

CHEMICAL COMPOSITION AND TRANSFORMATION
OF SUBMICRON AEROSOL PARTICLES IN THE
OUTFLOW OF MAJOR POPULATION CENTRES

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Alles hat seine Stunde.
Für jedes Geschehen unter dem Himmel
gibt es eine bestimmte Zeit.
Prediger 3,1

Dass alles seine Zeit hat,
heißt auch,
dass alles seine Zeit braucht.
(Ernst Ferstl)

ZUSAMMENFASSUNG

Urbane Ballungsräume tragen signifikant zur anthropogenen Luftverschmutzung bei. Aufgrund der stetig wachsenden Weltbevölkerung und des zunehmenden Urbanisierungsgrades ist zu erwarten, dass die Regulierung anthropogener Großstadtemissionen zukünftig wichtiger wird. Diese Arbeit fokussiert sich auf urbane Emissionen in Europa und Ostasien sowie auf die Prozesse in der Nähe von Ballungszentren, die die chemische Zusammensetzung von Aerosolpartikeln, deren Azidität und deren Oxidation bestimmen. Dafür wurden flugzeugbasierte in situ Messungen durchgeführt, um Aerosolpartikel im Größenbereich von 40 bis 800 nm mithilfe eines Aerosolmassenspektrometers zu messen. Die Analyse wird durch Spurengasmessungen und Daten aus verschiedenen Modellen zur Luftmassenherkunft ergänzt.

Als Ergebnis zeigten sich charakteristische Unterschiede in der regionalen und vertikalen Verteilung der Aerosolpartikelzusammensetzung. Die maximalen Aerosolmassenkonzentrationen wurden in den unteren 4 - 5 km gemessen, wobei höhere Werte von bis zu $34 \mu\text{g m}^{-3}$ in Ostasien vorkamen, während in Europa maximal $18 \mu\text{g m}^{-3}$ gemessen wurden. In Europa dominieren organische Verbindungen die Aerosolzusammensetzung, während in Ostasien Sulfat dominiert. Der Anteil an Nitrat im Aerosol steigt mit zunehmender Luftverschmutzung. Außerdem konnten Schwefelemissionsquellen von Kupferhochöfen, Schiffen und Vulkanen mithilfe der Aerosolazidität identifiziert werden. Die Analysen der Luftmassenherkunft zeigten, dass anthropogene Emissionen über weite Distanzen transportiert werden. Während des Transports werden organische Aerosolkomponenten oxidiert, was häufig mit den Zeitskalen der Prozessierung flüchtiger organischer Komponenten und anderer Spurengase korreliert. Dieser Zusammenhang liefert Informationen über das Alter der Luftmassen und wurde mit den mittleren Altern aus Dispersionsrechnungen kombiniert. Es wurden jedoch auch Unterschiede bei der Alterung von Aerosolpartikeln und Spurengasen festgestellt, da viele Faktoren beteiligt sind, die die Komplexität der Daten erhöhen.

Zusammenfassend wurden im Rahmen dieser Arbeit charakteristische Aerosolzusammensetzungen in den Regionen Europa und Ostasien sowie für ausgewählte urbane Ballungszentren festgestellt. Darüber hinaus wurde die Aerosolazidität untersucht und Einblick in die Oxidationsprozesse organischer Aerosolkomponenten in den Abluftregionen von Ballungsräumen gegeben. Diese Ergebnisse verbessern das Verständnis von Aerosolpartikeln in anthropogen beeinflussten Luftmassen.

ABSTRACT

Major population centres (MPCs) are significant contributors to anthropogenic air pollution. Since the worldwide population and degree of urbanisation are steadily increasing, the control of anthropogenic (mega-)city emissions is expected to become more important in the future. This study focuses on urban emissions in Europe and East Asia as well as processes controlling the aerosol particle chemical composition, acidity, and oxidation in the vicinity of MPCs. Therefore, airborne in situ measurements of aerosol particles in the size range of 40 - 800 nm were conducted using a compact time-of-flight aerosol mass spectrometer (C-ToF-AMS). The analysis is complemented by trace gas measurements and data from different air mass history models.

This study presents differences and characteristic signatures in the regional and vertical distribution of the aerosol particle composition. Maximum aerosol mass concentrations are observed in the lowest 4 - 5 km with larger values in East Asia of up to $34 \mu\text{g m}^{-3}$ compared with around $18 \mu\text{g m}^{-3}$ in Europe. Organic aerosol is the main aerosol component in Europe while sulfate aerosol dominates in East Asia. However, the fraction of nitrate aerosol increases with an increasing pollution level. Moreover, the aerosol acidity allowed the identification of sulfur emissions from copper smelters, ships, and volcanoes. Air mass history analysis shows that anthropogenic emissions are transported over long distances. During transport, organic aerosol compounds become oxidised which often correlates with processing scales of volatile organic compounds and other trace gases. This correlation gives insights on the age of air masses and is further combined with mean ages derived from dispersion modelling. However, differences in aerosol particle and trace gas ageing were found as many factors are involved increasing the complexity of the data.

In conclusion, this thesis reveals characteristic aerosol compositions for Europe and East Asia in general as well as for selected MPCs. In addition, an analysis of aerosol acidity is provided and valuable insights into oxidation processes of organic aerosol in the outflow of MPCs are offered. Together, these findings improve our knowledge of aerosol particles in anthropogenically influenced air masses.

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Part I

THESIS

INTRODUCTION

1.1 MOTIVATION

1.1.1 *Air quality and health effects*

Air pollution is a phenomenon that has always existed due to natural sources like volcanoes, fires, and sandstorms. Anthropogenic emissions began when wood was burned for the first time and land was cleared which promoted windblown dust. People were aware of the problem of air pollution and written accounts occurred quite early: Around 400 BC, Hippocrates mentioned the importance of good air quality and pure water to human health in the context of choosing settlement sites (Hippocrates, 400 BC). Furthermore, the citizens of ancient Rome described the situation of heavy pollution caused by emissions trapped under strong inversions as *gravioris caeli* (heavy heaven) and *infamis aer* (infamous air) (Hughes, 2014).

Air pollution is defined as the presence of at least one, mainly anthropogenic, pollutant in the atmosphere which exceeds its normal ambient concentration. The pollution could be comprised of a complex mixture of solid particles, liquid droplets, as well as gases. This results in a measurable and undesirable effect on humans, animals, vegetation, or materials and can cause a significant reduction of visibility. A polluted episode persists if the pollutants are emitted or formed faster than they are dispersed or removed from the atmosphere (Wallace and Hobbs, 2006; Seinfeld and Pandis, 2016). In other words, good air quality is defined as the absence of such harmful substances. The topic includes concerns about e.g. outdoor urban smog, indoor air pollution, acid deposition, Antarctic ozone depletion, global ozone reduction, and global warming (Jacobson, 2002). Most pollutants originate from human activities. Therefore, improvements are possible by tackling the key sources of outdoor air pollution like transport, energy generation, industry, and waste management.

Air quality is affected by several factors that can accumulate or distribute contaminants. For example, pollutants are moved by wind which can improve the air quality at one place within minutes while degrading conditions elsewhere. Geographic features like mountain ranges can cause air pollutants to accumulate upwind when the wind is blowing towards them. Furthermore, temperature plays a role. Vertical transport of emissions arise from ascending heated air in summer diluting ground-based concentrations while enhanced photochemistry can lead to more harmful ground-level ozone (O₃). In con-

trast, cold air can trap pollutants at the surface during winter. Locally, pollutants can be removed by chemical reactions or deposition. Another aspect influencing air quality is the presence of clouds which modify the concentrations by reducing photolysis or dissolving pollutants (Jacobson, 2002; Pérez et al., 2020).

At first, air pollution was thought to be a local problem like the Great Smog of London in 1952 which mainly affected urban centres and industrialised regions. Moreover, the focus of early policies was solely on improving health conditions although there had been concerns about ecological effects in some papers. This could also be due to the fact that high-quality and quantitative measurements have only existed for 150 years and numerical modelling for the last 40 years (Fowler et al., 2020). One solution to increase air quality in the 1950s was the installation of tall chimney stacks to prevent the accumulation of emissions in the cities. Meanwhile, the sulfur dioxide (SO_2) emissions in the countryside reached their maximum. It took until the end of the 1960s for air pollution to be recognised as a global problem as the Swedish scientist Svante Oden stated the consequences of long-range transported sulfur in Europe. More precisely, he presented that sulfur emissions from countries like Great Britain, Germany, France, and Poland were causing widespread acidification of lakes and rivers in Scandinavia resulting in loss of fish populations and furthermore, decline of forests (Fowler et al., 2020; Oden, 1968; Mosley, 2014). In June 1972, the first discussions about the earth's status were held at the United Nations conference on the human environment leading to the convention on long-range transboundary pollution in 1979 (Mosley, 2014). Nowadays, it is clear that air pollution affects all areas and that dense urban centres are just hotspots of globally distributed emissions.

Precise measurements and comparable standards are needed to track and improve air quality. First efforts to introduce such measures as a base for air quality policies were taken in the 1960s, e.g. the National Air Pollution Control Administration invented an air quality index in the USA in 1968 and used it in metropolitan statistical areas (Fensterstock et al., 1969; K and Kumar, 2022). One initial aim was to inform the public about air pollution issues but also to bring local officials into action. Apart from many local air quality indices, the World Health Organization (WHO) air quality guidelines (AQG) provide global thresholds for key air pollutants which are considered to be a health risk. These guidelines are based on evidence and of a high methodological quality. They were developed under a transparent process involving many specialists and are updated on a regular basis to include the latest research. The last update was published in 2021 and the actual global target values are listed in Table 1.

Clean air is fundamental to health and improving the air quality is essential to avoid health impacts that could be severe. Air pollutants with the strongest health concerns include particulate matter (PM), carbon monox-

Table 1: Actual WHO air quality guidelines (World Health Organization, 2021). The O_3 peak season value averages the daily maximum 8 h concentration over the six consecutive months with the highest concentrations.

POLLUTANT	AVERAGING TIME	AQG LEVEL
$PM_{2.5}$	annual	$5 \mu\text{g m}^{-3}$
PM_{10}	annual	$15 \mu\text{g m}^{-3}$
O_3	peak season	$60 \mu\text{g m}^{-3}$
NO_2	annual	$10 \mu\text{g m}^{-3}$
SO_2	24 h	$40 \mu\text{g m}^{-3}$
CO	24 h	4mg m^{-3}

ide (CO), O_3 , nitrogen dioxide (NO_2), and SO_2 (Wallace and Hobbs, 2006; Manisalidis et al., 2020; World Health Organization, 2021). However, they often increase simultaneously which makes it difficult to separate the health effects of the different pollutants. More on the sources of air pollution will be elaborated in Sect. 1.1.3. Almost every organ in the human body can be affected by air pollution. The main pathway of exposure is through the respiratory tract and fine PM is especially dangerous as those small particles can penetrate deep into the lungs. There, they enter the bloodstream and are distributed through the entire body where they potentially lead to systemic inflammation, oxidative stress, immunosuppression, and mutagenicity. As a consequence of impacting the lungs, heart, and brain among other organs, diseases like stroke, heart disease, lung cancer, and both chronic and acute respiratory diseases, including asthma, occur (Wallace and Hobbs, 2006; Schraufnagel et al., 2019; Manisalidis et al., 2020; World Health Organization, 2021). There is also evidence that ultrafine particles with diameters smaller than $0.01 \mu\text{m}$ have enhanced toxicity (Schraufnagel, 2020; Moreno-Ríos et al., 2022). Other factors need to be considered when analysing the health effects of air pollution like the nature of pollutants and their concentration as well as the exposure time. Additionally, the receptors age and health status matter, e.g. children, elderly, and pregnant women are more sensitive. Maternal exposure to air pollution is related to adverse birth outcomes but also premature deaths occur more frequently (Hansen et al., 2006; Dadvand et al., 2013; Lelieveld et al., 2015b; Nault et al., 2021).

Recently, an updated data set on the reduction of life expectancy was published confirming previous findings (Lelieveld et al., 2015b; Carvalho, 2019; Chowdhury et al., 2022; Pozzer et al., 2023). The data set comprises an Air Quality Life Index based on satellite-derived particulate matter with a diameter of $2.5 \mu\text{m}$ or less ($PM_{2.5}$) combined with detailed global population data and calculates the gain of life expectancy if the WHO guidelines were

met (Chen et al., 2013; Ebenstein et al., 2017; Greenstone and Hasenkopf, 2023). The main finding of this study is that long-term exposure to an additional $10 \mu\text{g m}^{-3}$ of $PM_{2.5}$ reduces life expectancy by 0.98 years (Ebenstein et al., 2017). Globally, a compliance with the WHO limits could raise average life expectancy by 2.3 years and compared to other risk factors air pollution is similar to that of smoking, more than three times worse than alcohol abuse or unsafe water, and more than five times worse than transport injuries (Greenstone and Hasenkopf, 2023). Furthermore, the most impacted areas are Bangladesh, India, Nepal, and Pakistan. Air pollution is reducing the life spans of South Asians by 5.1 years while China managed to reduce pollution by 42.3 % between 2013 and 2021. European policies reduced pollution levels by 23.5 % since 1998 and gained their citizens 4.5 months. Another 8.4 months would be possible if the actual WHO thresholds were met (Greenstone and Hasenkopf, 2023).

1.1.2 *Air pollution and megacity emissions*

Air pollution has a special significance when it comes to cities and urban environments due to several reasons. First, more than half of the world's population lives in urban areas with a persistent increasing trend (United Nations, 2019, 2022). Second, the required energy production together with economic and industrial performance make cities a strong local source with a high burden of anthropogenic pollution (Molina et al., 2020). Third, almost the entire global population (99 %) breathes air that exceeds WHO air quality limits and related to the 117 countries monitoring their air quality, 17 % of cities in high-income countries comply with the WHO guidelines for particulate matter while only 1 % of the cities in low- and middle-income countries fulfil them (World Health Organization, 2022; Yu et al., 2023). Fourth, mortality in urban environments is 50 % higher than in rural areas as air quality in the former can be significantly worse due to a concentration of anthropogenic emission sources (Lelieveld et al., 2015b).

In 2007, the urban population was as large as the one in rural areas for the first time (United Nations, 2019). Studies on urbanisation trends project that around 60 % of the world's population will live in cities by 2030 and almost 70 % by 2050 (United Nations, 2019). Furthermore, the world's cities are growing in size and number as well as emissions. Based on United Nations (2019), the level of urbanisation in Europe is already at 75 % and in Asia around 51 % which is expected to grow much faster (+15 %) than in Europe (+9 %) until the mid of the century. This growth of urban areas occurs on less than 3 % of the global land surface which results in a high population density (Liu et al., 2014). However, a slowdown of land take for residential use in Europe has been observed after the global financial crisis of 2008 although

the trend, elaborated from 331 European cities, is towards densification of cities (Cortinovis et al., 2022).

Various reasons make people move from the countryside to cities. For example, urbanised centres provide better job opportunities, higher education for children, or a broad range of goods and shared services. Urban environments are places that promote human development by the generation of knowledge, technology, and wealth (Gurjar and Lelieveld, 2005; Kraas, 2007). Furthermore, migration is caused by conflicts, land degradation, and the depletion of natural resources. However, the greatest contributor is the general population growth, especially in the developing world (Molina and Molina, 2004; Montgomery, 2008; Baklanov et al., 2016).

The largest urban agglomerations with more than 10 million inhabitants are often called megacities (Molina and Molina, 2004; Gurjar and Lelieveld, 2005). They are centres of growing economies and large sources of air pollutants. However, there is no exact definition of the boundaries of such megacities. Therefore, the term major population centre (MPC) can be utilised to describe a metropolitan area or converging urban conurbations which forms an important regional source of anthropogenic emissions. MPCs enclose people who do not reside within the cities' political boundaries but might share employment, housing, transportation, and security needs (Molina and Molina, 2004). In 1950, the world had only two megacities: New York and Tokyo (BISWAS, 2010). Their number has increased significantly in recent decades, especially in the developing countries, yielding 33 megacities and 48 urban agglomerations with 5 to 10 million inhabitants in 2018 (United Nations, 2019). MPCs are more abundant in Europe than megacities, of which there are only two (Istanbul and Moscow) today while Asia is having 20 megacities. According to projections, the number of megacities will increase to 43 in 2030, Delhi in India will overtake Tokyo in Japan as the world's largest city, and London will grow to the third megacity in Europe (United Nations, 2019).

Emission sources are different between (mega-)cities and, thus, are the impacts on air quality and climate. This is due to their level of economic development, e.g. emissions from transport, energy production, and industry are more important in developed cities, whereas biomass burning, mainly released during domestic activities like cooking and heating, is predominant in developing cities (Lelieveld et al., 2015b; Crippa et al., 2021). One disadvantage of megacity development is the strong release of air pollutants, often decreasing the air quality drastically both locally as well as in regions downwind (Molina and Molina, 2004). Furthermore, the heat island effect increases the temperature compared to the surrounding area which causes additional heat stress to the residents, especially in summer (Heaviside et al., 2017). Additionally, the release of long-lived greenhouse gases and short-lived climate-active pollutants are drivers of climate change (Folberth et al., 2015).

However, consequent policies and joint efforts can be a significant chance for climate change mitigation by reduced emissions. More efficient energy use, fast and convenient public transport, improved vehicle mileage standards, and more energy-efficient buildings are just some of the possibilities for improvement (Grimm et al., 2008; Parrish and Zhu, 2009).

Some examples of successful policies are given in the following: First, the "Air Pollution Control Act" in the USA in 1955 and the "Clean Air Act" in the United Kingdom in 1956 as well as their consecutive regulations were a direct reaction to heavy smog episodes aiming at a reduction and control of hazardous air pollutants (Jacobson, 2002). Second, the "Convention on Long-range Transboundary Air Pollution" in 1979 cut SO₂ emissions to fight the consequences of acid rain and also initiated various research and monitoring programmes. The initial document have been extended eight times to take additional pollutants, e.g. nitrogen oxides (NO_x), volatile organic compounds (VOCs), heavy metals, and ground-level O₃, into account. The environment in Europe is beginning to recover although forests face other problems like global warming today (Mosley, 2014; Grennfelt et al., 2020). Third, the "National Air Quality Action Plan" in China announced in 2013 managed to reduce particulate pollution by 42.3 % between 2013 and 2021 (Greenstone and Hasenkopf, 2023). Furthermore, the introduction of various restrictions and requirements (low-sulfur fuels, flue gas desulfurisation, etc.) initiated a general trend in China showing a decrease in SO₂ emissions over the last few years (McDuffie et al., 2020). Lastly, the "zero pollution action plan" was announced in May 2021 by the European Commission as part of the European Green Deal. It includes a vision for 2050 to reduce air, water and soil pollution to levels no longer considered harmful to health and natural ecosystems as well as interim targets for 2030, e.g. to reduce the number of premature deaths due to exposure to fine PM by 55 % by 2030 compared to 2005 (European Commission, 2021). Regular monitoring and outlook assessments will evaluate the progress for every section included in the plan, e.g. premature deaths were reduced by 45 % in 2020 (European Environment Agency, 2022).

In conclusion, air pollution is an important but avoidable risk to health globally. Accompanying effects of long-term exposure to pollutants often go unnoticed or are not easily measured. However, the public concern about air quality and climate change increases which also results in an enhanced interest on studies about the influence of MPC emissions and the development of adequate control strategies. Emission reductions can lead to improvements as the previously presented examples show. Therefore, air quality monitoring by automatic measurement stations and specific research programmes is essential to make further progress and adjust existing policies.

1.1.3 *Natural and anthropogenic emissions*

The air quality is influenced by emissions from both natural and anthropogenic sources. They emit a variety of primary aerosol species and precursor gases which will be introduced in the following.

Natural emissions

Natural as well as anthropogenic biomass burning (BB) of vegetation and forests results in a major source of combustion products like carbon dioxide (CO_2), CO, NO_x , methane (CH_4), non-methane hydrocarbons (like acetonitrile), black carbon (BC) and organic aerosol (OA). BC particles from vegetation fires can be thickly coated with organics and inorganics indicating an intense aerosol processing during transport (Kondo et al., 2011). The quantity and type of emissions are determined by the type of vegetation but also other factors play a role like moisture content, ambient temperature, humidity, and local windspeed (Seinfeld and Pandis, 2016).

Marine emissions often contain sea spray aerosol particles which are a complex mixture of organic matter and inorganic salts like sodium, chloride, magnesium, and calcium (Boucher, 2015). Aged sea spray aerosol particles can be internally mixed with secondary organics, nitrate, and sulfate based on reactions with organic, nitric, and sulfuric acids, respectively (Laskin et al., 2012). Another substance released from the ocean and produced by marine phytoplankton is dimethyl sulfide (DMS) which is processed to methane-sulfonic acid (MSA), sulfuric acid, and sulfate and thus is a contributor to new particle formation (Seinfeld and Pandis, 2016).

Plants and vegetation emit bioaerosols, such as pollen, bacteria, and plant particles as well as VOCs like isoprene which can be precursor gases for new particle formation (Churkina et al., 2017).

Volcanic emissions consist of a mixture of gases, aerosol, and silicate particles e.g. SO_2 , airborne ash, and sulfate aerosol. The duration of atmospheric and climatic effects depends on the injection altitude of pollutants, e.g. SO_2 in the stratosphere has a longer lifetime than in the lower atmosphere. Furthermore, volcanic emissions can transport halogens, trace metals, and metalloids which impact local environments at the ground substantially and can cause economic losses (Durant et al., 2010).

Dust episodes contribute directly to air pollution by increasing the PM concentrations. The major dust sources are natural and a consequence of wind acting on undisturbed source regions. They are located in the arid and semiarid regions of the world with the Sahara-Sahel region in Northern Africa being the largest global source (Boucher, 2015). However, human activities can also cause dust emissions as land-use changes modify and increase soil surfaces or climate modifications favour dust emissions by changes

in windspeeds, clouds and precipitation (Seinfeld and Pandis, 2016). The major removal processes for dust are gravitational settling and wet deposition. During their atmospheric lifetimes of about two weeks, dust particles can be transported far away from their source. Thus, they are an important transboundary component (Seinfeld and Pandis, 2016).

