pp. 3636–3658. DOI: 10.1175/JAS-D-13-0305.1.
- [145] W.-K. Tao, J.-P. Chen, Z. Li, C. Wang, and C. Zhang. “Impact of aerosols on convective clouds and precipitation”. *Reviews of Geophysics* 50.2 (2012). DOI: <https://doi.org/10.1029/2011RG000369>.
- [146] R.-J. Huang, Y. Zhang, C. Bozzetti, K.-F. Ho, J.-J. Cao, Y. Han, K. R. Daellenbach, J. G. Slowik, S. M. Platt, F. Canonaco, et al. “High secondary aerosol contribution to particulate pollution during haze events in China”. *Nature* 514.7521 (2014), pp. 218–222. DOI: <https://doi.org/10.1038/nature13774>.

- [147] C.-W. Liang, C.-C. Chang, and J.-J. Liang. “The impacts of air quality and secondary organic aerosols formation on traffic accidents in heavy fog–haze weather”. *Heliyon* 9.4 (2023), e14631. ISSN: 2405-8440. DOI: <https://doi.org/10.1016/j.heliyon.2023.e14631>.
- [148] F. H. B. Gene E. Likens and N. M. Johnson. “Acid Rain”. *Environment: Science and Policy for Sustainable Development* 14.2 (1972), pp. 33–40. DOI: 10.1080/00139157.1972.9933001.
- [149] P. Khillare, S. Balachandran, and B. R. Meena. “Spatial and temporal variation of heavy metals in atmospheric aerosol of Delhi”. *Environmental Monitoring and Assessment* 90 (2004), pp. 1–21. DOI: <https://doi.org/10.1023/B:EMAS.0000003555.36394.17>.
- [150] P. J. Crutzen. “Albedo enhancement by stratospheric sulfur injections: A contribution to resolve a policy dilemma?” *Climatic change* 77.3-4 (2006), p. 211. DOI: 10.1007/s10584-006-9101-y.
- [151] A. Robock, A. Marquardt, B. Kravitz, and G. Stenchikov. “Benefits, risks, and costs of stratospheric geoengineering”. *Geophysical Research Letters* 36.19 (2009). DOI: <https://doi.org/10.1029/2009GL039209>.
- [152] A. Robock. “Albedo enhancement by stratospheric sulfur injections: More research needed”. *Earth’s Future* 4.12 (2016), pp. 644–648. DOI: <https://doi.org/10.1002/2016EF000407>.
- [153] A. Pozzer, S. F. Reifenberg, V. Kumar, B. Franco, **M. Kohl**, D. Taraborrelli, S. Gromov, S. Ehrhart, P. Jöckel, R. Sander, V. Fall, S. Rosanka, V. Karydis, D. Akritidis, T. Emmerichs, M. Crippa, D. Guizzardi, J. W. Kaiser, L. Clarisse, A. Kiendler-Scharr, H. Tost, and A. Tsimpidi. “Simulation of organics in the atmosphere: evaluation of EMACv2.54 with the Mainz Organic Mechanism (MOM) coupled to the ORACLE (v1.0) submodel”. *Geoscientific Model Development* 15.6 (2022), pp. 2673–2710. DOI: 10.5194/gmd-15-2673-2022.
- [154] S. F. Reifenberg, A. Martin, **M. Kohl**, S. Bacer, Z. Hamryszczak, I. Tadic, L. Röder, D. J. Crowley, H. Fischer, K. Kaiser, J. Schneider, R. Dörich, J. N. Crowley, L. Tomsche, A. Marsing, C. Voigt, A. Zahn, C. Pöhlker, B. A. Holanda, O. Krüger, U. Pöschl, M. Pöhlker, P. Jöckel, M. Dorf, U. Schumann, J. Williams, B. Bohn, J. Curtius, H. Harder, H. Schlager, J. Lelieveld, and A. Pozzer. “Numerical simulation of the impact of COVID-19 lockdown on tropospheric composition and aerosol radiative forcing in Europe”. *Atmospheric Chemistry and Physics* 22.16 (2022), pp. 10901–10917. DOI: 10.5194/acp-22-10901-2022.