Anthropogenic emissions

Anthropogenic pollutants originate from agriculture, the energy sector, industry, housing, transport, and mining as well as fossil fuel, biomass, and waste combustion. In many regions, the sum of the anthropogenic emissions dominate over natural emissions and can cause severe air pollution locally or downwind (Jacobson, 2002).

CO is a toxic gas produced by the incomplete combustion of carbonaceous fuels like wood, petrol, charcoal, natural gas, and kerosene. It is predominantly emitted by combustion engines. CO is typically used as a tracer for anthropogenic and combustion-related pollution due to its relatively long lifetime of 1 - 3 months in comparison to other pollutants (Seinfeld and Pandis, 2016).

NO_x (nitric oxide (NO) and NO₂) is produced by high temperature combustion of fossil fuels like in heating, industry, power generation, and transportation as well as biomass burning. It is further emitted from soils, lightning, ammonia oxidation, aircraft emissions, and transport from the stratosphere (Wallace and Hobbs, 2006).

O₃ at ground-level is formed through the reaction with gases like VOCs, CO, and NO_x in the presence of sunlight (Crutzen, 1988). It is one of the major components of photochemical smog and is harmful to health.

Anthropogenic SO₂ is produced from the burning of fossil fuels for domestic heating, industries, and power generation. Furthermore, the smelting of sulfur-containing mineral ores is a source for SO₂ as well as international shipping which causes air pollution and is still poorly regulated (Jonson et al., 2020).

Anthropogenic VOCs are emitted e.g. by motor vehicles, mainly in the form of hydrocarbons. They are generated during the incomplete combustion and from the vaporisation of fuel (Wallace and Hobbs, 2006). VOCs undergo atmospheric oxidation and further chemical processing, leading to a reduction of saturation pressure and thus to aerosol formation (Zhang et al., 2007a; Hallquist et al., 2009; Jimenez et al., 2009; Shrivastava et al., 2017).

Anthropogenic PM is either derived from primary sources, e.g. combustion of fuels in power generation facilities, industries, and vehicles, or secondary sources like chemical reactions between gases (Jacobson, 2002). Further sources are household activities like cooking, residential heating, or lighting as well as particles generated from traffic and transportation, industrial

activities, power plants, construction sites, waste burning, or fires (Seinfeld and Pandis, 2016). PM comprises sulfate, nitrates, ammonium, sodium chloride, black carbon, or water. Globally, the fraction of anthropogenic PM is around 20 % by mass (Wallace and Hobbs, 2006).

The life cycle of trace species starts with the emission at the source, continues with transformation during transport, and ends with the removal from the atmosphere. The transport of substances is influenced by the large-scale circulation as well as local and regional weather phenomena (Pérez et al., 2020). Thereby, aerosol particles undergo physical and chemical changes, whereas trace gases either go through kinetic reactions or are photolysed (Pöschl, 2005). The atmospheric lifetimes of trace gases and thus their distribution in the atmosphere are dependent on their solubilities and reaction rates. Therefore, the lifetime varies from a few hours to days, like for NO_x , to weeks, as is the case with O_3 , up to several months, as for CO, or even several years as for CO_2 (Jacobson, 2002). On the contrary, the bulk of tropospheric aerosol particles has lifetimes between a day and two weeks, which are mostly influenced by their size and solubility (Jaenicke, 1980; Schmale et al., 2011; Kristiansen et al., 2016). Removal processes are wet and dry deposition.

1.1.4 *Regional to global impacts of air pollution*

Air pollution is observed from regional to global scales. Aerosol particles which are more short-lived are generally thought to undergo less transport and contribute more to regional pollution than long-lived trace gases which travel long distances. Nevertheless, observations and model simulations indicate that aerosol particles can also travel long distances in the atmosphere (Chin et al., 2007; Wagstrom and Pandis, 2011; Weinzierl et al., 2017). MPC emissions are processed chemically and physically while they are affected by mixing with natural, biogenic, and other anthropogenic emissions transported from regional or long-range sources (Lawrence et al., 2007).

Regional impacts

A portion of the emissions remain around the source where they contribute to an increase in pollution. MPCs and larger cities experience haze episodes regularly which reduce visibility significantly and have effects on human health, animals, and plants (Pandis et al., 1995; Wallace and Hobbs, 2006; Seinfeld and Pandis, 2016). Wet and dry deposition can lead to soil and (ground-)water acidification which erodes buildings as well as other materials and causes many environmental and economic losses (K and Kumar, 2022). The impacts are largest close to the source where the emissions are almost undiluted. Although anthropogenic emissions were reduced in recent

years, air quality levels are still often violated and air pollution occurs in several areas of Europe and around the world (European Environment Agency et al., 2019; Fowler et al., 2020).

Long-range transport

Emissions of MPCs do not only remain at the source location but can be transported over long distance in the (planetary) boundary layer (BL) and the free troposphere, thereby changing the atmospheric gas-phase and aerosol composition (Butler and Lawrence, 2009; European Environment Agency et al., 2019; World Health Organization, 2021). Thus, the regional boundaries of a metropolitan region do not confine the distribution of polluted air masses. Moreover, the emissions have an effect on the areas downwind of their source and can also contribute to global air pollution (Folberth et al., 2015; Baklanov et al., 2016). In model simulations, about half of the artificial $PM_{2.5}$ aerosol tracers originating in MPCs ended up more than 1000 km away from their source (Kunkel et al., 2013).

The transport paths of trace substances depend on the geographical location (Lawrence et al., 2007; Cassiani et al., 2013; Folberth et al., 2015). In the middle and high latitudes, air pollutants are mainly transported close to the surface within the BL. This was confirmed e.g. through a model study whereby a tracer originating in Eurasia was released with a lifetime of 10 d. A proportion of 17 – 34 % was transported long-range more than 1000 km and stayed below 1 km (Lawrence et al., 2007). In contrast, deep convection is the main process for long-range transport in the tropics.

Some examples of air pollution transport patterns are given in the following. European air quality is frequently influenced by long-range transport from North America (Huntrieser et al., 2005; Roiger et al., 2012). Furthermore, air masses originating in Asia are observed in the Mediterranean after circling the globe (Lelieveld et al., 2002; Lawrence and Lelieveld, 2010; de Meij et al., 2012). A driving force of vertical air mass distribution from Asia is the Asian summer monsoon pumping up natural emissions and anthropogenic pollutants which form the Asian tropopause aerosol layer. The pollutants are further distributed from there preferentially west and east due to the weaker wind gradients (Vernier et al., 2009; Pan et al., 2016; Zhang et al., 2019; Appel et al., 2022). In spring, Chinese anthropogenic pollution is transported to the Western Pacific after frontal and orographic lifting to the free troposphere while the transport in the BL occurs behind the cold front (Liu et al., 2003; Russo et al., 2003). Furthermore, BB emissions from South-East Asia are taken upwards in warm conveyor belts as well as deep convection and mix with the anthropogenic pollution (Liu et al., 2003; Hadley et al., 2007). While the BL outflow occurs mainly around 30 - 45° N, the frontal outflow to the North-West Pacific is strongest between 20 - 35° N and

often limited between 2 and 6 km altitude with a maximum at about 4 km (Liu et al., 2003). Low latitudes below 35° N and altitudes of 7 - 12 km are the typical locations for convective outflow of Asian emissions (Liu et al., 2003). Another example for long-range transport of pollutants caused by fossil-fuel combustion, smelting, and other industrial processes in Northern Europe and Russia are Arctic haze episodes. They develop primarily from December to April and can persist over some time due to a stable Arctic atmosphere which limits vertical mixing. Furthermore, precipitation is weak during this time which increases the lifetimes of pollutants (Wallace and Hobbs, 2006).

Global impacts and climate effects

Today, impacts of anthropogenic pollution are visible globally. For example, CO₂ has been increasing worldwide since the industrial revolution as well as other trace gases like CH₄ and nitrous oxide have been rising over the past 170 years (Wallace and Hobbs, 2006). Moreover, ocean acidification and stratospheric ozone loss are global problems brought on by air pollution (K and Kumar, 2022). Greenhouse gas emissions have continued to drive climate change and increase global mean surface temperature. In contrast, aerosol particles alter the earth's radiation balance by increasing the number of cloud condensation nuclei (CCN) and changing cloud properties. This has on average a negative radiative forcing and masks the global warming (Pandis et al., 1995; IPCC et al., 2023). Further risks caused by climate change will affect coastal cities in particular, e.g. sea level rise as well as an increased hurricane frequency and strength (Grimm et al., 2008; Holland and Bruyère, 2014; Bhatia et al., 2018; Bruyère et al., 2019).

1.2 ANTHROPOGENIC AEROSOL

1.2.1 *Formation and size distribution*

Urban aerosol particles have a clear source characteristic and are anthropogenic in origin. Sulfate, nitrate, ammonium, elemental and organic carbon, as well as crustal chemicals (silicon, aluminum, calcium, and iron oxides) are the major components of urban aerosol (Pandis et al., 1995).

A distinction is made between primary and secondary aerosol particles. Primary aerosol particles are emitted directly from the source into the atmosphere. BC and organic matter are components found in primary aerosol particles produced by human activities, e.g. during fuel combustion, industrial processes, or transportation (Tomasi et al., 2017). On the contrary, secondary aerosol particles are formed after chemical reactions in the gas phase. They are either formed by gas-to-particle conversion of NO_x, hydrocarbons,

ammonia, and SO_2 or condensation of gaseous compounds on pre-existing aerosol (Pandis et al., 1995; Monks et al., 2009).

Number concentrations of urban aerosol particles are usually greater than 10^5 cm^{-3} and size distributions are comprised of four modes: nucleation, Aitken, accumulation, and coarse mode (Pandis et al., 1995). The ambient aerosol size distribution is characterised by different processes occurring within the aerosol life cycle, e.g. direct particle emission, in situ formation, interactions between particles or gases, and deposition (Wu and Boor, 2021). Particles in the nucleation mode (3 to about 20 nm) are newly formed by the nucleation of gaseous molecules and ions (Brines et al., 2015). Primary emitted particles from combustion sources and condensation of secondary materials are found in the Aitken (roughly 20 to 100 nm) and accumulation mode (100 to 1000 nm) (Yue et al., 2009). Particles in the coarse mode are larger than 1000 nm and result from mechanical processes like aerodynamic resuspension and abrasion (Wu and Boor, 2021). Therefore, they can contain road dust as well as tire debris (Adachi and Tainosho, 2004; Almeida et al., 2006). In the urban environment, the particle number distribution is dominated by particles in the nucleation and Aitken mode due to the prevalence of primary emission sources. Close to the source, the concentrations of these fine particles are high and decrease quickly with distance (Zhu et al., 2002). Particles grow during transport by condensation of secondary materials or water. Generally, sinks of aerosol particles are related to their size. Particles in the nucleation mode are highly diffusive and therefore, can be removed quickly via coagulation (Hinds and Zhu, 2022). In addition, they can grow into the Aitken mode accompanying new particle formation (Cai et al., 2017). Coagulation and condensation of particles lead to further growing from the Aitken into the accumulation mode. Particles in the accumulation mode have the longest lifetime because their gravitational settling velocities are low and the coagulation rates among themselves are low, too (Wu and Boor, 2021). Coarse-mode particles have high gravitational settling velocities which results in 2 – 4 orders of magnitude lower number concentrations compared to the other modes (Wu and Boor, 2021).

1.2.2 *Global composition of aerosol particles*

A quantification of global aerosol sources can only be roughly determined. However, the uncertainties have decreased significantly in recent years due to the increasing availability of satellite data and specially developed global aerosol models (Kanakidou et al., 2005; Textor et al., 2006; Dubovik et al., 2008). The geographical distribution of the most important anthropogenic aerosol sources for the year 2000 is given in Roedel and Wagner (2013). Therein, source strengths for anthropogenic sulfate aerosol (SO_4) are enhanced over the continents, whereby they are greatest in the Northern

hemisphere, especially at the east coast of North America, in Central Europe, and East Asia. Chin et al. (1996, 2000) reported that in a year without a major volcanic eruption around 80 % of the sulfate precursor emissions are from anthropogenic sources while the rest is biogenic or volcanic. Roedel and Wagner (2013) further explained that maxima in the anthropogenic OA production occur in Central Europe, East Asia, and Brazil, followed by West Africa. Lastly, anthropogenic soot emissions are greatest over Central Europe as well as East Asia and enhanced in Brazil and West Africa (Penner et al., 2001; Roedel and Wagner, 2013).

OA represents a significant and often major fraction of the sub-micron aerosol mass. At continental mid-latitudes, the OA fraction can range between 20 and 50 % (Putaud et al., 2004) whereas up to 90 % are possible in tropical forested areas (Andreae and Crutzen, 1997). In a global compilation of ground-based measurements, OA comprised a major fraction of 18 – 70 % with an average of 45 % (Zhang et al., 2007a). Furthermore, SO_4 mass fractions contributed with 10 - 67 % (average: 32 %) and nitrate aerosol (NO_3) with 1.2 – 28 % (average: 10 %). The ammonium aerosol (NH_4) proportion was 6.9 – 19 % (average: 13 %) while chloride was often below its detection limit or accounted for maximum 4.8 % (average: 0.6 %) (Zhang et al., 2007a). In highly polluted megacities, like Beijing and Mexico City, the OA mass concentrations were 4 to 5 times higher compared to the average of the other cities included in the study (Zhang et al., 2007a).

The aerosol particle composition in the remote atmosphere was explored during the Atmospheric Tomography (ATom) mission. Hodzic et al. (2020) found that OA and SO_4 were the main aerosol components. In winter 2017, sea salt made up around 30 % in the marine BL while SO_4 was prevalent in the lower stratosphere with up to 70 %. Additionally, a strong seasonal and zonal variability was observed for OA with the highest levels measured in the lower troposphere in summer 2016 and over regions influenced by BB from Africa. Furthermore, the OA measured in the remote free troposphere was highly oxygenated (Hodzic et al., 2020).

1.2.3 *Processing of organic aerosol particles*

OA comprises a variety of chemical compounds that are either emitted directly as primary organic aerosol (POA) or formed from the oxidation products of gas-phase precursors as secondary organic aerosol (SOA) (Hallquist et al., 2009; Heald et al., 2011; Seinfeld and Pandis, 2016). Another way to define umbrella terms is to use the terms hydrocarbon-like organic aerosol (HOA) and oxygenated organic aerosol (OOA). HOA is associated with primary emissions from combustion while many studies link OOA to SOA in or downwind of urban areas (de Gouw et al., 2005; Zhang et al., 2005; Volkamer et al., 2006; Herndon et al., 2008). Furthermore, OOA was found to

dominate the OA fraction in the free troposphere downwind of East Asia (Bahreini et al., 2003; Heald et al., 2005; Zhang et al., 2007a). Based on Zhang et al. (2007a), HOA levels of around 36 % are comparable between the included ground-based stations in urban areas, except for the heavily polluted regions where HOA makes a greater contribution to the aerosol mass. On the contrary, OOA is prevalent in the urban downwind and especially in the rural/remote locations of the study with up to 95 % OOA because HOA was diluted with increasing distance (Zhang et al., 2007a).

Jimenez et al. (2009) described the connection between oxidation and volatility. During oxidative processing and SOA formation, precursor gases like VOCs are oxidised, forming substances of lower volatility which partition into the particle phase. Thus, OOA evolves from semi-volatile OOA (SV-OOA) to low-volatile OOA (LV-OOA) (Alfarra et al., 2004; Zhang et al., 2005; Zhang et al., 2007a; Canagaratna et al., 2007; Jimenez et al., 2009; Ng et al., 2010; Ortega et al., 2016). Nowadays, the terms "less-oxidised OOA (LO-OOA)" and "more-oxidised OOA (MO-OOA)" are preferred when no clear correlation between the oxygen-to-carbon ratio and the aerosol volatility is observed (Hildebrandt et al., 2010; Xu et al., 2014; Xu et al., 2015; Setyan et al., 2012).

Another aspect altered by ageing of particles and their precursors is hygroscopicity which increased with ageing. This directly influences particle properties like size, scattering and cloud condensation nuclei capability and thus has an impact on radiative forcing and climate (Jimenez et al., 2009; Lambe et al., 2011).

1.2.4 *Open research questions*

Some of the open research questions regarding air pollution, anthropogenic emissions, and OA should be mentioned in the following.

Urban air pollution remains a major environmental health threat. Therefore, continuous aerosol measurements are crucial to control the extent of urban air pollution, identify further pollution sources, and understand aerosol transformation mechanisms (Asmi et al., 2011; Peng et al., 2014). Further studies are needed to characterise human exposure and health effects to improve current and future air quality policies (Shiraiwa et al., 2017; Wu and Boor, 2021). The Global Atmosphere Watch report stated that the impact of emissions on air pollution and climate are still poorly understood and imprecisely quantified, especially representative airborne measurements are required (Zhu et al., 2012). Ongoing urbanisation increases the number of people living in urban areas. Research is necessary to understand how this trend influences air pollution as well as local and global climate (Baklanov et al., 2016). Furthermore, such integrated studies can help to develop climate

adaptation strategies for cities that already face hazards like storm surge, flooding, heat waves, and air pollution episodes (Baklanov et al., 2016).

Early air quality research was focused on the situation within megacities. Therefore, studies about the regional to global impacts are lacking as well as on the mechanisms driving the pollutant dispersion in and away from the cities (Lawrence et al., 2007; Butler and Lawrence, 2009; Butler et al., 2012). Furthermore, local and regional MPC emission inventories are often inconsistent (Butler and Lawrence, 2009; van der Gon et al., 2011) and prediction capabilities limit the accuracy of pollution transport patterns in downwind regions of MPCs (Kunkel et al., 2012). Therefore, aircraft measurements and satellite observations are useful for verifying elements of emission inventories and to capture the vertical dimension of emission dispersion as well as the transformation of aerosol particles and trace gases (Molina and Molina, 2004).

Further open questions involve OA. MPCs emit gas-phase and particulate pollutants, but one of the least understood aspects are the formation and evolution of SOA during the atmospheric transport as well as residence times of the emissions (Jimenez et al., 2009). Additionally, atmospheric models tend to overestimate the primary OA fraction while underestimating SOA (Hodzic et al., 2016). Moreover, the measured OA is 30 – 60 % more oxygenated than in actual models which can result in significant errors of the calculated OA concentrations (Hodzic et al., 2020).

1.3 OBJECTIVES AND STRUCTURE OF THE THESIS

The major objectives of this thesis are to improve the understanding of urban emissions in Europe and Asia as well as their influence on the atmospheric aerosol composition. Therefore, aircraft-based measurements were performed in the project EMeRGe (Effect of Megacities on the transport and transformation of pollutants on the Regional to Global scales). The aerosol particle measurements were conducted using an Aerodyne Compact Time-of-Flight Aerosol Mass Spectrometer (C-ToF-AMS) to study the composition, distribution, and atmospheric processing of aerosol particles in the vicinity of selected MPCs in Europe and East Asia. Thus, the following research questions are addressed in this thesis:

- Does the aerosol particle composition differ between Europe and East Asia and do aerosol-specific emission signatures exist for the selected MPCs?
- Which emission sources control the aerosol acidity in the vicinity of MPCs?

- Does ageing affect the aerosol particle composition in the outflow of MPCs?

To answer these research questions, the structure of the current thesis is the following: In Ch. 1, the current understanding of the sources and impacts of air pollution were introduced, the characteristics of anthropogenic aerosol particles were presented, and the existing knowledge gaps were emphasised. Chapter 2 describes the aircraft campaign EMeRGe, the meteorological conditions during the measurements, as well as the measurement platform High Altitude and LOng range research aircraft (HALO). The following Ch. 3 contains the instruments and methods which were used for this thesis. Accordingly, the aerosol mass spectrometer as well as the applied calibrations and corrections are explained. Furthermore, complementary instruments and models as well as the data analysis methods are introduced briefly. The results of the data analysis are presented in Ch. 4 with focus on the aerosol composition, acidity, and oxidation. In the final Ch. 5, the key findings are summarised and an outlook on further research activities on urban emissions is given.

MEASUREMENT MISSION EMeRGe

The Effect of Megacities on the Transport and Transformation of Pollutants on the Regional to Global Scales (EMeRGe) project was a joint mission of several institutes (see Table 12 in the appendix and Andrés Hernández et al., 2022) lead by the Institute of Environmental Physics, University of Bremen (IUP). It aimed at the investigation of major population centre (MPC) emissions. Two field campaigns were conducted with the High Altitude and LOng range research aircraft (HALO) to probe European and Asian MPC plumes in summer 2017 and spring 2018, respectively.

In the following, an overview on the EMeRGe project and its objectives is presented, mentioning the meteorological conditions during the measurements, introducing the research aircraft HALO and the instrumentation on board. Furthermore, particle losses inside the inlet and tubing system are discussed as well as the importance and issues for a precise flight planning are explained.

2.1 OVERVIEW AND OBJECTIVES OF THE MISSION

Three main scientific topics were addressed with the EMeRGe project: the composition and transport of selected MPC emissions, their photochemical evolution on local, regional, and hemispherical scales as well as model comparisons.

An overview of the conducted flight paths is given in Fig. 1. In summer 2017, an intensive measurement campaign with seven research flights took place over Europe between 10 and 28 July. The aircraft HALO was based in Oberpfaffenhofen, west of Munich, in the south of Germany. The second campaign took place in spring between 10 March and 09 April 2018 out of Tainan, Taiwan, covering the region above the East and South China Sea. Therefore, 12 scientific measurement flights were performed in this area plus four transfer flights to and from the home airport in Germany (two with measurements above Europe and two without measurements due to country-specific restrictions).

Target cities and regions were defined beforehand using tropospheric composites of NO₂ columns retrieved from satellites. A list of target regions assigned to the different flights is given in Tables 2 and 3. The selected targets are also marked with boxes in Fig. 1 and their geographical extents are listed in Table 8. During EMeRGe-Europe, emissions from MPCs like the Belgium-Netherlands-Ruhr area (BeNeRu), London, Munich, Northern

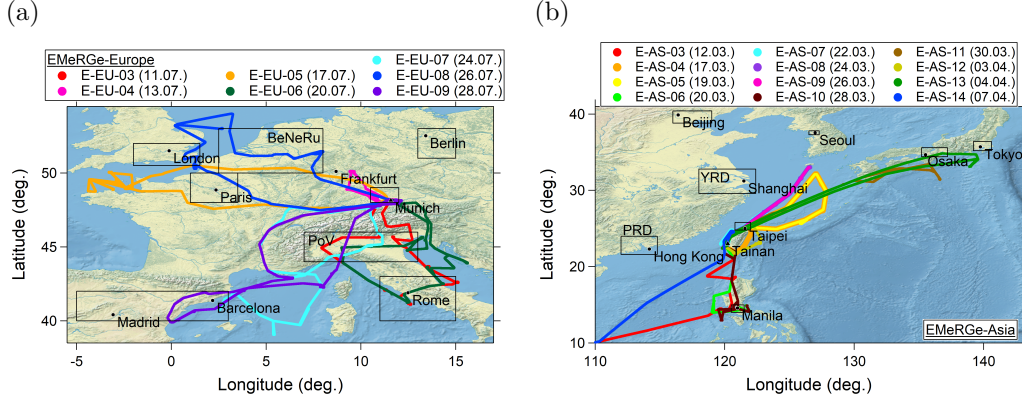


Figure 1: Flight paths of the EMeRGe campaigns in Europe in summer 2017 (a) and in East Asia in spring 2018 (b; excluding the transfer flights). The research aircraft HALO was based in Oberpfaffenhofen (22 km west of Munich), Germany and in Tainan, Taiwan, respectively. Selected cities are marked for orientation. Boxes indicate target regions around MPCs of interest, also used for the emission transport simulations described in Sect. 3.5.4. The map was created from public domain GIS data found on the Natural Earth website (<http://www.naturalearthdata.com>; last access: 12 September 2023).

Spain, Paris, the Po Valley (PoV), and Rome were studied, while during EMeRGe-Asia emission plumes from Beijing, Manila, Osaka, the Pearl River Delta (PRD), Seoul, Taipei, Tokyo, and the Yangtze River Delta (YRD) were investigated. The MPC outflows were probed upwind and downwind in a quasi-Lagrangian approach whenever possible and at different altitudes to investigate the extent of their emission plume. Further explanations and restrictions on the flight planning are given in Sect. 2.4. The comprehensive data set was analysed in combination with atmospheric transport modelling results further described in Sect. 3.5.4.

2.2 METEOROLOGICAL CONDITIONS

The month July was chosen for EMeRGe-Europe to investigate the transformation of MPC emissions under active photochemistry due to the usually high temperatures and solar radiation in summer. However, mean anomalies of the 850 hPa temperature (see Fig. 2a) during the campaign period show negative values for Northern Europe compared with a 1991 - 2020 climatology based on National Centers for Environmental Prediction (NCEP) reanalysis data (Kalnay et al., 1996). Further, the conditions in Central Europe were influenced by increased precipitation (see Fig. 2b) also affecting the flight planning as further explained in Sect. 2.4. In contrast, the situation in Southern Europe was characterised by higher temperatures, intense heat waves throughout the year, and forest fires in the Mediterranean region (Por-

Table 2: Overview of all flights of the EMeRGe campaign in Europe in 2017 including flight number, date, duration, and target regions. The flights E-EU-01 and -02 were certification and test flights which are not further discussed here.

FLIGHT	DATE IN 2017	DURATION	TARGET REGIONS
E-EU-03	11.07.	6 h 33 min	PoV, Rome
E-EU-04	13.07.	4 h 22 min	HALO-FAAM intercomparison
E-EU-05	17.07.	7 h 59 min	Paris, London, BeNeRu
E-EU-06	20.07.	8 h 28 min	PoV, Rome
E-EU-07	24.07.	8 h 34 min	PoV, Barcelona, Southern France
E-EU-08	26.07.	7 h 37 min	Paris, London, BeNeRu
E-EU-09	28.07.	8 h 35 min	Barcelona, Madrid, Southern France

Table 3: Overview of all flights of the EMeRGe campaign in Asia in 2018 including flight number, date, duration, and target regions. The flights E-AS-01 to -03 and E-AS-14 to -16 were transfer flights.

FLIGHT	DATE IN 2018	DURATION	TARGET REGIONS
E-AS-01	10.03.	7 h 54 min	(Rome, Athens, Egypt, Saudi Arabia)
E-AS-02	11.03.	-	no measurements
E-AS-03	12.03.	6 h 29 min	(Bangkok, Manila)
E-AS-04	17.03.	8 h 36 min	Taiwan, East China Sea (ECS)
E-AS-05	19.03.	8 h 5 min	Taiwan, ECS
E-AS-06	20.03.	6 h 49 min	Manila
E-AS-07	22.03.	5 h 45 min	Taiwan, ECS
E-AS-08	24.03.	8 h 52 min	ECS, Chinese outflow, Korea
E-AS-09	26.03.	9 h 2 min	ECS, Chinese outflow, Korea
E-AS-10	28.03.	8 h 39 min	Taiwan, Manila
E-AS-11	30.03.	9 h 23 min	Japan, ECS
E-AS-12	03.04.	6 h 0 min	Taiwan
E-AS-13	04.04.	8 h 57 min	Japan, ECS
E-AS-14	07.04.	7 h 40 min	(Taiwan, Bangkok)
E-AS-15	08.04.	-	no measurements
E-AS-16	09.04.	8 h 44 min	(Saudi Arabia, Egypt, Athens, Rome)

tugal, Spain, Southern France, and Italy). The fires were further intensified by dry soils and a lower precipitation rate compared to the climatological mean. Based on ERA-Interim data (Copernicus, 2018), 2017 was the fourth warmest year globally (until 2022) with an European mean surface air temperature 0.8°C higher than the climatological average (1981 - 2010) and even 1.7°C higher for South-Western Europe. The European annual precipitation was close to average (-0.02 mm d^{-1}) but soil moisture was the second lowest on record (Copernicus, 2018).

Spring 2018 was selected for EMERGe-Asia to measure the continental Asian outflow over the Pacific which reaches a maximum in this season (Bey et al., 2001; Liu et al., 2003). Further BB emissions from Indochina were expected, as well as mixing of MPC emissions with dust from the Gobi desert and observations of transpacific transport. The outflow patterns during the campaign period are depicted by the 500 hPa wind composite mean in Fig. 2c. The mean temperature was higher during the campaign in North-Eastern Asia and lower above the South China Sea (see Fig. 2d), compared with the NCEP climatology (1991 - 2020; Kalnay et al., 1996). Due to flight restrictions, the measurements took place at a greater distance to the sources in comparison to Europe. Thus, the East Asian air masses show other ageing characteristics than emissions from European MPCs as further analysed in Sect. 4.3.3.

Figure 3 presents an overview on some meteorological parameters with vertical profiles to compare the situation at the two campaign regions. The temperature profile is almost constant in the lowest 2 km around 292 K for EMERGe-Europe and 289 K for EMERGe-Asia while the temperature is decreasing with altitude above 2 km. The virtual potential temperature during EMERGe-Europe shows a stable stratification between 1.75 and 7.5 km which is enclosed by almost neutral layers in the lowest altitudes and above 7.5 km. In contrast, the virtual potential temperature during EMERGe-Asia increases with altitude and is interrupted by more neutral layers between 1.75 and 3.25 km as well as 6 to 8.25 km. The vertical profiles of the RH start with around 80 % at ground for both campaigns and decrease with altitude. During EMERGe-Europe, two RH minima occurred at around 4.75 and 7.25 km as well as two local maxima at around 2.25 and 5.5 km. The latter indicating the variable cloudiness during the summer 2017 at different altitudes. The RH increases again at higher altitudes above 7.5 km for EMERGe-Europe. During EMERGe-Asia, RH minima arise at around 6.5 and 9 km. The RH is stable around 40 % between 2 and 4 km for both campaigns and around 30 % above 10 km for EMERGe-Asia with a slightly increase above 12.5 km. Based on the vertical profiles, an average BL height of about 2 km can be estimated. The wind speed varies with altitude, having lower values of around 6 m s^{-1} in the BL for both campaigns and increasing to around 26 m s^{-1} at 7.75 km during EMERGe-Europe and around 42 m s^{-1} at 12 km during EMERGe-Asia.

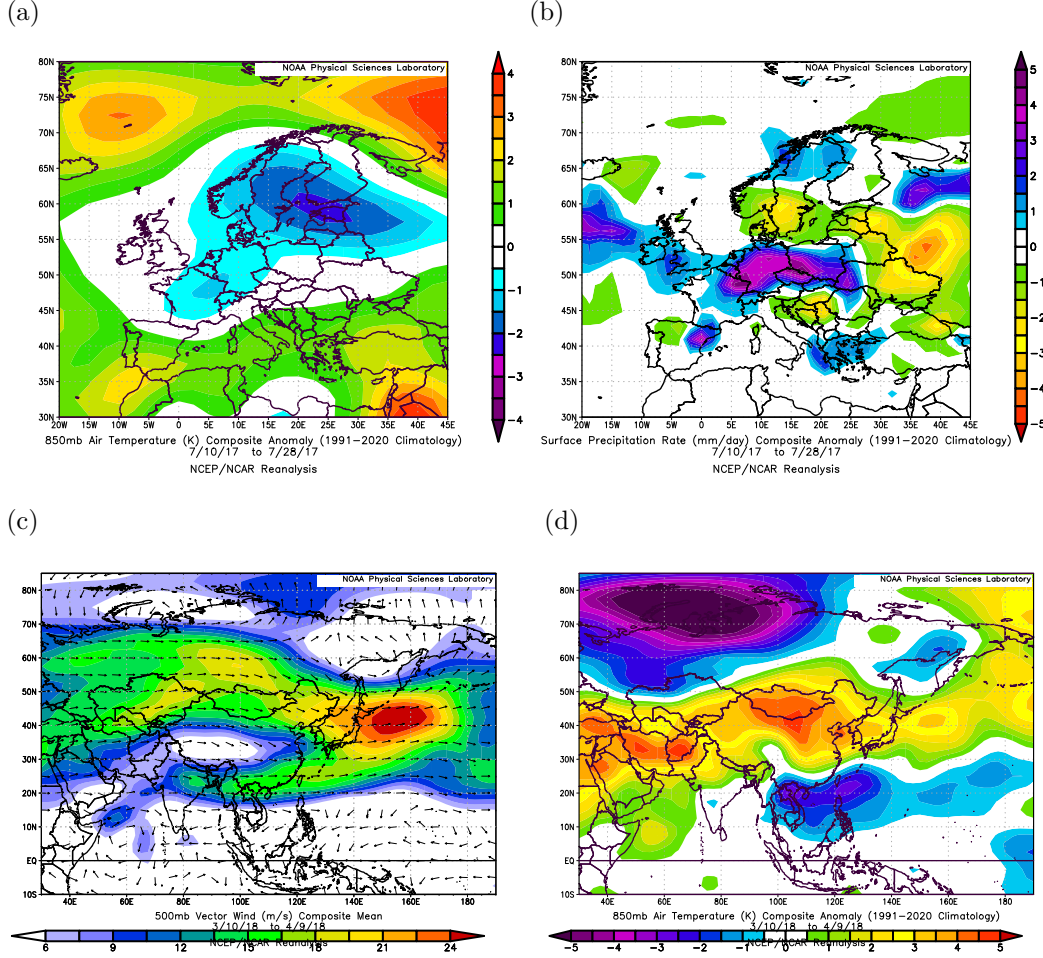


Figure 2: Mean anomaly of the 850 hPa temperature (a) and surface precipitation rate (b) for 10 to 28 July 2017. 500 hPa vector wind (c) and mean anomaly of the 850 hPa temperature (d) for 10 March to 09 April 2018. Anomalies are calculated with respect to a 1991–2020 climatology of the same time periods based on NCEP reanalysis data (Kalnay et al., 1996). Please note the reversed colour code for the precipitation rate anomaly. Images of the NCEP-NCAR Reanalysis 1 data (Kalnay et al., 1996) are provided by the NOAA Physical Sciences Laboratory, Boulder Colorado from their website (<https://psl.noaa.gov/>; last access: 12 September 2023).

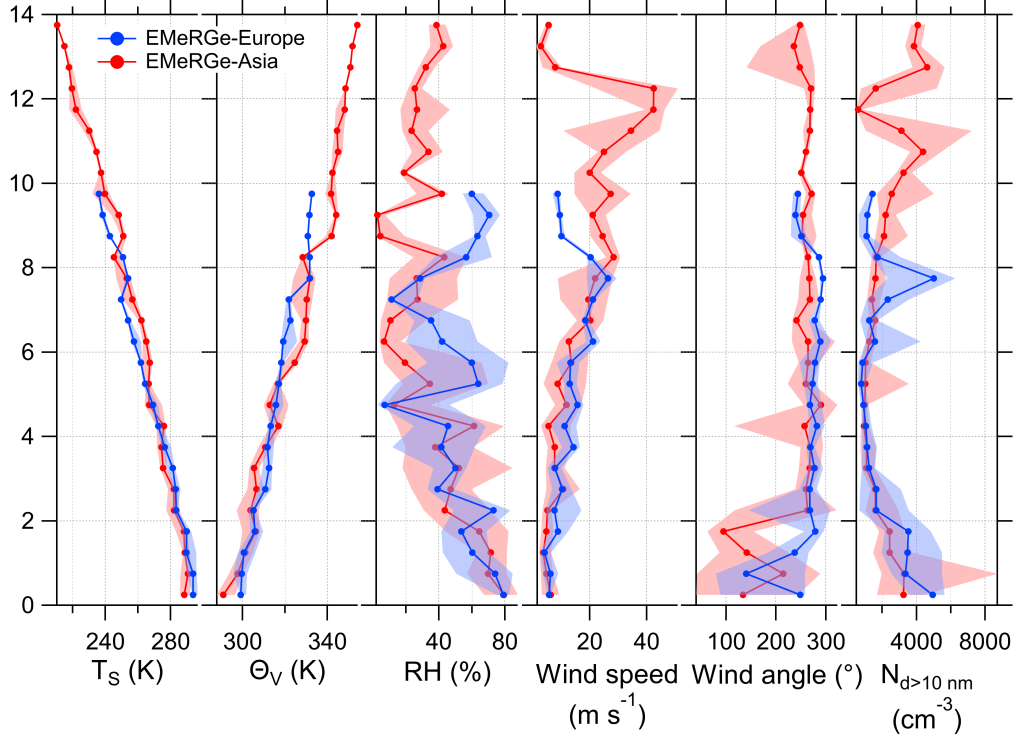


Figure 3: Vertical profiles of the static temperature (T_S), virtual potential temperature (θ_v), relative humidity (RH) with respect to water, horizontal wind speed, wind angle, and aerosol number concentration for particle diameters larger than 10 nm ($N_{d>10 \text{ nm}}$), considering all flights for the European part and flights E-AS-03 to E-AS-14 for the Asian part of the EMeRGe project. Medians (connected dots) with interquartile ranges (shaded area) are shown for each plot in 500 m altitude bins. The meteorological parameters were taken from BAHAMAS and the aerosol number concentration from the AMETYST.



Figure 4: Picture of the German research aircraft HALO and the aerosol inlet HASI mounted on top of the fuselage.

The wind direction is fairly constant above the BL but changes significantly within the lowest 2 km. The vertical profiles of aerosol particles larger than 10 nm decrease with altitude within the BL for both regions. In Europe, the number of this smaller particles stayed low above the BL except for two peaks at around 6.25 and 7.75 km possibly initiated by convective outflow as stated by Andrés Hernández et al. (2022). In Asia, an increase with altitude of these smaller particles started above 6 km and is interrupted by a steep decrease at 11.75 km. This minimum is caused by the absence of a pollution layer and fewer newly formed particles, as this altitude bin is quickly crossed during the transfer sections on the way to the MPCs Osaka and Tokyo.

2.3 MEASUREMENT PLATFORM HALO

The German research aircraft HALO is a modified business jet, type Gulfstream G550 (see Fig. 4). It is operated by the Flight Experiments facility of the German Aerospace Center (DLR-FX) and has its home base in Oberpfaffenhofen, Germany. As the name implies, the main advantages of the aircraft are its exceptionally long range of up to 12 500 km (or more than 10 flight hours) and a maximum flight altitude up to 15.5 km can be reached (Krautstrunk and Giez, 2012). However, the flight altitudes during EMeRGe primarily were below 3 km.

The scientific payload on board HALO was a set of state-of-the-art instruments for the measurement of trace gases and aerosol particles, as described in the following Sect. 2.3.1 and in detail in Andrés Hernández et al. (2022) for EMeRGe-Europe in 2017.

2.3.1 *Instrumentation*

Besides the trace gas and radiation measuring instruments, several aerosol instruments were installed inside the aircraft HALO during EMERGE to measure the aerosol properties such as number concentration and chemical composition: the Aerosol MEasurement sYSTEM (AMETYST), the CCN rack together with the Single Particle Soot Photometer (SP2), and the compact time-of-flight aerosol mass spectrometer (C-ToF-AMS) including an optical particle counter (OPC). A detailed description of all the instruments, measured species and parameters, as well as further references are given in Table 12 in the appendix. The complementary data used for the analysis in this thesis is further discussed in Sect. 3.5.

2.3.2 *HALO Aerosol submicrometer inlet*

The aerosol instruments were connected to the HALO Aerosol Submicrometer Inlet (HASI), a special inlet system for the sampling of ambient aerosol particles under cloud-free conditions. The inlet is mounted on top of the fuselage (see the enlargement in Fig. 4) but outside of the aircraft boundary layer. By regulating the airflow in the sample lines according to the aircraft speed, near-isokinetic sampling conditions can be achieved. The upper cut-off diameter is 2 to 3 μm (Wendisch et al., 2016; Minikin et al., 2017; Andreae et al., 2018).

2.3.3 *Particle loss calculation*

Particle loss is a crucial factor for conducting aerosol measurements. The transport of aerosol particles from outside of the aircraft to the instrument requires two components: a sampling inlet and a tubing system. On both parts particle losses can occur influencing the aerosol characteristics and reducing the sampling efficiency. Therefore, the overall efficiency combines the sampling and the transport efficiency. Mechanisms for particles loss are diffusion, sedimentation or inertial deposition. The effect of each mechanism depends on the particle size. Small particles are most affected by diffusion while larger particles sediment and impact (inertial deposition).

The transmission efficiency calculation was performed with the particle loss calculator after von der Weiden et al. (2009). Version 2.0 of this tool, developed at the Max Planck Institute for Chemistry (MPIC), now also considers the ambient temperature and pressure. The calculations are not applicable to shrouded inlets like HASI. Therefore, only the sampling line inside the aircraft was considered, neglecting possible losses at the HASI inlet itself. An overview of the used parameters is given in Table 13 in the appendix. The total stainless steel tubing line between the HASI and the C-ToF-AMS inlet

Table 4: Static pressure and temperature values which are taken from BAHAMAS data at ground level and maximum altitude for both campaigns.

	LOWER LEVEL		MAX. ALTITUDE	
	p_s (hPa)	T_s ($^{\circ}\text{C}$)	p_s (hPa)	T_s ($^{\circ}\text{C}$)
E-EU	953	27	301	-37
E-AS	1015	25	162	-63

measured 3.62 m and is divided into 16 separate sections depending on their inclination and bending angles. Further, the flow varies inside the different sections depending on the instruments connected to it. Besides the last section 16, where only the C-ToF-AMS is responsible for a flow of 0.1 L min^{-1} , all the other sections are connected to both the C-ToF-AMS and the OPC (with a flow of 1.2 L min^{-1}) and for the first four sections also the bypass flow back to the AMETYST has to be considered. The bypass flow was used to keep the sampling conditions isokinetic and is correlated with the true air speed of the aircraft which is proportional to the flight altitude. For both campaigns, a minimum and maximum bypass flow value was taken at ground level and the highest flight altitude and added to the flow of the other two instruments for the first four sections (see Table 14 in the appendix). The effect of this minimum and maximum bypass flow is negligible in comparison to the greater influence of pressure. Further, a shape factor of 1 (assuming spherical particles) was used for the calculation as well as an aerosol density of 1.2 g cm^{-3} (Cross et al., 2007; Nakao et al., 2013) which corresponds to OA, a major aerosol component globally. Laminar flow was achieved in all tube sections. For both campaigns, a minimum and maximum value of pressure and temperature were taken from the BAsic HALO Measurement And sensor System (BAHAMAS) data at ground level and the highest flight altitude (listed in Table 4) to get an impression of the range of the transmission efficiency.