- [155] **M. Kohl**, J. Lelieveld, S. Chowdhury, S. Ehrhart, D. Sharma, Y. Cheng, S. N. Tripathi, M. Sebastian, G. Pandithurai, H. Wang, and A. Pozzer. “Numerical simulation and evaluation of global ultrafine particle concentrations at the Earth’s surface”. *Atmospheric Chemistry and Physics* 23.20 (2023), pp. 13191–13215. DOI: 10.5194/acp-23-13191-2023.

- [156] M. Kuntic, I. Kuntic, D. Cleppien, A. Pozzer, D. Nußbaum, M. Oelze, T. Junglas, L. Strohm, H. Ubbens, S. Daub, M. T. Bayo Jimenez, S. Danckwardt, T. Berkemeier, O. Hahad, **M. Kohl**, S. Steven, A. Stroh, J. Lelieveld, T. Münzel, and A. Daiber. “Differential inflammation, oxidative stress and cardiovascular damage markers of nano- and micro-particle exposure in mice: Implications for human disease burden”. *Redox Biology* (2025), p. 103644. ISSN: 2213-2317. DOI: <https://doi.org/10.1016/j.redox.2025.103644>.
- [157] C. Xenofontos, **M. Kohl**, S. Ruhl, J. Almeida, H. M. Beckmann, L. Caudillo-Plath, S. Ehrhart, K. Höhler, M. Kaniyodical Sebastian, W. Kong, et al. “The impact of ammonia on particle formation in the Asian Tropopause Aerosol Layer”. *npj Climate and Atmospheric Science* 7.1 (2024), p. 215. DOI: 10.1038/s41612-024-00758-3.
- [158] M. Wang, M. Xiao, B. Bertozzi, G. Marie, B. Rörup, B. Schulze, R. Bardakov, X.-C. He, J. Shen, W. Scholz, et al. “Synergistic HNO₃–H₂SO₄–NH₃ upper tropospheric particle formation”. *Nature* 605.7910 (2022), pp. 483–489. DOI: 10.1038/s41586-022-04605-4.
- [159] F. Ploeger, P. Konopka, K. Walker, and M. Riese. “Quantifying pollution transport from the Asian monsoon anticyclone into the lower stratosphere”. *Atmospheric Chemistry and Physics* 17.11 (2017), pp. 7055–7066. DOI: 10.5194/acp-17-7055-2017.
- [160] B. Vogel, R. Müller, G. Günther, R. Spang, S. Hanumanthu, D. Li, M. Riese, and G. P. Stiller. “Lagrangian simulations of the transport of young air masses to the top of the Asian monsoon anticyclone and into the tropical pipe”. *Atmospheric Chemistry and Physics* 19.9 (2019), pp. 6007–6034. DOI: 10.5194/acp-19-6007-2019.
- [161] L. L. Pan, S. B. Honomichl, D. E. Kinnison, M. Abalos, W. J. Randel, J. W. Bergman, and J. Bian. “Transport of chemical tracers from the boundary layer to stratosphere associated with the dynamics of the Asian summer monsoon”. *Journal of Geophysical Research: Atmospheres* 121.23 (2016), pp. 14, 159–14, 174. DOI: 10.1002/2016JD025616.
- [162] A. Hünig, O. Appel, A. Dragoneas, S. Molleker, H.-C. Clemen, F. Helleis, T. Klimach, F. Köllner, T. Böttger, F. Drewnick, J. Schneider, and S. Borrmann. “Design, characterization, and first field deployment of a novel aircraft-based aerosol mass spectrometer combining the laser ablation and flash vaporization techniques”. *Atmospheric Measurement Techniques* 15.9 (2022), pp. 2889–2921. DOI: 10.5194/amt-15-2889-2022.