The results of the particle loss calculator are displayed in Fig. 5. The same tubing line was used for both campaigns. Thus, the transmission efficiency curves are mainly influenced by the different flight altitudes. The curves at ground level are more or less similar for the two campaigns and show a transmission efficiency greater than 90 % for particles with a diameter between 50 and 2000 nm. The transmission is reduced for smaller and larger particles and the cut-off diameter at 50 % efficiency (d_{50}) is around 10 nm. The particle losses for small and large particles are slightly higher with increasing altitude but still an efficiency above 90 % is reached at the maximum flight altitudes for particle sizes between 65 and 1770 nm for EMeRGe-Europe and between 78 and 1500 nm for EMeRGe-Asia. The d_{50} is 14.5 nm at 301 hPa

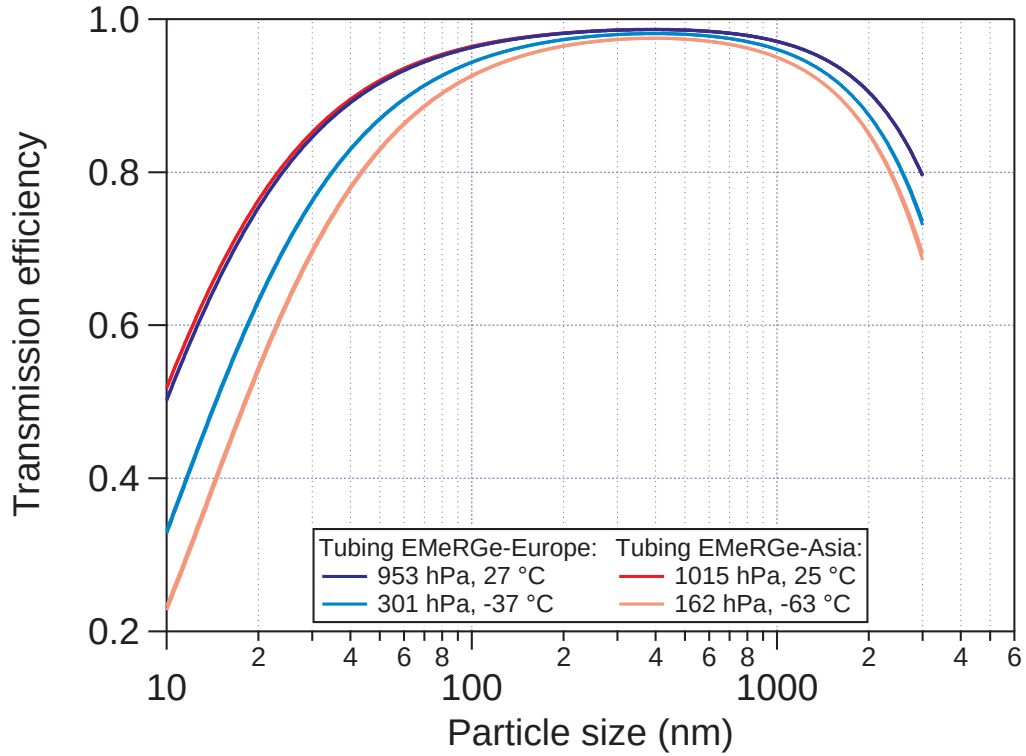


Figure 5: Transmission efficiencies for different particle sizes for different altitudes of both campaigns.

and 18 nm at 162 hPa. In the measurement range of the C-ToF-AMS (40 to 800 nm), the transmission efficiency in all altitudes is greater than 80 %.

2.4 FLIGHT PLANNING

For the measurement campaign EMeRGe, it was important to know exactly where to find the emission plumes of the target areas geographically and at which altitude before handing in the flight plan for approval. During one flight, many different airspaces were crossed, meaning several air traffic controls being in charge and announcing their restrictions on blocked airspaces or busy flight routes (e.g. flight restrictions in the Chinese airspace made it necessary to fly mainly above sea). Further, it was not possible to change the flight pattern spontaneously during the flight, so the route had to be planned early in advance considering all the requests. Flying with HALO at an average speed of 115 m s^{-1} for a flight altitude around 1.5 km implies having one data point every 3.45 km for an instrument time resolution of 30 s. So missing the exact position of the emissions could reduce the number of valuable data points significantly. Other factors influencing the flight planning are to avoid clouds, to consider restrictions on climb and sink rates, and to find a compromise on the different sampling strategies for in situ and

remote sensing instruments. In conclusion, a precise flight planning is needed to address all the measurement goals sufficiently.

The flight planning during the EMeRGe project was supported by forward trajectories using Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT) (Stein et al., 2015; HYSPLIT, 2022) combined with CO emission data (calculations conducted by the DLR Institute of Atmospheric Physics (DLR-IPA)) and chemical forecasts for CO and O₃ provided by the Copernicus Atmosphere Monitoring Service (CAMS) (Flemming et al., 2019; CAMS, 2022). The dispersion of CO was used because it is a long-lived tracer for anthropogenic pollution (1 - 3 months; Seinfeld and Pandis, 2016). O₃ was considered as a tracer for stratospheric intrusions, often coinciding with long-range transport. Additionally, for some specific flights, perfluorocarbon (PFC) tracers (principal investigator: DLR-IPA) were released at distinct locations on the ground to follow the path of the emissions. Those PFCs are chemically and biologically inert, show no interaction with aerosols and clouds, and are detectable at concentrations of a few ppq because their natural background concentrations are very low. The weather forecasts were taken from the European Centre for Medium-Range Weather Forecast (ECMWF) (ECMWF, 2022) and the NCEP (NCEP, 2022).

A typical flight pattern includes transfer sections at high altitudes to reach the area of interest quickly and to save time to measure the urban emission plumes within the atmospheric BL or in the lower troposphere, and extended flight segments close to MPCs. One way to capture their emissions is to perform vertical "shuttles" downwind of the anthropogenic source regions, i.e. repeating flight transects at two or three different altitudes stacked at the same location.

A total of 53 flight hours were carried out during EMeRGe-Europe and around 111 flight hours during EMeRGe-Asia (flights E-AS-03 to -14). Although the flight planning tried to avoid clouds, 2.52 % and 3.42 % of the time could have been influenced by clouds during EMeRGe-Europe and -Asia, respectively, based on the RH data greater than 98 % (see Fig. 59 in the appendix; Ström et al., 2003; Korolev and Isaac, 2006). Based on HYSPLIT simulations for the selected target regions, most of the MPC emissions were located below 3 km and consequently between 60 % and 67 % of the measurements were performed in this altitude range (see Fig. 6; Andrés Hernández et al., 2022; Förster et al., 2023).

The flights are named after Andrés Hernández et al. (2022) as E-EU-FN or E-AS-FN, where E stands for the project EMeRGe, EU or AS indicate the campaign region Europe or Asia, and FN are the two digits of the flight number.

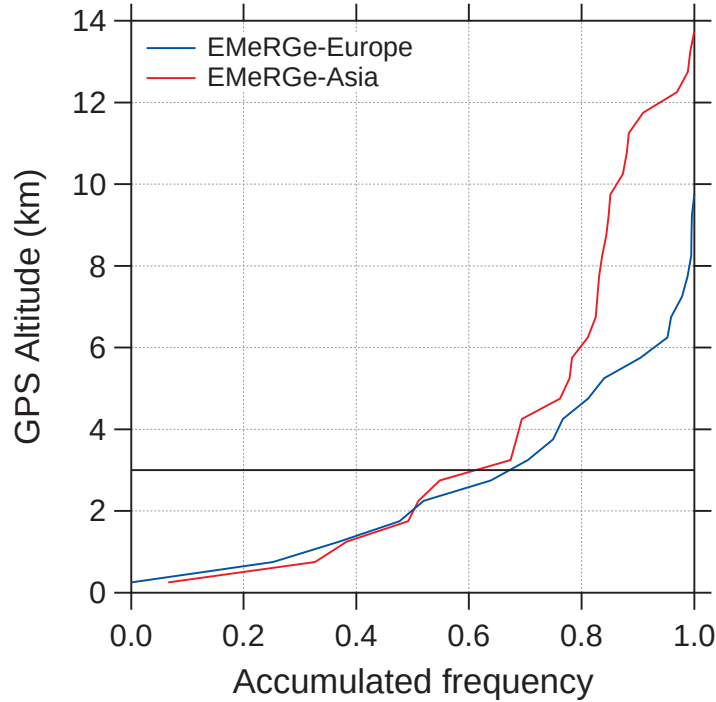


Figure 6: Accumulated frequency of flight altitudes during EMeRGe in 500 m bins, considering all flights for the European part and flights E-AS-03 to E-AS-14 for the Asian part of the project.

For EMeRGe-Europe three main flight regions were investigated considering the flight restrictions (described in detail in Andrés Hernández et al. (2022)):

- *Southern Europe and Italy*
Flights: E-EU-03, E-EU-06
Targets: Po Valley, Rome
Additional measurements: BB from Southern Italy and Croatia, dust from Northern Africa
- *London and Central Europe*
Flights: E-EU-05, E-EU-08
Targets: London, BeNeRu, Paris
Special flight E-EU-04: blind intercomparison with another research aircraft (see Sect. 3.4), transcontinental transport of Canadian fire emissions
- *South-Western Europe*
Flights: E-EU-07, E-EU-09
Targets: Spain, Southern France, Po Valley
Additional measurements: BB from Southern France, Corsica, and Portugal, dust from Northern Africa

For EMeRGe-Asia four main flight regions were investigated considering the flight restrictions:

- *Manila and Pearl River Delta*
Flights: E-AS-03, E-AS-06, E-AS-10
- *Japan*
Flights: E-AS-11, E-AS-13
- *East China Sea*
Flights: E-AS-04, E-AS-05, E-AS-07, E-AS-08, E-AS-09
Targets: also outflow from South Korea and the YRD
- *Taiwan*
Flights: especially E-AS-12, E-AS-14, and all other flights during take-off and landing

METHODS

The main focus of the thesis is the characterisation of aerosol particles in the outflow of MPCs. Therefore, the C-ToF-AMS was deployed in the HALO aircraft. In the following sections, the main working principle is described briefly and the data evaluation is presented including calibrations, applied corrections, and the detection limit calculation. Furthermore, complementary measurements of additional aerosol instrumentation and different trace gases as well as the atmospheric transport models are introduced. In addition, the results of a instrument comparison flight are described. In the data analysis section, different tracer ions used for the identification of aerosol types are presented as well as a VOC-based separation of air mass origin and age. The chapter is accomplished by a section on aerosol acidity and the conversion ratio of sulfur.

3.1 COMPACT TIME-OF-FLIGHT AEROSOL MASS SPECTROMETER

Section 3.1.1 summarises the measurement principle of the C-ToF-AMS and its inlet system. The instrument was integrated in HALO during both EMeRGe campaigns. The measured aerosol mass concentration and composition are the basis for the data evaluation in Ch. 4. The performance and data coverage of the C-ToF-AMS during EMeRGe is described in Sect. 3.1.2.

3.1.1 *Measurement principle*

The chemical composition of non-refractory submicron aerosol particles was measured with a C-ToF-AMS as depicted in Fig. 7 and explained in detail e.g. in Drewnick et al. (2005), DeCarlo et al. (2006), Canagaratna et al. (2007), and Schulz et al. (2018). A brief description is given below.

Ambient aerosol particles of up to 2 - 3 μm in diameter were sampled with near-isokinetic flow through the HASI (Minikin et al., 2017) that was mounted on top of the fuselage of HALO. It is connected to the aerosol instruments with stainless steel tubing and conductive silicone tubing is used for the bypass line. After passing the HASI, the aerosol particles enter the C-ToF-AMS via a constant pressure inlet (CPI), which maintains a constant mass flow of $1.3 \text{ cm}^3 \text{ s}^{-1}$ into the instrument (Molleker et al., 2020). The CPI is followed by an aerodynamic lens operated at 2.5 hPa (1.9 Torr) focusing the particles into a narrow beam passing the chopper (Liu et al., 1995a,b). The several differential pumping stages maintain a pressure of less

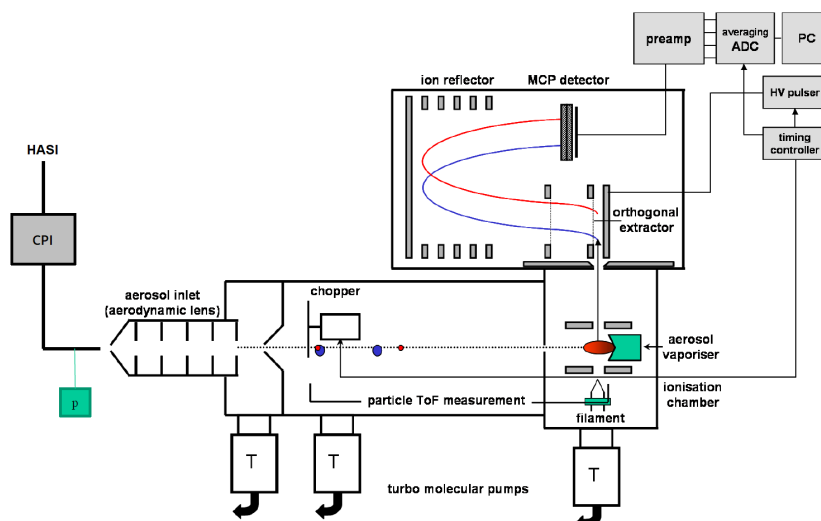


Figure 7: Scheme of the C-ToF-AMS, adapted from Drewnick et al. (2005)¹. Abbreviations: HASI is the near-isokinetic aircraft inlet, CPI the constant pressure inlet of the instrument, p the pre-pump, T the turbo pumps, MCP stands for micro-channel plate, ADC for analog-to-digital conversion data acquisition card, PC for personal computer, and HV for high voltage. Only three of the five turbo molecular pumps T are drawn in the scheme. One of the not depicted turbo pumps is located in the vacuum chamber and the other one at the mass spectrometer.

than 3×10^{-6} hPa in the ionisation region. In the vacuum chamber, the non-refractory species are flash vaporised on an inverted cone porous tungsten vaporiser, operated at around 600 °C, and the generated gas molecules are ionised by 70 eV electron impact ionisation. The ions are extracted, separated by their mass to charge ratio (m/z), and collected by a micro-channel plate (MCP) detector of the time-of-flight mass spectrometer.

The spectrometer gives quantitative mass concentrations of particulate OA, NH₄, NO₃, SO₄, and chloride (in unit mass resolution) with an overall accuracy of about 30 % (Canagaratna et al., 2007; Middlebrook et al., 2012). The charge symbols are not utilised for the nominally inorganic species, as organic compounds could have (mostly small) contributions to these species, e.g. organonitrates and organosulfates (Farmer et al., 2010). The mass concentrations of the C-ToF-AMS are differential signals calculated by subtracting the background signal in vacuum from the signal of the total incoming air including aerosol particles. The separation is accomplished by a metal chopper plate that is frequently moved in and out of the particle beam in the vacuum chamber by a servo motor.

The C-ToF-AMS was operated with a time resolution of 30 s for most of the flights. During EMERGE-Asia, the time resolution could be increased to

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15 s for flights E-AS-08 to E-AS-14 due to higher mass concentrations on site. The time stamps are placed at the end of the intervals averaging over the selected time.

Due to the CPI, the data represent standard temperature and pressure (STP) conditions (273.15 K, 1013.25 hPa) independent of the flight altitude. The transmission of the CPI is known to be reduced at lower altitudes due to the harder squeezing of the O-ring (Schulz, 2019; Molleker et al., 2020; Mei et al., 2020). This was also observed during the instrument comparison flight E-EU-04 on 13 July 2017 (see Sect. 3.4; Schumann, 2021).

The transmission efficiency through the sampling line was determined with the particle loss calculator (von der Weiden et al., 2009) and yield values greater 80 % for the highest flight altitudes and almost 90 % for ground conditions inside the C-ToF-AMS measurement range (40 - 800 nm) as further described in Sect. 2.3.3.

The C-ToF-AMS has not been pumped continuously since the aircraft had power only 4.5 h prior to take-off on a flight day. This leads to increased background signals, especially influencing the organic signal in the beginning of some flights. The background decreased with progressing pumping time to a usual level and is considered in the detection limit (DL) calculation (see Sect. 3.2.3).

The C-ToF-AMS data were analysed using the software package Squirrel v1.60 within Igor Pro 6 (WaveMetrics) (DeCarlo et al., 2006; Sueper and collaborators, 2022).

3.1.2 Performance during EMeRGe

The performance of the C-ToF-AMS during both EMeRGe campaigns is summarised in Tables 18 and 19 in the appendix. As described later in Sect. 3.2.1, filter measurements are needed to correct for fragmentation patterns from the gas phase. Thus, at least one filter measurement was performed for each flight ranging between 9 and 20 min for EMeRGe-Europe, and between 10 and 32 min for EMeRGe-Asia. During flight E-EU-07, problems with the data saving occurred and accumulated to data gaps of 117.5 min. For the last transfer flight E-AS-16, the measurements could only be started with a delay of 113 min due to reduced preparation time before take-off, resulting in a coverage of around 74.5 % for those two flights. In total, the data coverage is 93.3 % and 94.8 % for EMeRGe-Europe and EMeRGe-Asia, respectively.

3.2 DATA EVALUATION METHODS

The following section outlines the calibration procedures applied to the C-ToF-AMS data, describes an additional background correction, and explains the determination of the DLs. A detailed description of the calibra-

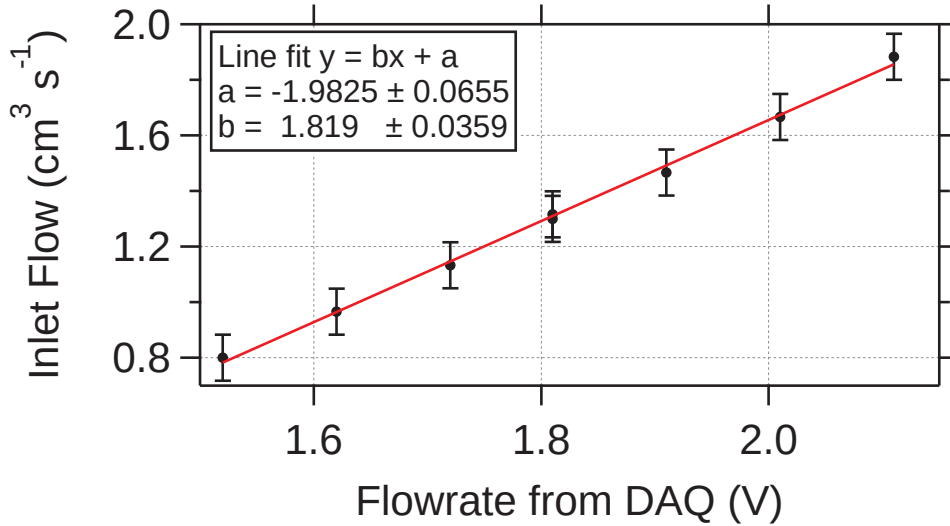


Figure 8: Flow calibration during the EMeRGe-Asia campaign listing the linear fit coefficients. Exact values are given in Table 15 in the appendix.

tions can be found in e.g. Jayne et al. (2000), Allan et al. (2003), Hings (2007), and Kimmel (2011).

3.2.1 Calibrations

Lens adjustment

After installing the C-ToF-AMS in HALO, the aerodynamic lens was adjusted to focus the particle beam in the optimal position. Therefore, ammonium nitrate (NH_4NO_3) particles were generated with an mobile atomiser and dried with a diffusion dryer. The lens was moved slightly and successively in the x and y direction to find the maximum in the NO_3 concentration.

Flow calibration

The inlet flow was calibrated on the ground under ambient pressure conditions once during EMeRGe-Asia using a TSI flowmeter. The calibration was retrospectively applied to the European data set. Figure 8 shows the fit curve and used fit values that were entered into the data acquisition (DAQ) software of the C-ToF-AMS.

Relative ionisation efficiency (RIE) calibration

The C-ToF-AMS was calibrated with monodisperse NH_4NO_3 and ammonium sulfate ($(\text{NH}_4)_2\text{SO}_4$) particles several times during the campaigns to determine the *RIE* for NO_3 and SO_4 . The results are given in Table 5.

Table 5: Relative ionisation efficiencies (*RIEs*) used for the data analysis. The *RIEs* for ammonium and sulfate were determined from calibrations. The other *RIEs* are literature values (Alfarra et al., 2004; Takegawa et al., 2005; Canagaratna et al., 2007).

	NO ₃	NH ₄	SO ₄	OA	CHLORIDE
EMeRGe-Europe	1.1	4.05	1.28	1.4	1.3
EMeRGe-Asia	1.1	3.76	1.25	1.4	1.3

Ionisation efficiency (IE) calibration

The *IE* was obtained in the brute force single particle (BFSP) mode with size-selected NH₄NO₃ particles and is defined as the ratio of detected ions per molecule introduced into the ionisation region. The measurements were performed four times for EMeRGe-Europe and five times for EMeRGe-Asia with particles of 300 and 350 nm (particle mobility diameter (d_{mob})) resulting in campaign averages of 7.62×10^{-7} and 9.56×10^{-7} for EMeRGe-Europe and -Asia, respectively. Each flight refers to the *IE* calibration taken shortly before the flight and uses the mean of both sizes (see Tables 16 and 17 in the appendix). Furthermore, an air beam signal (*AB*) correction is performed to account for changes of the *IE*, detector gain, and inlet flow.

Particle size calibration

The particle vacuum aerodynamic diameter (d_{va}) measurement was calibrated with NH₄NO₃ particles size-selected with a differential mobility analyser (DMA) and polystyrene latex (PSL) spheres ranging from 100 to 800 nm (geometric diameter). The results for both campaigns differ only slightly and are depicted in Fig. 9.

Collection efficiency (CE)

The *CE* was set to 0.5 in the analysis software after Matthew et al. (2008) and Middlebrook et al. (2012). It is the default value valid for measurements of ambient aerosol particles.

Fragmentation table adjustment

Pure gas-phase measurements were realised during each flight by switching the inlet line to pass a particle filter. They were used to diminish interference between gas and non-refractory particle phase materials within the mass spectra by adjusting the fragmentation table (Allan et al., 2004).