- [163] F. Friedl-Vallon, T. Gulde, F. Hase, A. Kleinert, T. Kullessa, G. Maucher, T. Neubert, F. Olschewski, C. Piesch, P. Preusse, H. Rongen, C. Sartorius, H. Schneider, A. Schönfeld, V. Tan, N. Bayer, J. Blank, R. Dapp, A. Ebersoldt, H. Fischer, F. Graf, T. Guggenmoser, M. Höpfner, M. Kaufmann, E. Kretschmer, T. Latzko, H. Nordmeyer, H. Oelhaf, J. Orphal, M. Riese, G. Schardt, J. Schillings, M. K. Sha, O. Suminska-Ebersoldt, and J. Ungermann. “Instrument concept of the imaging Fourier transform spectrometer GLORIA”. *Atmospheric Measurement Techniques* 7.10 (2014), pp. 3565–3577. DOI: 10.5194/amt-7-3565-2014.

- [164] M. Riese, H. Oelhaf, P. Preusse, J. Blank, M. Ern, F. Friedl-Vallon, H. Fischer, T. Guggenmoser, M. Höpfner, P. Hoor, M. Kaufmann, J. Orphal, F. Plöger, R. Spang, O. Suminska-Ebersoldt, J. Ungermann, B. Vogel, and W. Woiwode. “Gimballed Limb Observer for Radiance Imaging of the Atmosphere (GLORIA) scientific objectives”. *Atmospheric Measurement Techniques* 7.7 (2014), pp. 1915–1928. DOI: 10.5194/amt-7-1915-2014.
- [165] E. E. McDuffie, S. J. Smith, P. O’Rourke, K. Tibrewal, C. Venkataraman, E. A. Marais, B. Zheng, M. Crippa, M. Brauer, and R. V. Martin. “A global anthropogenic emission inventory of atmospheric pollutants from sector- and fuel-specific sources (1970–2017): an application of the Community Emissions Data System (CEDS)”. *Earth System Science Data* 12.4 (2020), pp. 3413–3442. DOI: 10.5194/essd-12-3413-2020.
- [166] L. L. Pan, E. L. Atlas, S. B. Honomichl, W. P. Smith, D. E. Kinnison, S. Solomon, M. L. Santee, A. Saiz-Lopez, J. C. Laube, B. Wang, R. Ueyama, J. F. Bresch, R. S. Hornbrook, E. C. Apel, A. J. Hills, V. Treadaway, K. Smith, S. Schauffler, S. Donnelly, R. Hendershot, R. Lueb, T. Campos, S. Viciani, F. D’Amato, G. Bianchini, M. Barucci, J. R. Podolske, L. T. Iraci, C. Gurganus, P. Bui, J. M. Dean-Day, L. Millán, J.-M. Ryoo, B. Barletta, J.-H. Koo, J. Kim, Q. Liang, W. J. Randel, T. Thornberry, and P. A. Newman. “East Asian summer monsoon delivers large abundances of very short-lived organic chlorine substances to the lower stratosphere”. *Proceedings of the National Academy of Sciences* 121.12 (2024), e2318716121. DOI: 10.1073/pnas.2318716121.
- [167] H. K. Alladi, P. Satheesh Chandran, and V. R. M. “Impact of ENSO on the UTLS chemical composition in the Asian Summer Monsoon Anticyclone”. *Atmospheric Research* 309 (2024), p. 107551. ISSN: 0169-8095. DOI: <https://doi.org/10.1016/j.atmosres.2024.107551>.
- [168] E. Salati and P. B. Vose. “Amazon Basin: A System in Equilibrium”. *Science* 225.4658 (1984), pp. 129–138. DOI: 10.1126/science.225.4658.129.
- [169] M. Gloor, L. Gatti, R. Brien, T. R. Feldpausch, O. L. Phillips, J. Miller, J. P. Ometto, H. Rocha, T. Baker, B. de Jong, R. A. Houghton, Y. Malhi, L. E. O. C. Aragão, J.-L. Guyot, K. Zhao, R. Jackson, P. Peylin, S. Sitch, B. Poulter, M. Lomas, S. Zaehle, C. Huntingford, P. Levy, and J. Lloyd. “The carbon balance of South America: a review of the status, decadal trends and main determinants”. *Biogeosciences* 9.12 (2012), pp. 5407–5430. DOI: 10.5194/bg-9-5407-2012.
- [170] E. A. Davidson, A. C. de Araújo, P. Artaxo, J. K. Balch, I. F. Brown, M. M. C. Bustamante, M. T. Coe, R. S. DeFries, M. Keller, M. Longo, et al. “The Amazon basin in transition”. *Nature* 481.7381 (2012), pp. 321–328. DOI: 10.1038/nature10717.
- [171] M. O. Andreae, O. C. Acevedo, A. Araújo, P. Artaxo, C. G. G. Barbosa, H. M. J. Barbosa, J. Brito, S. Carbone, X. Chi, B. B. L. Cintra, N. F. da Silva, N. L. Dias, C. Q. Dias-Júnior, F. Ditas, R. Ditz, A. F. L. Godoi, R. H. M. Godoi, M. Heimann, T. Hoffmann, J. Kesselmeier, T. Könemann, M. L. Krüger, J. V. Lavric, A. O. Manzi, A. P. Lopes, D. L. Martins, E. F. Mikhailov, D. Moran-Zuloaga, B. W. Nelson, A. C. Nölscher, D. Santos Nogueira, M. T. F. Piedade, C. Pöhlker, U. Pöschl, C. A. Quesada, L. V. Rizzo, C.-U. Ro, N. Ruckteschler, L. D. A. Sá, M. de Oliveira Sá, C. B. Sales, R. M. N. dos Santos, J. Saturno, J. Schöngart, M. Sörgel, C. M. de

- Souza, R. A. F. de Souza, H. Su, N. Targhetta, J. Tóta, I. Trebs, S. Trumbore, A. van Eijck, D. Walter, Z. Wang, B. Weber, J. Williams, J. Winderlich, F. Wittmann, S. Wolff, and A. M. Yáñez-Serrano. “The Amazon Tall Tower Observatory (ATTO): overview of pilot measurements on ecosystem ecology, meteorology, trace gases, and aerosols”. *Atmospheric Chemistry and Physics* 15.18 (2015), pp. 10723–10776. DOI: 10.5194/acp-15-10723-2015.
- [172] M. L. Pöhlker, C. Pöhlker, F. Ditas, T. Klimach, I. Hrabec de Angelis, A. Araújo, J. Brito, S. Carbone, Y. Cheng, X. Chi, R. Ditz, S. S. Gunthe, J. Kesselmeier, T. Könemann, J. V. Lavrič, S. T. Martin, E. Mikhailov, D. Moran-Zuloaga, D. Rose, J. Saturno, H. Su, R. Thalman, D. Walter, J. Wang, S. Wolff, H. M. J. Barbosa, P. Artaxo, M. O. Andreae, and U. Pöschl. “Long-term observations of cloud condensation nuclei in the Amazon rain forest – Part 1: Aerosol size distribution, hygroscopicity, and new model parametrizations for CCN prediction”. *Atmospheric Chemistry and Physics* 16.24 (2016), pp. 15709–15740. DOI: 10.5194/acp-16-15709-2016.
- [173] A. Ringsdorf, A. Edtbauer, J. Vilà-Guerau de Arellano, E. Pfannerstill, S. Gromov, V. Kumar, A. Pozzer, S. Wolff, A. Tsokankunku, M. Soergel, et al. “Inferring the diurnal variability of OH radical concentrations over the Amazon from BVOC measurements”. *Scientific Reports* 13.1 (2023), p. 14900. DOI: 10.1038/s41598-023-41748-4.