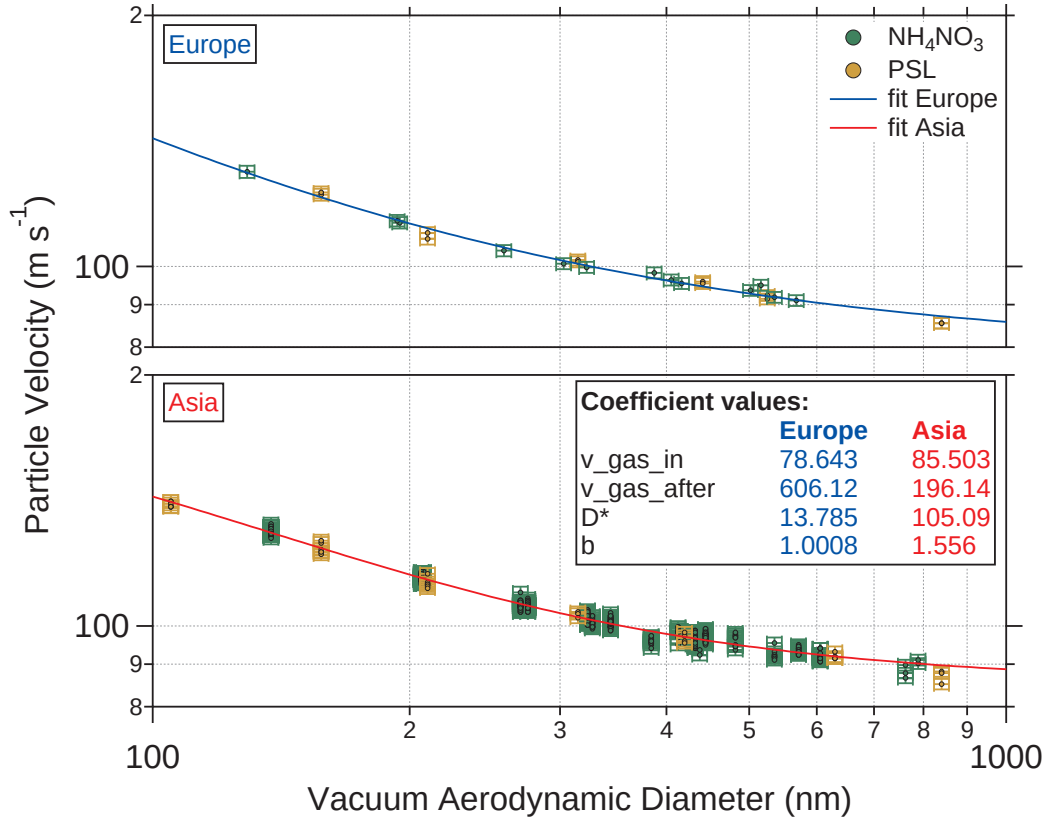


Figure 9: Results of the size calibrations using NH_4NO_3 and PSL particles for EMeRGe-Europe (top) and -Asia (bottom). The used fit function is: $f(d_{\text{aero}}) = v_{\text{gas_in}} + (v_{\text{gas_after}} - v_{\text{gas_in}}) / (1 + (d_{\text{aero}} / D^*)^b)$ and was performed for a combination of the two substances.

Single ion (SI) calibration

Before every flight and calibration, the *SI* value was taken within the data acquisition software (ToF-AMS DAQ, Kimmel (2011)) and is needed to convert the signal intensity to units of mass concentration.

Software

The data analysis was performed with Igor Pro 6.37 (Wave Metrics) and the software Squirrel 1.60B including the performance of the m/z calibration and baseline correction.

3.2.2 *Background correction*

After performing all the calibrations and corrections as described in the previous Sect. 3.2.1, there was still a small signal of up to $0.24 \mu\text{g m}^{-3}$ remaining during the filter measurement periods. Therefore, an additional background correction was performed by subtracting the average mass concentration of each species during the filter time from the total time series if the remaining signal was greater than $0.001 \mu\text{g m}^{-3}$. This procedure has mainly affected the organic and nitrate signal but also the sulfate and chloride signal of some flights. The applied values are listed in Tables 20 and 21 in the appendix.

3.2.3 *Detection limits*

The DLs for all C-ToF-AMS species were calculated as three times the standard deviation from the background signal (chopper closed) using a time-dependent cubic spline function gaining a DL for each data point (Reitz, 2011). According to Schulz (2019), this method is the most representative calculation of the DLs suitable for aircraft measurements because long-term trends influencing the background signal are excluded and only the short-term noise persists. The DLs averaged over the whole campaign are given in Table 6 and agree with values from DeCarlo et al. (2006) and Schulz (2019).

Figures 10 and 11 display averaged vertical profiles of the C-ToF-AMS species and their corresponding DLs for the EMeRGe-Europe and -Asia campaigns, respectively. The DL values are divided by the square root of the number of data points per bin. Almost all mass concentrations during EMeRGe-Europe exceed the DLs which is also due to the great amount of averaged data points. An exception are the OA and nitrate means in an altitude range between 7.5 and 9 km. The ammonium is close to the DL above 7.5 km. For EMeRGe-Asia, the organics, sulfate, and nitrate are greater than the DL, as well as the ammonium below 6 km. Two DL average profiles are displayed for the EMeRGe-Asia campaign depending on the sampling time

Table 6: Campaign averaged DLs in $\mu\text{g m}^{-3}$ for all C-ToF-AMS species. The time resolution Δt for the EMeRGe-Europe campaign is 30 s. The EMeRGe-Asia parts consider the flights E-AS-03 to E-AS-07 with a time resolution of 30 s and E-AS-08 to E-AS-14 with 15 s, leaving out the transfer flights over Europe.

	Δt	NO_3	NH_4	SO_4	OA	CHLORIDE
Europe	30 s	0.04	0.09	0.03	0.15	0.02
Asia (E-AS-03 to -07)	30 s	0.02	0.07	0.02	0.25	0.01
Asia (E-AS-08 to -14)	15 s	0.03	0.13	0.03	0.46	0.02

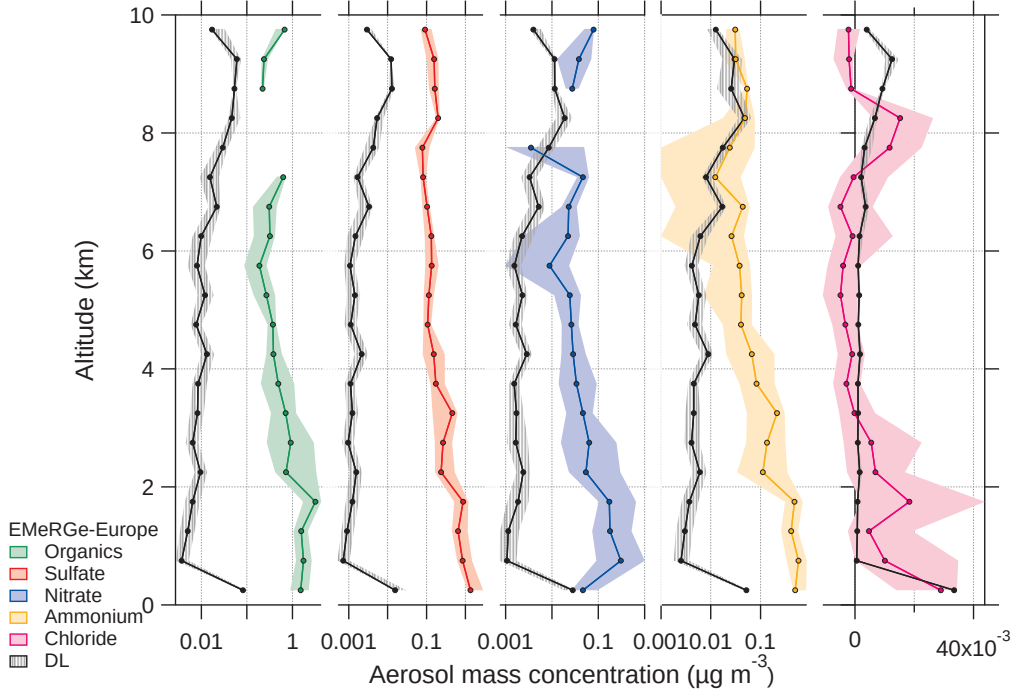


Figure 10: Averaged vertical profiles of the C-ToF-AMS species and their corresponding DLs (divided by the square root of the number of data points per bin) for all EMeRGe-Europe flights in 500 m altitude bins. The dots represent the medians and the shaded areas the interquartile ranges. Note that the axes for the mass concentrations are logarithmic (except for chloride) to emphasise the smaller DL values. Gaps in the profiles are due to negative values that can not be displayed in the logarithmic representation.

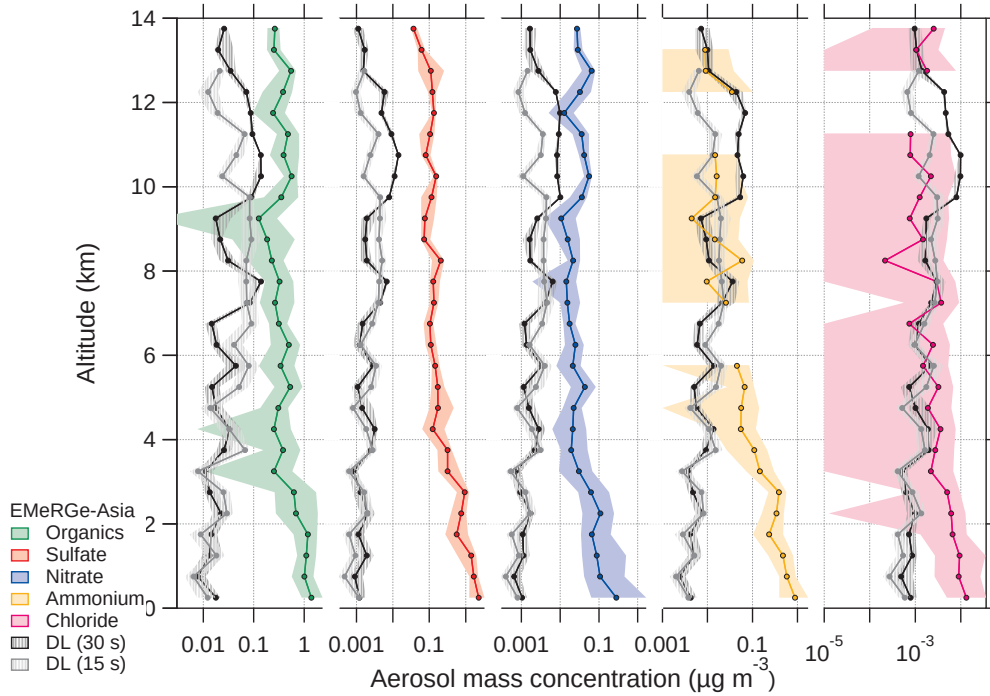


Figure 11: Same as Fig. 10 but for EMeRGe-Asia flights E-AS-03 to E-AS-14. The DL profiles are separated in the flights E-AS-03 to E-AS-07 and E-AS-08 to E-AS-14 considering their different time resolutions of 30 and 15 s, respectively.

that was reduced to 15 s for some flights. The chloride mass is very low for both campaigns and only exceeds the DL below 3 km and around 8 km during EMeRGe-Europe and below 5.5 km for EMeRGe-Asia. Therefore, chloride is not taken into account in the following analysis or just for the lower altitudes and BL measurements. In general, negative mass concentrations can occur as the C-ToF-AMS measurements are differential signals and even the additional background correction can not compensate for every variability. These values are considered to be below the DL.

3.3 OPTICAL PARTICLE COUNTER

Additionally to the aerosol composition of the C-ToF-AMS, the particle number concentration and size distribution were measured with a optical particle counter (OPC) (model 1.129 Sky-OPC, GRIMM, Ainring, Germany) for particles in a size range of 0.25 to 32 μm in 31 size channels. However, the maximum diameter is limited by the upper cut-off of the aerosol inlet at around 2 - 3 μm . The instrument is connected with the C-ToF-AMS tubing and has a time resolution of 6 s (GRIMM, 2008). The sample flow is maintained at a constant volume flow by a critical orifice and an external vacuum pump.

3.3.1 STP correction

A STP correction is applied to the OPC data to make it comparable to the C-ToF-AMS measurements using Eq. 1.

$$dp_{STP} = dp \cdot \frac{T_{ipOPC}}{T_{STP}} \cdot \frac{p_{STP}}{p_{ipOPC}} \quad (1)$$

In this equation dp is the particle number concentration in each size channel per time step, T_{ipOPC} and p_{ipOPC} are values for temperature in K and pressure in hPa taken from internal sensors and interpolated on the OPC time series, and T_{STP} and p_{STP} are the STP values for temperature and pressure (273.15 K and 1013.25 hPa).

The correction is applied to each measurement point and also valid for every quantity derived from the binned number concentrations (e.g. total number concentration or volume of particulate matter with a diameter of 1 μm or less (PM_1) particles).

3.3.2 Aerosol surface, volume, and mass calculations

Based on the original data matrix dp also aerosol surface dS , volume dV , mass dM , and total number concentration dN can be derived by summing up over the different size channels for every time step:

$$dS = \sum_{i=0}^{31} dp[i] \cdot \pi \cdot D_{Mean}^2[i] \quad (2)$$

$$dV = \sum_{i=0}^{31} dp[i] \cdot \frac{\pi}{6} \cdot D_{Mean}^3[i] \quad (3)$$

$$dM = dV \cdot \rho \quad (4)$$

$$dN = \sum_{i=0}^{31} dp[i] \quad (5)$$

Depending on the upper size limit, these calculations differentiate between PM_1 , $PM_{2.5}$, particulate matter with a diameter of 10 μm or less (PM_{10}), or the total measurable range of the OPC. D_{Mean} is the mean diameter of each size channel and ρ the assumed density depending on the aerosol type (e.g. 2.5 g cm^{-3} for dust or 1.5 g cm^{-3} for submicron non-refractory aerosol particles; Pitz et al., 2003; Khlystov et al., 2004; Wagner et al., 2009; Froyd et al., 2019).

3.4 QUALITY ASSURANCE

Quality assurance is an important part of providing reliable data and ensuring a critical evaluation of the measurements. Therefore, one section of flight



Figure 12: Picture of the FAAM-BAe 146 aircraft flying next to the HALO during the comparison flight E-EU-04 on 13 July 2017.

E-EU-04 was used as a blind comparison with instruments on the UK research aircraft BAe 146 from the Facility for Airborne Atmospheric Measurements (FAAM). The flight was performed over the temporary restricted area (TRA) Allgäu in Southern Germany and the comparison period consisted of a racetrack pattern in three different altitudes (4.7 (flight level (FL)150), 3.2 (FL100), and 1.2 km (FL040)) starting in the dry and cloud-free troposphere stepping down into the polluted convective BL. HALO was flying in front while the FAAM-BAe aircraft was following closely with a mean horizontal (vertical) distance of only 106 m (8 m) (see Fig. 12). The comprehensive results of the comparison flight can be found in Schumann (2021) complemented with model results and ground-based data from the Meteorological Observatory at Hohenpeißenberg which is operated by the Deutscher Wetterdienst (German Meteorological Service).

The focus of this section is the comparison of the two aerosol mass spectrometers, one mounted in each aircraft, both of the type compact time-of-flight. The different aerosol particle species were under investigation as well but for simplicity we proceed here with the total sum of all species. Figure 13 shows the correlation of the total aerosol mass concentration from the FAAM and HALO C-ToF-AMS for the three comparison altitudes. The data differ by a factor of 2.4, 1.8, and 1.9 on FL040, FL100, and FL150, respectively.

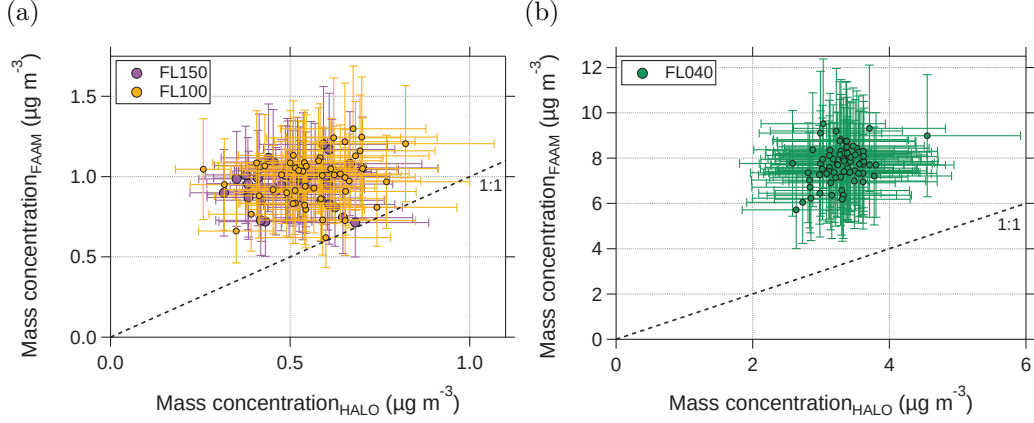


Figure 13: Correlation of the total aerosol mass concentration from the revised FAAM and HALO C-ToF-AMS for the three comparison altitudes: FL100 and FL150 (a), FL040 (b). FAAM data (revised data set) were provided by University Manchester (Schumann, 2021). Error bars indicate the uncertainty range of 30 % (Canagaratna et al., 2007; Middlebrook et al., 2012). During the discussion of the comparison report, a revised data set for the FAAM C-ToF-AMS was released due to issues with the calibration equipment, reducing the original data by a factor of 0.623. The correlation plot using the original FAAM data can be found in Fig. 60 in the appendix for comparison.

To examine these differences in detail, three independent measurements were utilised, namely a Scanning Mobility Particle Sizer (SMPS) mounted on the FAAM-BAe and two OPCs on board HALO. Therefore, their number and volume distributions were combined to one size distribution covering a bigger size range (see Fig. 14). This was possible because the distributions matched quite good in the overlapping size range although being measured on different aircraft (inlet issues between the two aircraft can be excluded as a general cause for the discrepancies). The second OPC (integrated in the AMETYST) was exploited to double check the results of the first OPC (integrated in the HALO C-ToF-AMS). Further, the combined volume distribution was used to calculate an expected aerosol mass concentration $dM_{expected}$ for the C-ToF-AMSs using Eq. 6:

$$dM_{expected} = dV_{SMPS+OPC<0.8\mu m} \cdot \rho - dM_{BC} \quad (6)$$

Thereby, an average aerosol density ρ of 1.5 g cm^{-3} was assumed and only the measurable size range (up to $0.8 \mu\text{m}$) for the C-ToF-AMSs was taken into consideration. In addition, the refractory black carbon (rBC) mass concentration was subtracted (data provided by MPIC Multiphase Chemistry Department within Schumann (2021)) because it is not detectable by the C-ToF-AMSs. The mean values are listed in Table 22 in the appendix.

Figure 15 demonstrates that the C-ToF-AMSs measure reasonable values compared to the calculated expected mass concentration considering

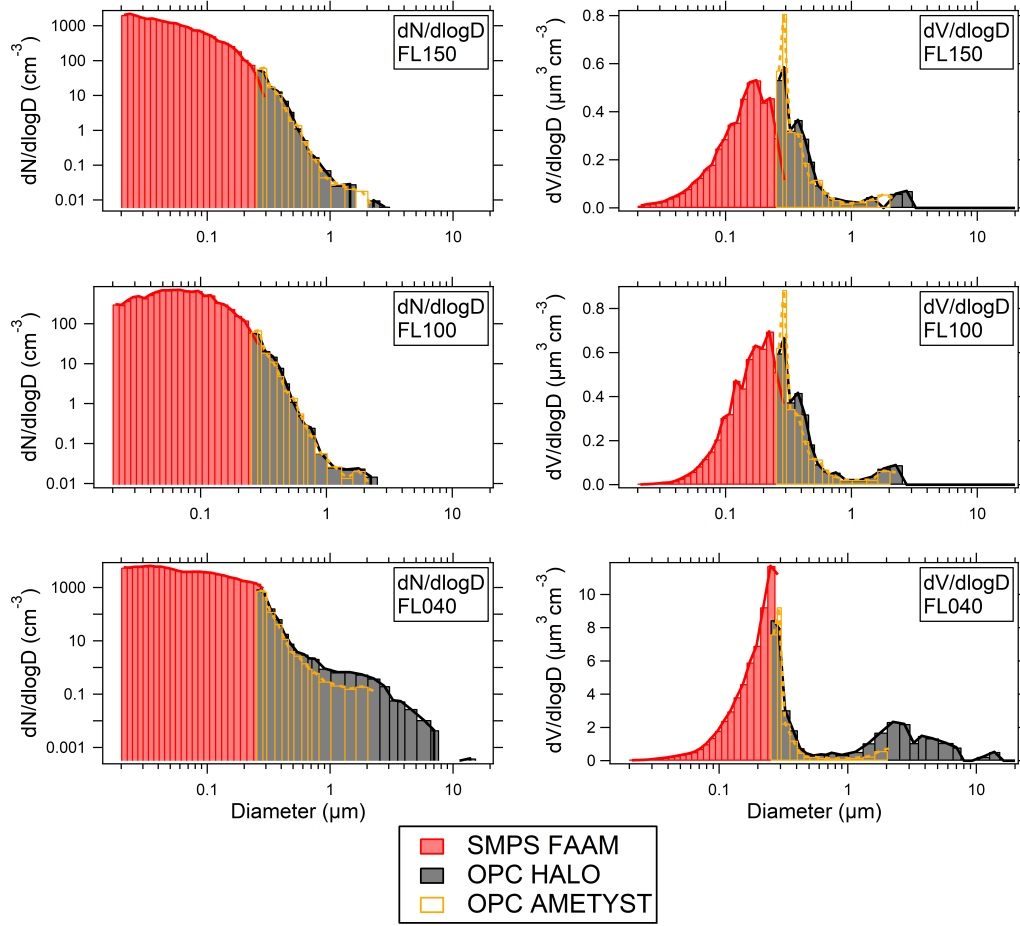


Figure 14: Number and volume size distributions from the FAAM SMPS and the two HALO OPCs for the three comparison altitudes. SMPS data were provided by University Manchester and data of the AMETYST OPC by DLR-IPA (Schumann, 2021).

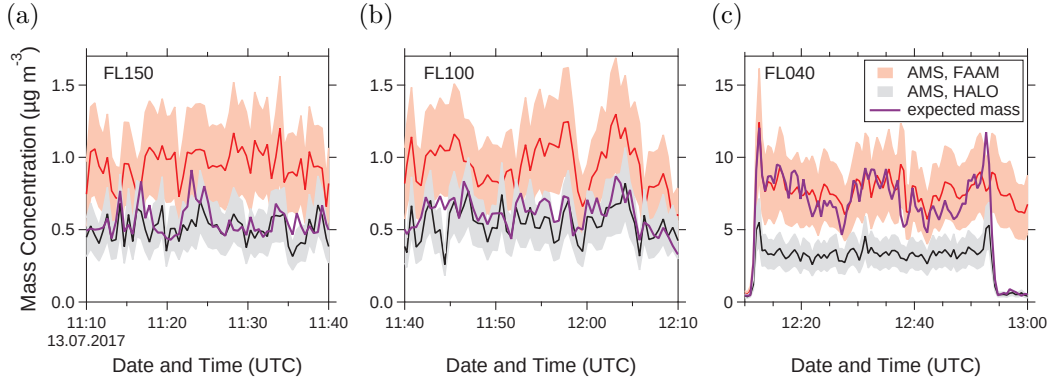


Figure 15: Time series of the revised FAAM and HALO total aerosol mass concentration for the three comparison altitudes including the calculated expected mass concentration. FAAM data (revised data set) were provided by University Manchester (Schumann, 2021). Shaded areas indicate the uncertainty range of 30 % (Canagaratna et al., 2007; Middlebrook et al., 2012). Note the modified y-axis for FL040.

the C-ToF-AMS uncertainty range of 30 % (Canagaratna et al., 2007; Middlebrook et al., 2012). Therefore, we can safely assume that the HALO C-ToF-AMS measured correctly at FL100 and FL150 while the FAAM C-ToF-AMS measured the expected mass concentration at FL040. Nevertheless, a substantial difference between the two instruments is evident. At FL100 and FL150, the FAAM C-ToF-AMS overestimates the expected mass concentration by almost 70 % (see Fig. 15a and b; Table 22 in the appendix). This might be explained by the different aerosol inlet used on the FAAM aircraft which is a Rosemount inlet (Foltescu et al., 1995). It has been shown that these inlets enrich aerosol concentrations, especially in the supermicron size range above a diameter of $0.6 \mu\text{m}$ (Trembath, 2013; Hodgson et al., 2018). Thereby, the enrichment is dependent on the aerosol bulk density, e.g. up to a factor of 10 enhanced aerosol concentrations were observed for Saharan dust. Furthermore, the deviation between the two instruments might be related to differences in the sampling lines or the different inlets of the instruments, especially the different inlet pressure regulation systems. The larger deviation of about 50 % between expected and measured mass concentration by the HALO C-ToF-AMS at the lowest FL040 (see Fig. 15c) can be explained by inlet specific issues of this instrument. The transmission of the constant pressure inlet is reduced below 2.5 km due to the squeezed O-ring that keeps the mass flow constant. This was already observed during other measurement campaigns (Schulz, 2019; Mei et al., 2020) and systematically tested in Molleker et al. (2020). The data have not been corrected for the transmission reduction at lower altitudes because the exact size and height dependency is unclear and the squeezed O-ring shape is irregular and not reproducible.

3.5 COMPLEMENTARY DATA

The following section depicts the complementary data (meteorological parameters, other aerosol measurements, trace gas data, air mass history modelling) and their basic working principles used for the data evaluation of the EMeRGe campaign in Ch. 4 supporting the aerosol particle composition measurements of the C-ToF-AMS. Table 7 gives an overview of the used parameters from the additional instruments. Further details can be found in Sect. 2.3.1, Andrés Hernández et al. (2022), and the references listed in the text. The modelling tools HYSPLIT and flexible trajectory model (FLEXTRA) as well as a method using emission inventories to identify source regions (described in Förster et al. (2023)) are presented helping to gain a comprehensive analysis of the MPC emissions and their air mass history.

3.5.1 *Basic meteorological and aircraft data*

Basic meteorological and aircraft data were taken from the BAHAMAS, operated and maintained by DLR Flight Experiments (DLR-FX). The data set includes e.g. basic aircraft position data (Global Positioning System (GPS) altitude, latitude, and longitude), 3-D wind field, pressure, and temperature (Krautstrunk and Giez, 2012). Typical accuracy values of the data are 0.22-0.39 K for temperature (increasing with altitude), 0.15 hPa for pressure, and 0.14-0.42 m s⁻¹ for the horizontal wind speed (increasing with altitude) (Giez et al., 2023). The RH was measured by the Sophisticated Hygrometer for Atmospheric ResearCh (SHARC) based on direct absorption measurement by a tunable diode laser system (Krautstrunk and Giez, 2012). The humidity measurements have an accuracy of 5 % but at least 1 ppmv (Kaufmann et al., 2018; Giez et al., 2023).

3.5.2 *Other aerosol measurements*

Particle number concentration and size distribution

Particle number concentrations and size distributions for aerosol particles greater 10 nm and 250 nm were measured by a condensation particle counter (CPC) and an OPC, respectively, both integrated in the AMETYST and operated by DLR-IPA. The OPC data was used to validate the data of the OPC attached to the C-ToF-AMS in Sect. 3.4.

Table 7: Overview of complementary data from different instruments used in this thesis, together with their units, time resolution, measurement range, and accuracy/precision. Further references and data operators are listed in Table 12.

INSTRUMENT	USED PARAMETER (s)	UNIT	MEAN TIME RESOLUTION	ACCURACY (A), PRECISION (P)
AENEAS	NO	ppbv	1 s	A: 7 %, P: 5 %
	NO _y	ppbv	1 s	A: 15 %, P: 10 %
AMETYST*	particle number	cm ⁻³	1 s	
AMTEX	CO	ppbv	1 s	A: 2.4 %, P: 1.5 ppbv
	O ₃	ppbv	1 s	A: 10 %, P: 6 ppbv
BAHAMAS	static <i>p</i> and <i>T</i>	hPa and K	1 s	A: 0.3 hPa and 0.5 K
	Latitude, Longitude	°	1 s	A: 4.75×10^{-5} °
	GPS Altitude	m	1 s	
	θ_v and RH	K and %	1 s	RH: 15 %
	wind speed, angle	m s ⁻¹ and °	1 s	A: 0.5 m s ⁻¹
CATS	SO ₂	pptv	2 s	7 - 13 % at high VMR, 25 - 31 % close to DL
CCN*	CCN number	cm ⁻³	1 s	6 - 10 %
HKMS	Acetonitrile, Benzene, Isoprene, Toluene	pptv	60 s, 1/61 Hz per VOC	A: 8 - 9 %
SP2*	rBC mass	µg m ⁻³	1 s	A: 10 %

* The upper particle size cut-off is approximately 3 µm owing to the particle transmission efficiency of the main aerosol inlet (Sect. 2.3.2), although some instruments might have a higher upper cut-off.
Data by the AMETYST, CCN, and SP2 are presented at STP 273.15 K and 1013.25 hPa.

Black carbon

Mass concentrations of rBC were measured with an eightchannel SP2 (manufactured by Droplet Measurement Techniques, Longmont, CO, USA) (Stephens et al., 2003; Schwarz et al., 2006; Holanda et al., 2020), operated by the MPIC-Multiphase Chemistry Department. The rBC mass was added to the C-ToF-AMS species to obtain the total aerosol mass for PM_1 . The accuracy of the SP2 rBC mass concentration is 10 % (Laborde et al., 2012).

Cloud condensation nuclei

The number concentration of CCN was measured with a two-column CCN counter (CCNC) (model CCN-200, manufactured by Droplet Measurement Techniques, Longmont, CO, USA) (Roberts and Nenes, 2005; Rose et al., 2008; Krüger et al., 2014), operated by the MPIC-Multiphase Chemistry Department. The CCNC also sampled from the HASI and measured at a constant supersaturation of 0.38 ± 0.05 % with a time resolution of 1 Hz. The error is around 6 % to 10 % (Pöhlker et al., 2016).

3.5.3 *Trace gas data**NO and NO_y*

NO and total reactive nitrogen (NO_y) were measured with Atmospheric Nitrogen oxides mEAsuring System (AENEAS) operated by DLR-IPA using a dual-channel chemiluminescence detector (CLD-SR, Eco Physics), a second detector combined with a heated gold converter, and a standard HALO trace gas inlet. The time resolution is 1 s. The precision and accuracy of the measurements depend on the ambient concentrations, with typical values of 8 % and 5 % (NO) and 15 % and 10 % (NO_y) for levels of 0.1 and 1 ppbv, respectively (Ziereis et al., 2000, 2004).

CO and O₃

CO and O₃ were both measured with the Atmospheric Tracer EXperiment (AMTEX) instrument operated by DLR-IPA. Measurements of CO were conducted with an AeroLaser 5002 vacuum-UV resonance fluorescence spectrometer and have a precision of 1.5 ppbv and an accuracy of 2.4 % (Gerbig et al., 1996, 1999). O₃ was obtained using UV absorption with a precision of 6 ppbv and an accuracy of 5 - 10% (Baehr et al., 2003). The time resolution of both trace gases is 1 Hz.

Volatile organic compounds

The VOCs were measured by the HALO Karlsruhe Mass Spectrometer (HKMS), a proton transfer reaction mass spectrometer operated by the Karlsruhe Institute of Technology (KIT) (Brito and Zahn, 2011). The following VOCs were detected in a sequential measurement cycle with a duration of 60 s with an accuracy of 8 to 18 %: Formaldehyde, acetaldehyde, methanol, acetonitrile, acetone, methyl ethyl ketone, methyl vinyl ketone, isoprene, dimethyl sulfide, benzene, toluene, and xylene. Ratios of different VOCs are used to estimate the photochemical processing of air masses (see Sect. 3.6.2 for further explanation).

SO₂

SO₂ was measured with the CATS instrument using chemical ionisation ion trap mass spectrometry operated by DLR-IPA. The overall uncertainty is 13 % at high SO₂ mixing ratios and 31 % close to the DL (Eirenschmalz, 2021).

*3.5.4 Atmospheric transport modelling**HYSPLIT tagged CO plumes*

HYSPLIT dispersion model simulations were used (HYSPLIT4, revision SVN 804; Stein et al., 2015) to estimate the distribution of emissions from specific target regions and their proportion to the measurements. For this, a CO increase over the background concentration was modelled for each flight and interpolated on the flight path. The elevated volume mixing ratios (VMRs) are caused by fresh CO emissions originating from the predefined target regions (listed in Table 8 and shown in Fig. 1). The modelled emissions are accumulated over the past 144 h and as well split up in partial CO mixing ratios in eight different age-classes (0-3 h, 3-6 h, 6-12 h, 12-24 h, 24-48 h, 48-72 h, 72-96 h, 96-144 h). The simulations also deliver a mean age for the trajectories. The simulations are based on the meteorology of the ECMWF operational forecast (0-11 h forecast, 12-hourly updated, interpolated at 0.1° horizontally, 137 model levels, 1-hourly output). The initial CO emission rates were taken from the Emissions Database for Global Atmospheric Research (EDGAR) Hemispheric Transport of Air Pollution (HTAP) V2 emission inventory (monthly means 2010, 0.1° resolution; Janssens-Maenhout et al., 2015). The trajectory calculations achieve accuracies of 10 to 20 % of the transport distance (Draxler and Hess, 1998).

The following equation was used to calculate a weighted mean age to combine the modelled CO influence of different target regions:

$$mean\ age\ weighted = \sum^{regions} \frac{CO(region)}{CO(sum)} * mean_age(region) \quad (7)$$

FLEXTRA trajectories

The kinematic flexible trajectory model (FLEXTRA) 5.0 (Stohl et al., 1995; Stohl and Seibert, 1998) was used to determine the origin and transport history of air masses measured at flight altitude and to link the results to specific emission sources. The accuracy of the backward trajectories is around 20 % of the travel distance (Stohl, 1998). The trajectories were calculated at IUP Bremen (by Andreas Hilboll and Anna B. Kalisz Hedegaard) using meteorological reanalysis data of the fifth generation from the ECMWF (ERA5; Hersbach et al., 2020) at 0.25° horizontal resolution. The trajectories start every minute of flight time and reach back 240 h in time. Furthermore, they were linearly interpolated at a strict 5 min temporal resolution from the original trajectories.

Identification of source regions

Förster et al. (2023) developed a combined method using FLEXTRA backward trajectories and the EDGAR CO emission inventory v6.1 (Crippa et al., 2022) to identify source regions and their contributions to the measured data. For this, they used the residence time of the trajectories inside the BL over a specific area and accumulated the CO emission rates during those times gaining an emission contribution potential for the defined source regions and MPCs. Förster et al. (2023) extended the set of defined target regions and MPCs (listed in Table 8) to a broader collection including more regions all over the world. They considered only anthropogenic CO emissions (excluding BB) due to the focus of the EMeRGe project. The results of the method give a maximum CO emission contribution potential because loss processes during transport like oxidation and dilution are neglected.

3.6 DATA ANALYSIS

The following section outlines the methods used for the data analysis in Ch. 4 after integrating and applying all the calibrations and corrections mentioned in Sect. 3.2. Namely, the utilisation of distinct marker ions in the aerosol mass spectrum to examine the oxidation state of OA and the combination with photochemical ageing properties of specific VOCs. Other ways to determine the air mass age, so-called chemical clocks, are listed. The section

Table 8: Geographical extents of the target regions and MPCs defined for the EMeRGe campaigns and used for the HYSPLIT model simulations. Abbreviations: Belgium-Netherlands-Ruhr area (BeNeRu), Po Valley (PoV), Pearl River Delta (PRD), and Yangtze River Delta (YRD).

EMERGE-EUROPE		
Target region	Latitude	Longitude
BeNeRu	50° - 53° N	2.5° - 8.0° E
Berlin	51° - 53° N	13° - 15° E
London	50.5° - 52.0° N	2.0° W - 1.5° E
PoV	44° - 46° N	7° - 13° E
Munich	48° - 49° N	10.5° - 12.0° E
Paris	48° - 50° N	1.0° - 3.5° E
Rome	40° - 43° N	11° - 15° E
Spain	40° - 42° N	5° W - 3° E
EMERGE-ASIA		
Target region	Latitude	Longitude
Beijing	38.8° - 40.4° N	116° - 119° E
Manila	14° - 15° N	120.5° - 121.5° E
Osaka	34.4° - 35.6° N	135.2° W - 137.2° E
PRD	21.6° - 24.0° N	112.0° - 114.8° E
Seoul	37.3° - 37.8° N	126.5° - 127.3° E
Taipei	24.8° - 25.8° N	120.8° - 122.0° E
Tokyo	35.2° - 36.4° N	139.2° - 140.6° E
YRD	29.6° - 32.8° N	118.0° - 122.4° E

is accomplished by a part on BB markers, aerosol acidity, and a method to estimate the conversion ratio of SO_2 to SO_4 .

3.6.1 *Oxidation state of OA*

Distinct marker ions are utilised to analyse the organic mass spectrum and specify the type of aerosol further. They are characteristic e.g. for BB (see Sect. 3.6.4) or SOA formation and OA evolution/ageing, as described here. The OOA spectrum consists of mainly smaller fragments below m/z 55 (just 15 % above m/z 55; Zhang et al., 2005). A marker peak for photochemical ageing is m/z 44 (Alfarra et al., 2004; de Gouw et al., 2005; Aiken et al., 2008; Kleinman et al., 2008).

The degree of oxidation is analysed by the ratio of the signal at m/z 44 (main ion: CO_2^+) to the total organic signal (f_{44}) in combination with the ratio of the signal at m/z 43 (main ion: $\text{C}_2\text{H}_3\text{O}^+$) to the total organic signal (f_{43}) (Ng et al., 2010, 2011). Atmospheric measurements of OA fall into a triangular shape in the so-called "triangle plot" of f_{44} versus f_{43} (dashed lines in Fig. 16 are empirical, taken from Ng et al., 2010). Primary, less oxidised and photochemically younger OA particles are located at the bottom of the triangle in Fig. 16. The variability in the data points arises likely from different sources and chemical pathways. Secondary formed LO-OOA components concentrate in the lower half and MO-OOA undergo an evolution to the left upper corner of the triangle while the average and range in f_{43} decrease making the OOA particles more chemically similar regardless of the original source (Ng et al., 2010). In conclusion, oxidation decreases f_{43} and increases f_{44} . Thus, the f_{44} axis can be considered as an indicator of atmospheric ageing.

As Ng et al. (2010) stated, additional measures are needed to assign a particular source to the OOA components or to different parts of the triangular space, we compared the C-ToF-AMS results with other photochemical clocks of trace gas measurements as described in the next Sect. 3.6.2 and 3.6.3 and combined them with trajectory analyses.

3.6.2 *Photochemical ageing of VOCs*

VOCs originate from both biogenic and anthropogenic sources. Therefore, their emissions can be used to relate measurements to specific origins. For example, the long-lived VOC acetonitrile is used as a tracer for BB (Holzinger et al., 1999) or the short-lived VOCs benzene and toluene indicate anthropogenic as well as BB influence. In Ch. 4, VOCs are used on the one hand to identify plumes and specify their sources and on the other hand to separate fresh and aged air masses using the ratio of toluene to benzene.

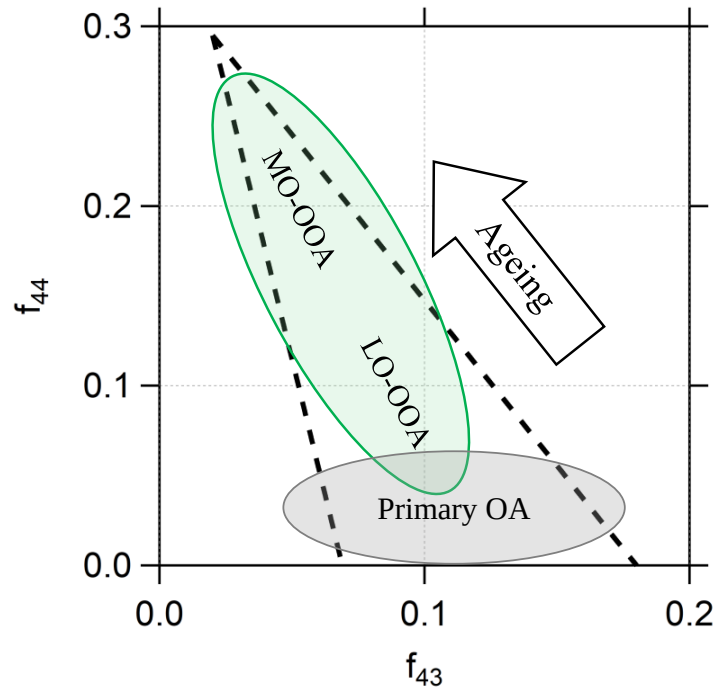


Figure 16: Triangle plot of f_{44} versus f_{43} to gain information on the oxidation state of OA particles, adapted from Ng et al. (2010), used under CC BY 3.0. The dashed lines are empirical and border atmospheric values taken at ground-based sites in the Northern Hemisphere (Ng et al., 2010) and aircraft data over Europe (Morgan et al., 2010).

Table 9: VOCs and their characteristics (tropospheric lifetime, DL, instrumental noise, and calculated threshold as $DL+3*\text{noise}$) used for the identification of plumes of different origin after Förster et al. (2023) (updated values). Values for EMeRGe-Europe/-Asia.

VOC	LIFETIME	ORIGIN	DL (pptv)	NOISE (pptv)	THRESHOLD (pptv)
Acetonitrile	6 months	BB	145*	13/18	184/199
Benzene	10 d	anthr., BB	29/15	10/15	59/60
Isoprene	2 h	biogenic	49/25	13/11	88/58
Toluene	2 d	anthr., BB	20/17		

* For acetonitrile an estimated atmospheric background is utilised (derived from the north hemispheric mean concentration measured with IAGOS-CARIBIC in winter (DJF) 2012 - 2016; Brenninkmeijer et al., 2007 and supplement of Förster et al., 2023). For the other VOCs the DL is used instead due to their shorter atmospheric lifetimes. The lifetimes mentioned are the tropospheric lifetimes as defined for a 12 h daytime average hydroxide (OH) concentration of 2.0×10^6 molecules cm^{-3} (Atkinson, 2000).

The identification of plumes and additional distinction between background and anthropogenically influenced air was done in accordance with Förster et al. (2023). There, a threshold is calculated from the DL or background value plus 3 times one sigma of the instrumental noise during flight (see Tab. 9). Combining the thresholds of up to three VOCs assigns the air mass to a specific source (see definitions in Tab. 10). For example, tagging pure anthropogenic emissions is done by using the benzene data above the threshold while excluding BB influence of acetonitrile. The differentiation between anthropogenic and background air was done utilising the absence of benzene, acetonitrile, and isoprene to define the background (also excluding fresh biogenic emissions). Further, the ratio between toluene and benzene is an appropriate marker for photochemical ageing (Roberts et al., 1984; Warneke et al., 2007). The values shift towards benzene with time because toluene degrades faster. Close to a source and the fresh emissions, the ratio is greater than one, for aged and more processed air masses it is less than one.

3.6.3 Other chemical clocks

Various other chemical clocks are used throughout the literature, considering ageing of aerosol particles as well as processing and dilution of trace gases. A few of them are listed here for reference and the NO_y to CO ratio is also applied during the analysis of this thesis.

Table 10: Definitions to distinguish between background and anthropogenically influenced air as well as different ages after Förster et al. (2023) (updated values). Values for EMeRGe-Europe/-Asia. "anthr."=anthropogenic. Background means clean air conditions without any anthropogenic, BB, or biogenic signatures.

SOURCE / AGE	DEFINITION
Pure anthr.	benzene > 59/60 pptv & acetonitrile < 184/199 pptv
Strong anthr.	benzene > 130 pptv & acetonitrile < 184/199 pptv
BB influence	acetonitrile > 184/199 pptv
Fresh biogenic	isoprene > 88/58 pptv
Background	benzene < 59/60 pptv & acetonitrile < 184/199 pptv & isoprene < 88/58 pptv
Fresh [*]	toluene/benzene > 1
Aged [*]	toluene/benzene < 1

^{*} Before forming the ratio of toluene to benzene, values below the DL were filtered out for both VOCs separately. This reduces the number of data points.

For instance, others used the (logarithmic) ratio of NO_x to NO_y as ageing timescale. Hereby, lower values indicate air masses close to the source and the ratio increases with ageing (Zellweger et al., 2003; Kleinman et al., 2007; Freney et al., 2014). Kleinman et al. (2007) correlated this clock to exposure times of OH, concentrations of OA, VMRs of CO, and f_{44} . They found NO_x to NO_y ratios ranging from 0.5 in the morning to 0.16 in the afternoon for airborne measurements near urban centres in New England and the Mid-Atlantic States. Zellweger et al. (2003) found that this clock performs best close to the emissions sources on timescales less than one day. In addition, Freney et al. (2014) derived correlations for the ratios OA/BC and OA/CO with increasing ageing time of NO_x to NO_y during aircraft measurements around Paris in summer. However, NO_x was not measured during EMeRGe and the distances to the sources are too large to capture the evolution of emissions on these timescales.