- [174] J. Shen, D. M. Russell, J. DeVivo, F. Kunkler, R. Baalbaki, B. Mentler, W. Scholz, W. Yu, L. Caudillo-Plath, E. Sommer, et al. “New particle formation from isoprene under upper-tropospheric conditions”. *Nature* 636.8041 (2024), pp. 115–123. DOI: 10.1038/s41586-024-08196-0.
- [175] J. Curtius, M. Heinritzi, L. J. Beck, M. L. Pöhlker, N. Tripathi, B. E. Krumm, P. Holzbeck, C. M. Nussbaumer, L. Hernández Pardo, T. Klimach, K. Barmounis, S. T. Andersen, R. Bardakov, B. Bohn, M. A. Cecchini, J.-P. Chaboureaud, T. Dauhut, D. Dienhart, R. Dörich, A. Edtbauer, A. Giez, A. Hartmann, B. A. Holanda, P. Joppe, K. Kaiser, T. Keber, H. Klebach, O. O. Krüger, A. Kürten, C. Mallaun, D. Marno, M. Martinez, C. Monteiro, C. Nelson, L. Ort, S. S. Raj, S. Richter, A. Ringsdorf, F. Rocha, M. Simon, S. Sree Kumar, A. Tsokankunku, G. R. Unfer, I. D. Valenti, N. Wang, A. Zahn, M. Zauner-Wieczorek, R. I. Albrecht, M. O. Andreae, P. Artaxo, J. N. Crowley, H. Fischer, H. Harder, D. L. Herdies, L. A. T. Machado, C. Pöhlker, U. Pöschl, A. Possner, A. Pozzer, J. Schneider, J. Williams, and J. Lelieveld. “Isoprene nitrates drive new particle formation in Amazon’s upper troposphere”. en. *Nature* 636.8041 (Dec. 2024). Publisher: Nature Publishing Group, pp. 124–130. ISSN: 1476-4687. DOI: 10.1038/s41586-024-08192-4.
- [176] R. Vella, M. Forrest, J. Lelieveld, and H. Tost. “Isoprene and monoterpene simulations using the chemistry–climate model EMAC (v2.55) with interactive vegetation from LPJ-GUESS (v4.0)”. *Geoscientific Model Development* 16.3 (2023), pp. 885–906. DOI: 10.5194/gmd-16-885-2023.
- [177] J. Lelieveld, S. Gromov, A. Pozzer, and D. Taraborrelli. “Global tropospheric hydroxyl distribution, budget and reactivity”. *Atmospheric Chemistry and Physics* 16.19 (2016), pp. 12477–12493. DOI: 10.5194/acp-16-12477-2016.

- [178] C. M. Nussbaumer, **M. Kohl**, A. Pozzer, I. Tadic, R. Rohloff, D. Marno, H. Harder, H. Ziereis, A. Zahn, F. Obersteiner, A. Hofzumahaus, H. Fuchs, C. K nstler, W. H. Brune, T. B. Ryerson, J. Peischl, C. R. Thompson, I. Bourgeois, J. Lelieveld, and H. Fischer. “Ozone Formation Sensitivity to Precursors and Lightning in the Tropical Troposphere Based on Airborne Observations”. *Journal of Geophysical Research: Atmospheres* 129.14 (2024), e2024JD041168. DOI: <https://doi.org/10.1029/2024JD041168>.
- [179] C. E. Scott, S. A. Monks, D. V. Spracklen, S. Arnold, P. Forster, A. Rap, M.  ij l , P. Artaxo, K. Carslaw, M. Chipperfield, et al. “Impact on short-lived climate forcers increases projected warming due to deforestation”. *Nature communications* 9.1 (2018), p. 157. DOI: 10.1038/s41467-017-02412-4.