In addition, the age distribution of aerosol species can be inferred from chemical transport models. Wagstrom and Pandis (2009) found a large variety of ages influenced by the factors location, time, and aerosol species. Furthermore, a distinction between primary and secondary aerosol was observed. The results gave average ages of around 24 h for primary aerosol particles over polluted continental regions in the Eastern USA during summer. In contrast, secondary aerosol particles had lifetimes of 48 - 72 h near the surface (Wagstrom and Pandis, 2009). Additionally, average ages increased

vertically and horizontally with increasing distance to the source. Particle size became important for non-volatile species (Wagstrom and Pandis, 2009).

NO_y to CO ratio

An ageing method applied in this thesis is the ratio of NO_y to CO as it further assesses the anthropogenic input to air masses (Parrish et al., 1991). It is based on different atmospheric lifetimes, e.g. CO is long-lived with respect to deposition while NO_y lifetimes are shorter with about 2 d (Stohl et al., 2002; Simon et al., 2018). Therefore, air masses close to the surface become enriched in CO relative to NO_y as they age which results in smaller NO_y to CO ratios (Parrish et al., 1991; Stohl et al., 2002; Zellweger et al., 2003; Simon et al., 2018). Zellweger et al. (2003) found pollution events with NO_y to CO ratios greater than 0.01 and a seasonal variation of the ratio with lower values in winter due to the longer CO lifetime. The method is suitable for airmasses within the BL as only 3 - 5 % of the NO_y emissions reach altitudes above 3 km (Stohl et al., 2002). Furthermore, the evolution of fresh emissions can be tracked on a timescale of 5 - 6 d because after this time NO_y from anthropogenic sources becomes equal to in situ formation (Stohl et al., 2002).

3.6.4 *Biomass burning*

Similarly to the marker ions for aerosol ageing defined in Sect. 3.6.1, a marker ion for BB is observed within the organic mass spectrum, namely m/z 60 (Schneider et al., 2006; Alfarra et al., 2007). Levoglucosan (C₆H₁₀O₅) is supposed to produce signals at m/z 60 but also at m/z 73 (Chen et al., 2022). Other substances influencing m/z 60 are e.g. carboxylic acids from SOA (DeCarlo et al., 2008; Docherty et al., 2008; Ulbrich et al., 2009; Aiken et al., 2010), fatty acids in cooking POA, and to a smaller extent motor vehicle exhaust (Mohr et al., 2009). The ratio of the signal at m/z 60 (main ion: C₂H₄O₂⁺) to the total organic signal (f_{60}) is calculated accordingly to f_{43} and f_{44} . The ageing of BB influenced OA particles can be analysed within a triangle plot, as depicted in Fig. 17 (Cubison et al., 2011). Atmospheric and laboratory measurements of OA were observed within the triangle and showed decreasing f_{60} and increasing f_{44} during ageing. In addition, acetonitrile is known to correlate well with fresh BB OA and the evolution of f_{60} (Aiken et al., 2009, 2010; Lack et al., 2013). According to Förster et al. (2023), acetonitrile values greater than 184 pptv are used to filter the EMeRGe-Europe data for BB influence and values greater than 199 pptv are applied for EMeRGe-Asia.

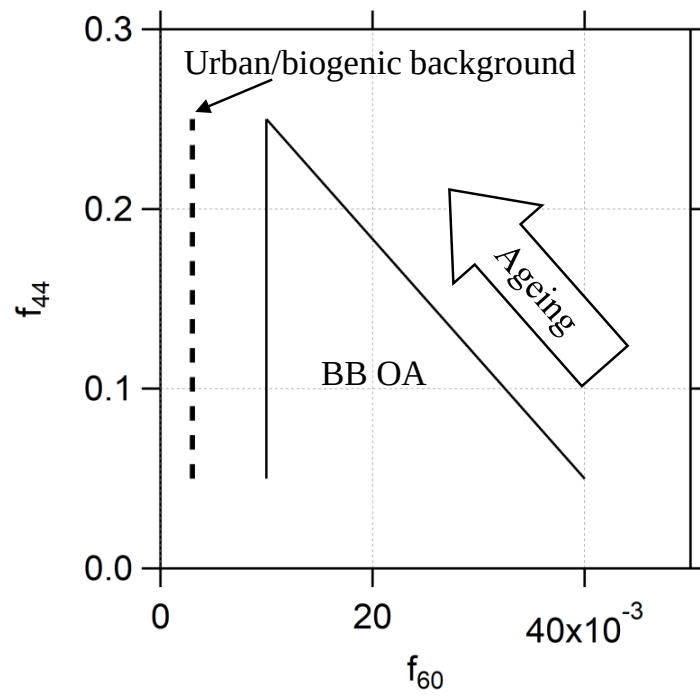


Figure 17: Triangle plot of f_{44} versus f_{60} to gain information on the oxidation state of BB influenced OA particles, adapted from Cubison et al. (2011), used under CC BY 3.0. Atmospheric and laboratory BB OA values accumulate inside the triangle and approach the dashed line by ageing which is the urban/biogenic background of approximately 0.3 %.

3.6.5 Acidity

Acidic aerosol particles are ubiquitous in the atmosphere having significant implications for the atmospheric lifetime of pollutants, the climate, and human health. For example, atmospheric acidic aerosol particles influence visibility and climate as they are prone to be more hygroscopic than their neutralised form enhancing the ability to scatter light and act as CCN (Khlystov et al., 2005; Pathak et al., 2004). Furthermore, particle phase acidity could enhance SOA formation significantly as measured in the atmosphere and in chamber studies (Jang et al., 2002; Edney et al., 2005). In Pye et al. (2020), a detailed review on the atmospheric processes influenced by acidity and the involved aspects in the Earth system is given. Acidity values of atmospheric aerosol particles and cloud droplets cover a high range when given in units of pH and vary as a function of particle size (Zhang et al., 2007b; Pye et al., 2020). Anthropogenic emissions can both increase (e.g. SO₂, NO_x, and organic acids) and reduce (e.g. ammonia, non-volatile cations, and amines) acidity (Pye et al., 2020).

Acidity calculations are performed using the inorganic fraction of the C-ToF-AMS data. The method after Zhang et al. (2007b) determines whether the aerosol particles are neutralised or acidic by using ammonium that takes part in the neutralisation of inorganic species measured by the C-ToF-AMS (i.e. sulfate, nitrate, and chloride). Equation 8 yields the amount of ammonium needed to neutralise the measured inorganic species (NH_{4,neut}). The particle acidity is evaluated based on this value related to the measured ammonium (NH_{4,meas}) (see Eq. 10).

$$\text{NH}_{4,\text{neut}} = \frac{2 \cdot 18}{96} \cdot \text{SO}_4 + \frac{18}{62} \cdot \text{NO}_3 + \frac{18}{35.5} \cdot \text{chloride} \quad (8)$$

$$\Delta \text{NH}_{4,\text{neut}} = \sqrt{\left(\frac{2 \cdot 18}{96} \cdot \Delta \text{SO}_4\right)^2 + \left(\frac{18}{62} \cdot \Delta \text{NO}_3\right)^2 + \left(\frac{18}{35.5} \cdot \Delta \text{chloride}\right)^2} \quad (9)$$

$$\frac{\text{NH}_{4,\text{meas}}}{\text{NH}_{4,\text{neut}}} \begin{cases} \geq 1 \pm \sigma, & \text{neutralised particle periods} \\ \approx 0.75, & \text{equal mole conc. of } (\text{NH}_4)_2\text{SO}_4 \text{ and } \text{NH}_4\text{NO}_3 \\ < 0.75 \pm \sigma, & \text{more acidic particle periods,} \end{cases} \quad (10)$$

whereby σ is the analytical error based on error propagation of the measurement uncertainty and noise for NH₄, SO₄, NO₃, and chloride; on average, 4 % of the corresponding ratio (Zhang et al., 2007b).

The results of the acidity calculation for EMeRGe-Europe and -Asia are presented in Sect. 4.2. In general, this type of analysis does not consider

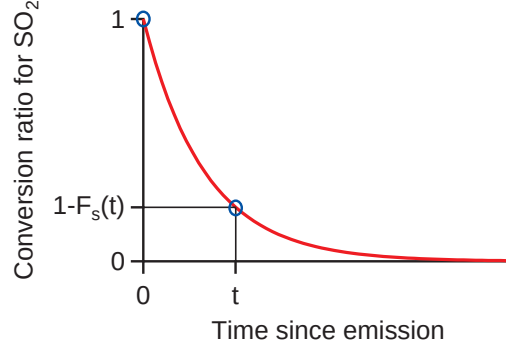


Figure 18: Calculation of the e-folding time (T_e) using two known points on the conversion curve of SO_2 marked with the blue circles: 100 % SO_2 at the emission source at time $t = 0$ and the reduced ratio F_s for SO_2 at the time t of the measurement.

that there are other neutralising compounds such as amines (Ge et al., 2011). Organic substances can also be neutralising (as in organic sulfates), but they can also act as acids (e.g. organic acids) (Zhang et al., 2007b; Froyd et al., 2010). However, organics are not included here.

3.6.6 Conversion ratio of SO_2

Grosjean and Friedlander (1975) and Kadowaki (1986) defined the conversion ratio of sulfur (F_s) as

$$F_s = \frac{\text{SO}_4}{\text{SO}_2 + \text{SO}_4} \quad (11)$$

where SO_4 is the particulate sulfate concentration (converted from $\mu\text{g m}^{-3}$ to ppbv) and SO_2 is the gas phase SO_2 VMR in ppbv. The ratio indicates the fraction of sulfur that has been converted to sulfuric acid and condensed to particulate sulfate.

The e-folding time (T_e) can be calculated with Eq. 12 and as illustrated in Fig. 18 by knowing two points along the exponential conversion curve of SO_2 : 100 % SO_2 at time $t = 0$ emitted at the source (e.g. a volcano or power plant) and the reduced ratio F_s for SO_2 at the measurement point after time t .

$$T_e = \frac{-t}{\ln(N(t)/N(0))} \quad (12)$$

with t the time between measurement and emission, $N(t) = 1 - F_s(t)$ the conversion ratio of SO_2 at the time t , $N(0) = 1$ the initial ratio at the time of emission (in case of a volcano the eruption time).

This method is applied to the EMERGe-Europe and -Asia data in Sect. 4.2.4.

RESULTS

In this chapter, the results of the C-ToF-AMS data analysis from the EMeRGe campaigns are presented. According to the objectives of the project mentioned in Sect. 2.1 and to address the central questions of this thesis (see Sect. 1.3), the aerosol particle composition in the vicinity of MPCs is presented. Furthermore, an enhanced acidity of aerosol particles due to SO₂ emissions is discussed and the oxidation state of aerosol particles is evaluated. The EMeRGe-Asia data set is restricted to the flights E-AS-03 to E-AS-14, disregarding the first and last transfer flights to focus only on the emissions in East Asia.

4.1 AEROSOL PARTICLE COMPOSITION IN THE VICINITY OF MPCs

This section gives an overview on the regional and vertical distribution of the aerosol particle composition to gain a general impression of the conditions in Europe and East Asia during the measurement periods. Then, a comparison between anthropogenic influenced and background air is presented. Additionally, a distinct pollution event in the Gulf of Thailand is analysed. Furthermore, the composition of individual MPCs in Europe and East Asia is examined. The section closes by relating the results to the literature. Although the data set is biased towards MPC emissions, owing to the scientific objectives of EMeRGe and the according flight strategy (see Fig. 6), the comparison is useful to understand differences in the aerosol composition between the two areas.

4.1.1 *Regional distribution*

The averaged aerosol compositions in relation to different mass concentration ranges indicated similarities and differences for EMeRGe-Europe and -Asia (see Fig. 19). The sum of all aerosol species is comparable between the regions for the first two bars up to 10 $\mu\text{g m}^{-3}$ while the total mass concentration between 10 and 20 $\mu\text{g m}^{-3}$ is higher in East Asia than in Europe and even values in the greatest subset (20 - 35 $\mu\text{g m}^{-3}$) are measured. The chloride concentrations are included but are below 0.01 $\mu\text{g m}^{-3}$ (0.6 % of the total mass) in Europe and 0.25 $\mu\text{g m}^{-3}$ (1.16 % of the total mass) in East Asia. The mean total mass concentration ('All' bars in Fig. 19) is a little lower during EMeRGe-Asia but this overall average is not representative because

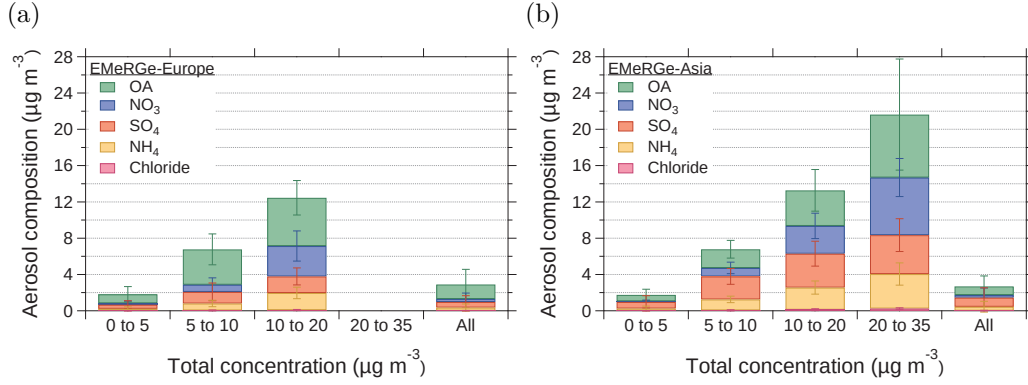


Figure 19: Average aerosol composition from C-ToF-AMS data as a function of binned concentration ranges for EMeRGe-Europe (a) and -Asia (b; flights E-AS-03 to E-AS-14). The bar for 20 - 35 $\mu\text{g m}^{-3}$ for Europe is missing because the maximum measured concentration is 18.7 $\mu\text{g m}^{-3}$. The 'All' bar averages all data and is not filtered by concentration. Error bars are the standard deviation of the mean value. The y-axes are aligned for both campaigns to make the bars easily comparable. The relative shares of each bar are listed in Table 23 in the appendix.

the aircraft flew higher more often to reach the areas of interest faster and covered greater distances during the flights than for the European part.

OA is the prevalent species in Europe (see Fig. 19a) making up more than half of the composition for concentrations up to 10 $\mu\text{g m}^{-3}$ and still accounts for 42.7% at higher concentrations. All fractions are listed in Table 23 in the appendix. The second largest aerosol species is SO_4 with a decreasing relative contribution from 24.6 to 14.6% for increasing total mass concentrations while NO_3 is increasing from 8.7 to around 27%. NH_4 is observed in all concentration subsets with an amount of 11.3 to 15.2%. The EMeRGe-Asia data set in Fig. 19b generally indicates an aerosol composition with more SO_4 and less organics (up to 41.3% and 39.7% in the smallest concentration subset, respectively). The fraction of both species is decreasing with increasing total concentration while NO_3 is gaining importance with up to 30% for the highest concentration subset. NH_4 is measured with an average of around 16%.

The geographical distribution of the C-ToF-AMS concentrations during EMeRGe-Europe is depicted in Fig. 20 and in Fig. 61 in the appendix. Figure 20 contains exemplarily the OA mass concentration because it makes up the largest fraction of the aerosol species in Europe. The greatest OA values are found over the Mediterranean, especially close to Spain, France, and Italy (in the PoV and over the Adriatic Sea). Other areas of elevated concentrations occur over the North Sea, the target region BeNeRu (defined in Table 8 and Fig. 1a), the English Channel, and south-west of Paris. The panels of Fig. 61 in the appendix comprise the colour-coded flight paths for the other C-ToF-AMS species during EMeRGe-Europe, showing the strongest signal

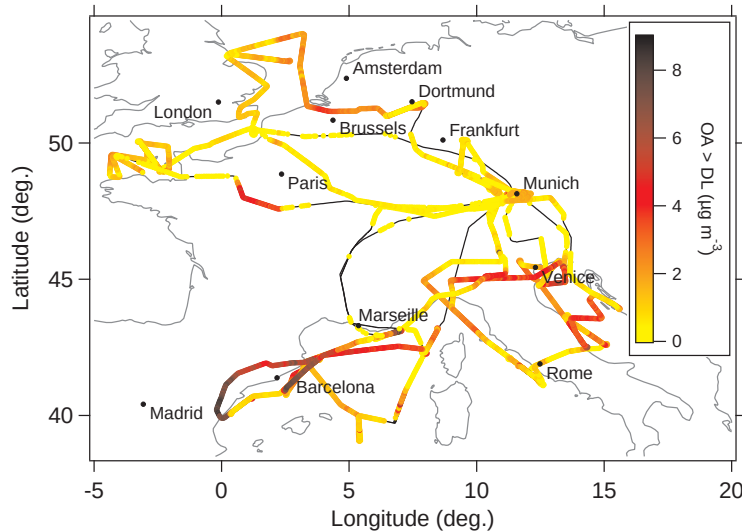


Figure 20: Flight paths of the EMeRGe-Europe campaign in summer 2017 colour-coded with the OA mass concentration. The data is filtered for values below the DL to increase clarity and filter measurements are left out, too. The colour scale goes up to $9 \mu\text{g m}^{-3}$, although the OA maximum is $13 \mu\text{g m}^{-3}$.

for SO_4 north-west of Rome over the Tyrrhenian Sea (further investigated in Sect. 4.2.1) and other smaller mass concentration increases over the Mediterranean and the BeNeRu. The NO_3 mass concentration is greatest in the BeNeRu, the PoV, and the English Channel. The distribution of the NH_4 mass concentration is comparable to the OA but the elevated concentrations close to Spain, France, and Paris are less pronounced. Only a few data points are available for chloride because the values often are below the DL showing some small signatures on the east coast of Great Britain and over the BeNeRu.

The geographical distribution of the C-ToF-AMS concentrations during EMeRGe-Asia is depicted in Fig. 21 and in Fig. 62 in the appendix. Figure 21 contains exemplary the OA mass concentration in East Asia. As the flight patterns in Figs. 1b and 21 imply, the same path is used during many flights making it difficult to see elevated concentrations because the flight routes overlap each other. Nevertheless, some increases for the OA are observed in the Gulf of Thailand (during the two transfer flights), over the Philippines close to and west of Manila, and around Taiwan. Other maxima can only be guessed and require further in-depth analysis on a flight-to-flight basis. The same applies to the other C-ToF-AMS species (see Fig. 62 in the appendix).

4.1.2 Vertical distribution

The mean vertical profiles for the five C-ToF-AMS species were already displayed in Figs. 10 and 11 in Sect. 3.2.3 together with their corresponding

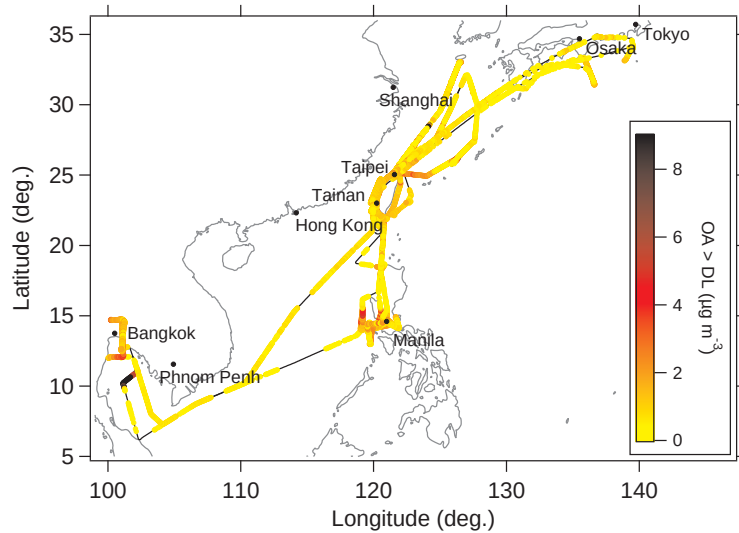


Figure 21: Flight paths of the EMeRGe-Asia campaign flights E-AS-03 to E-AS-14 in spring 2018 colour-coded with the OA mass concentration. The data is filtered for values below the DL to increase clarity and filter measurements are left out, too. The colour scale goes up to $9 \mu\text{g m}^{-3}$ (to make it comparable to the European values), although the OA maximum is $27 \mu\text{g m}^{-3}$.

DLs. Referring back to those figures, the aerosol mass concentrations of the C-ToF-AMS species are higher at altitudes below 4 km and decrease with increasing altitude. One of the most prominent differences between Europe and East Asia is the higher SO_4 mass concentration in the Asian BL (almost doubled in the lowest altitudes), which is accompanied, but less pronounced, by the NH_4 mass concentration. Anthropogenic SO_2 emissions and resulting SO_4 formation are mainly caused by the combustion of fossil fuels (coal and oil). However, extensive interventions in recent decades, such as the desulfurisation of power plant emissions in Western Europe, as well as the industrial collapse in Eastern Europe and the former Soviet Union, have led to a significant reduction in anthropogenic SO_2 emissions in these regions. In contrast, strong industrialisation in East and South Asia has led to a further increase in SO_2 emissions. In comparison, the averaged OA mass concentrations are markedly higher in Europe than in East Asia. Above 4 km, the mass concentrations remain constantly low until OA and NO_3 increase again slightly above 9 km. The chloride mass concentrations are very low for both campaigns and the median only exceeds the DL below 3 km and around 8 km during EMeRGe-Europe as well as below 5.5 km for EMeRGe-Asia.

The profiles of the relative compositions in Fig. 22 accompany the results of the averaged vertical profiles in Sect. 3.2.3 and assign numerical values to the findings. The fraction of OA below 4 km is 40 - 76 % and 27 - 52 % for EMeRGe-Europe and -Asia, respectively, while the SO_4 mass fraction ranges from 12 to 44 % and 30 to 42 % for the two regions in this altitude range.