- [180] J. Weber, J. A. King, N. L. Abraham, D. P. Grosvenor, C. J. Smith, Y. M. Shin, P. Lawrence, S. Roe, D. J. Beerling, and M. V. Martin. “Chemistry-albedo feedbacks offset up to a third of forestation’s CO₂ removal benefits”. *Science* 383.6685 (2024), pp. 860–864. DOI: 10.1126/science.adg6196.
- [181] P. O. Wennberg, K. H. Bates, J. D. Crounse, L. G. Dodson, R. C. McVay, L. A. Mertens, T. B. Nguyen, E. Praske, R. H. Schwantes, M. D. Smarte, J. M. St Clair, A. P. Teng, X. Zhang, and J. H. Seinfeld. “Gas-Phase Reactions of Isoprene and Its Major Oxidation Products”. *Chemical Reviews* 118.7 (2018). PMID: 29522327, pp. 3337–3390. DOI: 10.1021/acs.chemrev.7b00439.
- [182] P. Joppe, J. Schneider, K. Kaiser, H. Fischer, P. Hoor, D. Kunkel, H.-C. Lachnitt, A. Marsing, L. R der, H. Schlager, L. Tomsche, C. Voigt, A. Zahn, and S. Borrmann. “The influence of extratropical cross-tropopause mixing on the correlation between ozone and sulfate aerosol in the lowermost stratosphere”. *Atmospheric Chemistry and Physics* 24.13 (2024), pp. 7499–7522. DOI: 10.5194/acp-24-7499-2024.
- [183] S. Ohlwein, R. Kappeler, M. Kutlar Joss, N. K nzli, and B. Hoffmann. “Health effects of ultrafine particles: a systematic literature review update of epidemiological evidence”. eng. *International Journal of Public Health* 64.4 (May 2019), pp. 547–559. ISSN: 1661-8564. DOI: 10.1007/s00038-019-01202-7.
- [184] T. Yang, Y. Guo, R. Zhang, J. Zhong, Z. Xu, L. Liu, Z. Peng, F. Wang, Y. Jiang, Y. Zhu, Q. Liu, Y. Wu, Q. Meng, Z. Duoji, M. Han, X. Meng, R. Chen, H. Kan, C. Liu, and F. Hong. “Associations between long-term exposure to ultrafine particles and type 2 diabetes: A large-scale, multicenter study in China”. *Journal of Hazardous Materials* 488 (2025), p. 137364. ISSN: 0304-3894. DOI: <https://doi.org/10.1016/j.jhazmat.2025.137364>.
- [185] Z. A. Pond, P. K. Saha, C. J. Coleman, A. A. Presto, A. L. Robinson, and C. Arden Pope III. “Mortality risk and long-term exposure to ultrafine particles and primary fine particle components in a national U.S. Cohort”. *Environment International* 167 (2022), p. 107439. ISSN: 0160-4120. DOI: <https://doi.org/10.1016/j.envint.2022.107439>.
- [186] F. Bouma, N. A. Janssen, J. Wesseling, S. van Ratingen, M. Strak, J. Kerckhoffs, U. Gehring, W. Hendricx, K. de Hoogh, R. Vermeulen, and G. Hoek. “Long-term exposure to ultrafine particles and natural and cause-specific mortality”. *Environment*

International 175 (2023), p. 107960. ISSN: 0160-4120. DOI: <https://doi.org/10.1016/j.envint.2023.107960>.

- [187] M. Lloyd, T. Olaniyan, A. Ganji, J. Xu, A. Venuta, L. Simon, M. Zhang, M. Saeedi, S. Yamanouchi, A. Wang, A. Schmidt, H. Chen, P. Villeneuve, J. Apte, E. Lavigne, R. T. Burnett, M. Tjepkema, M. Hatzopoulou, and S. Weichenthal. “Airborne Nanoparticle Concentrations Are Associated with Increased Mortality Risk in Canada’s Two Largest Cities”. *American Journal of Respiratory and Critical Care Medicine* 210.11 (2024). PMID: 38924496, pp. 1338–1347. DOI: 10.1164/rccm.202311-20130C.