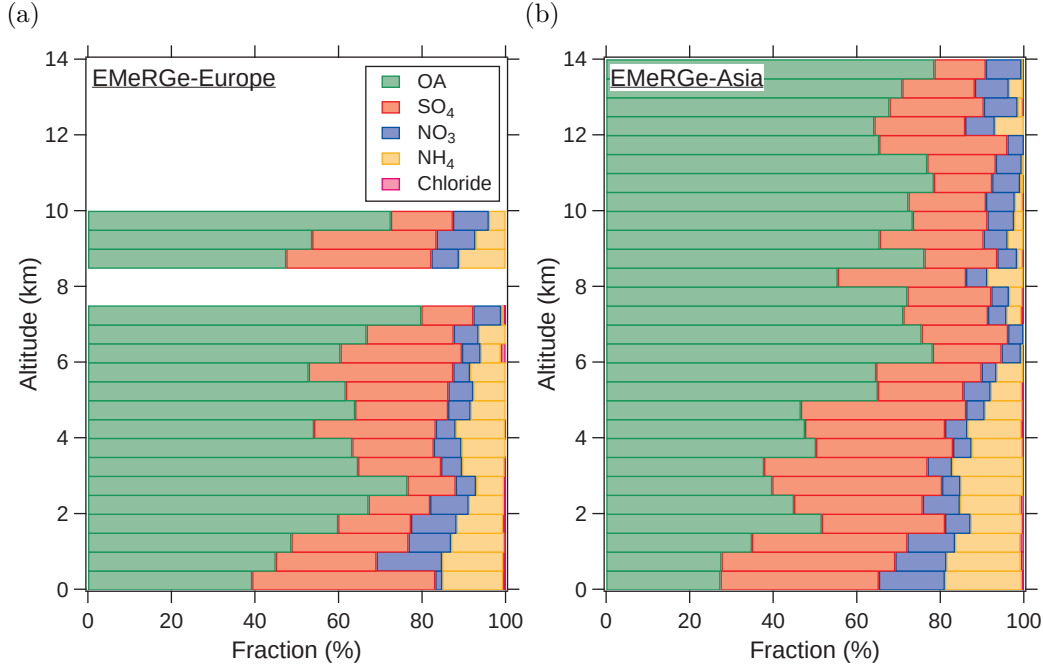


Figure 22: Vertically resolved relative aerosol composition from C-ToF-AMS data for EMeRGe-Europe (a) and -Asia (b; flights E-AS-03 to E-AS-14) in 500 m altitude bins. Negative values were set to zero before calculating the fractions and the bins around 8 km in (a) are left out due to negative OA mass concentrations.

The maximum NH₄ fraction accounts for around 15% in Europe and 18% in East Asia in the lower levels. The NH₄ proportion remains above 4% up to the maximum flight altitude of around 10 km for EMeRGe-Europe while the NH₄ fraction is smaller above 6 km in East Asia. The NO₃ contribution is maximum at around 16% with an average of approximately 6% for both campaigns. The chloride fraction is small and contributes with a maximum of 1%.

Meridional intersections are appropriate to compensate for the shortcoming of overlapping flight paths of the map representation in the previous section. For EMeRGe-Europe, the location of the Alps is visible in Fig. 23a by the data gap up to 5 km with raised flight altitudes between 46 and 48° N. The total aerosol mass concentration is higher below 4 km south of the Alps and below 3 km north of the Alps implying a slightly elevated BL in Southern Europe. This is in accordance with the meteorological conditions described in Sect. 2.2, presenting enhanced precipitation rates in Northern Europe and dry conditions in the south, favouring wet deposition of aerosol particles and strengthened upward transport of emissions, respectively. The greatest concentrations are not always measured close to the surface but are transported to or formed at higher altitudes, like the values between 2 and 4 km observed between 40 and 42° N over Spain and south of France or the signal

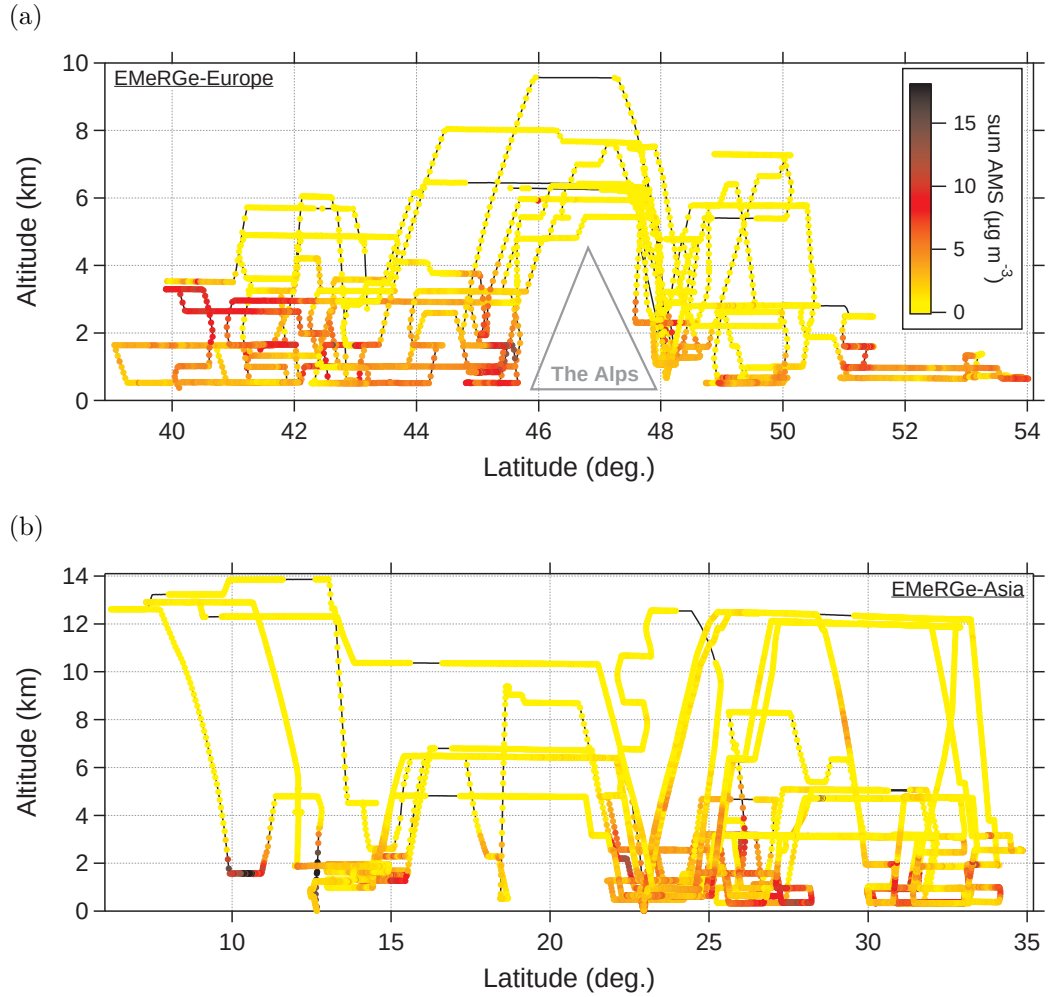


Figure 23: Meridional cross sections of the sum of all C-ToF-AMS species for the whole campaigns EMeRGe-Europe (a) and -Asia (b; flights E-AS-03 to E-AS-14). Missing data points are due to filter measurements. The colour scale is applicable to both plots and has a maximum of $18 \mu\text{g m}^{-3}$, although the local maxima are $18.7 \mu\text{g m}^{-3}$ and $33.9 \mu\text{g m}^{-3}$ for EMeRGe-Europe and -Asia, respectively.

at around 51°N over the BeNeRu between 1 and 2 km. This is contrary to the signature of the PoV aerosol particles which is observed from the lowest flight altitude up to almost 4 km at around 45°N . Figure 23b gives similar results for the EMeRGe-Asia campaign with increased aerosol mass concentrations below 4 - 5 km. The greatest values are measured in vicinity of the U-Tapao international airport in Thailand between 10 and 13°N where the aircraft made a stopover during the transfer flights to and from Taiwan. Around 15°N , increased concentrations are due to Manila emissions. The dip at 18.5°N is performed to validate the CO_2 and CH_4 measurements at the Total Carbon Column Observing Network (TCCON) site in Burgos, Philippines (Ohyama et al., 2020). An aerosol layer is observed there between 2 and 3 km during flight E-AS-03 with air masses originating mainly in India and mixing with local emissions from the Northern Philippines and the adjacent East China Sea (based on the FLEXTRA backward trajectories and the calculated source region contributions in Förster et al. (2023), introduced in Sect. 3.5.4). Many data points cluster below 3 km between 22 and 28°N due to the intense measurements around and north of Taiwan where the aircraft base is located. Apparently, the situation in this area is not homogeneous, but depends on a number of different factors and requires more detailed analyses like in Sect. 4.2.3 and 4.3. Another area of intense measurements is located at 30 to 35°N over the East China Sea involving stacked flight patterns to study the transported outflow from different MPCs. Furthermore, pollution layers are crossed while changing altitudes like the one measured during flight E-AS-11 between 7 and 8 km altitude at around 29°N originating mainly in Southeast Asia/Indochina mixing with smaller emissions from Southern China and India (based on the FLEXTRA backward trajectories and the calculated source region contributions in Förster et al. (2023)).

4.1.3 Anthropogenic influence

A first comparison of the pollution levels between the two campaigns is given in Fig. 24. Here, a scatter plot of the OA mass concentration and the CO VMR as a tracer for anthropogenic emissions is depicted, showing a clear difference between the regions. This is mainly caused by the higher CO values in East Asia originating from fires and vehicle/industrial emissions rising up to almost 2000 ppbv while the maximum CO VMR in Europe is around 320 ppbv. Thus, the majority of the European data points is accumulated below 150 ppbv CO which is in accordance to other aircraft measurements over Europe (Xue et al., 2023). The higher proportion of OA during EMeRGe-Europe becomes once again evident except for one event over the Gulf of Thailand during EMeRGe-Asia with OA mass concentrations increasing to $27\text{ }\mu\text{g m}^{-3}$. This is explained in more detail in Sect. 4.1.4. Despite performing

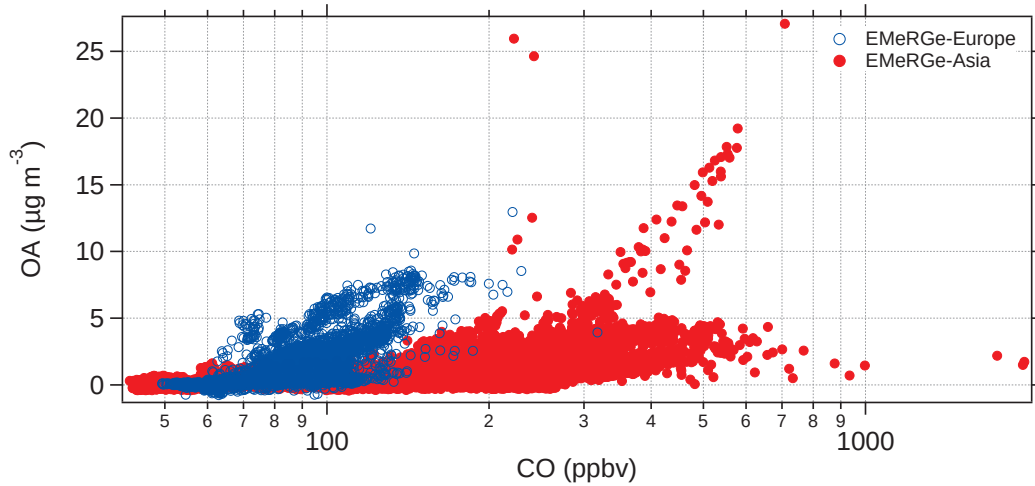


Figure 24: Scatter plot of the OA mass concentration versus the logarithmic CO VMR colour-coded to separate the two EMeRGe campaigns. The Asian part is restricted to flights E-AS-03 to E-AS-14.

around 30 % of the flight sections under cleaner conditions at altitudes above 4 km (see Fig. 6), the enhanced pollution levels in East Asia are remarkable.

Referring to the main focus of EMeRGe, the vertical profiles of the aerosol species are now separated for anthropogenically influenced air masses utilising external VOC data in accordance to Sect. 3.6.2. Based on Förster et al. (2023), around 18 % of the flight time anthropogenic emissions and in 40 % background air has been sampled during EMeRGe-Europe. The rest of the European measurements are influenced by BB (2 %), pure biogenic origin (1 %), or could not be attributed due to VOC data gaps (39 %). During EMeRGe-Asia, approximately 32 % of the measurements are anthropogenically influenced, 15 % classified as background, and around 37 % could not be attributed (Förster et al., 2023). The remaining 16 % are related to BB.

Higher concentrations occur in the anthropogenically influenced air masses compared to the background in the vertical profiles of EMeRGe-Europe (see Fig. 25a). However, the direct influence of anthropogenic emissions on the aerosol components in the free troposphere above 4 km becomes small or is rather negligible as there are no data points attributed to anthropogenic pollution above 5.5 km. The OA increases around 3 km which is mainly caused by one particular measurement period above Spain during flight E-EU-09. Thereby, anthropogenic emissions from Madrid are lifted and transported to the measurement location (Andrés Hernández et al., 2022; see Sect. 4.1.5). Further possible explanations might be lower temperatures favouring SOA formation in this altitude range or a longer conversion time of VOCs to SOA in comparison to the conversion time for inorganic aerosol precursor gases. In contrast to the OA profile, the inorganic aerosol components, especially sulfate and ammonium, show a steady decrease with altitude in the

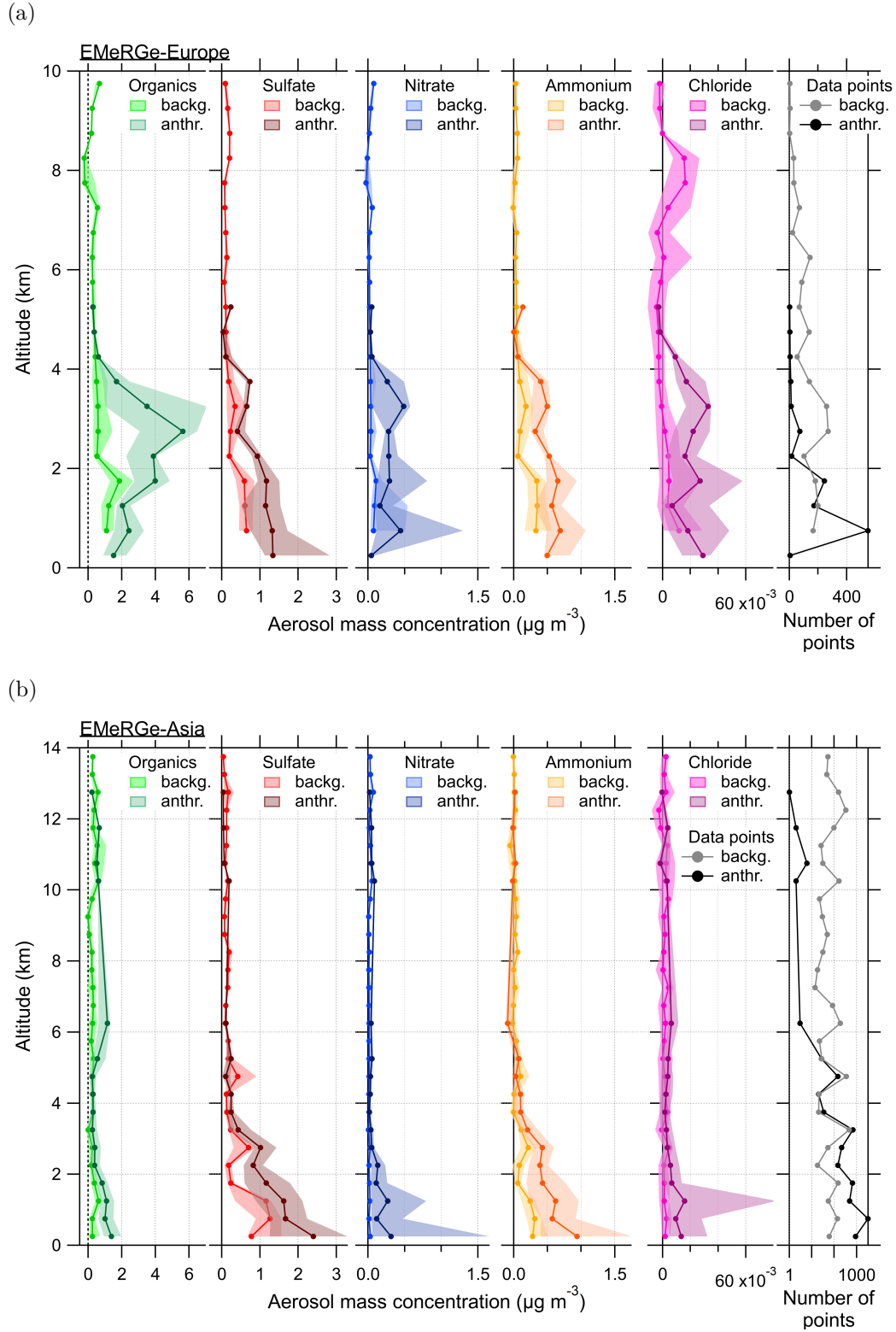


Figure 25: Averaged vertical profiles of the C-ToF-AMS species for the EMeRGe-Europe (a) and -Asia (b; flights E-AS-03 to E-AS-14) campaigns in 500 m altitude bins. The dots represent the medians and the shaded areas the interquartile ranges. The vertical profiles are differentiated for anthropogenic and background origin. Axis ranges are aligned for the C-ToF-AMS species. The last profile contains the number of data points per altitude bin, given on a logarithmic scale for (b).

anthropogenically polluted air. The NO_3 below 4 km is predominantly anthropogenic as the background NO_3 is almost zero. Only below 2 km, a BL structure is existent in the background NO_3 mass concentrations. Chloride is showing a similar behaviour although the mass concentrations are small. In general, the lowest bin (0 - 500 m) has poor statistics and should be treated carefully, more precisely it does not contain any data for the background and also the anthropogenic median consists of only 5 data points. The number of anthropogenic data points is sufficient up to 3.5 km, above are less than 10 per altitude bin. The background statistics are better with still 29 points at 8.25 km.

In contrast to the European part, the separated vertical profiles for EMeRGe-Asia in Fig. 25b contain a few anthropogenically influenced data points at higher altitudes above 5 km, although the values are quite small, do not exceed the background mass concentrations, and contain a maximum of 6 data points per bin. The particles could have been removed by wet deposition in these altitudes. An exception is the anthropogenic OA data point around 6 km exceeding the background profile, probably induced by long-range transport. In the lower 4 km, the anthropogenic aerosol mass concentrations are highest at ground and decrease with increasing altitude. The OA mass concentrations are significantly lower than in Europe and no maximum occurs around 3 km. NO_3 and chloride mass concentrations are higher below 2.5 km and mostly anthropogenic. SO_4 and NH_4 behave similarly and decrease with altitude, showing a smaller maximum at around 2.75 km. Statistics are good for the background profiles with an average of 98 and a minimum of 14 points per bin while the main anthropogenic influence reaches up to 5.5 km. The shape of the background profiles is comparable for both regions even though the exact values differ. More precisely, there is a BL reaching up to around 2 km followed by another layer ranging up to 4 km of slightly increased mass concentrations. This is especially observed for the background SO_4 and NH_4 but also the OA shows the BL effect.

4.1.4 Characterisation of a distinct pollution event - Gulf of Thailand

The greatest OA mass concentrations measured during EMeRGe have already been introduced in Sect. 4.1.3 and their regional as well as vertical extension can be seen from Figs. 21 and 23. The event occurred during flight E-AS-03 on 12 March 2018 right after the start around 04:50 UTC and could be observed until 05:47 UTC. The corresponding scatter plot of the OA mass concentration versus the CO VMR is depicted in Fig. 26a showing a clear correlation between the two variables (Pearson r of 0.96) as they are increasing about one order of magnitude. The flight path in Fig. 26b is colour-coded with the OA mass concentration, showing elevated concentrations directly after take-off from the international airport in U-Tapao in

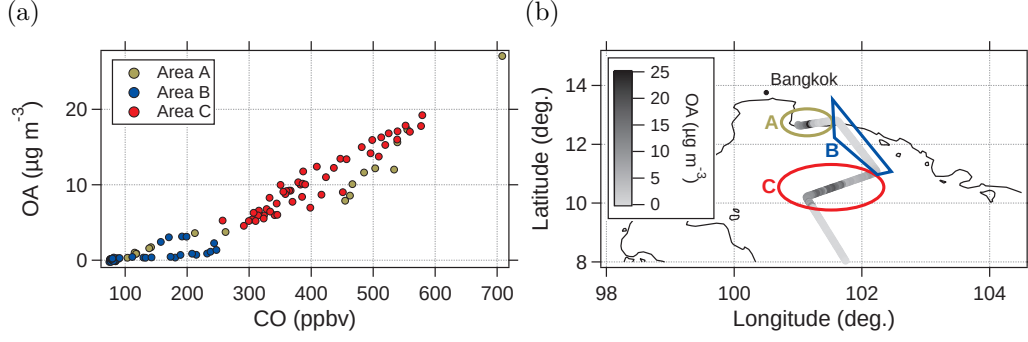


Figure 26: (a) Scatter plot of the OA mass concentration versus the logarithmic CO VMR for flight E-AS-03 04:50 - 05:57 coordinated universal time (UTC). The data are colour-coded to separate the three different areas marked on the map of the Gulf of Thailand (b) where the flight track is shaded with the OA mass concentration.

area A and over the ocean south-east of Bangkok in area C. The maximum of the OA mass concentration is $27.1 \mu\text{g m}^{-3}$ and the total non-refractory aerosol mass concentration (approximately PM_1) is $33.9 \mu\text{g m}^{-3}$. Kim Oanh et al. (2013) studied air pollution in Bangkok in 2010 at different ground sites in the vicinity of roads in urban and suburban areas. The dry season values (December and January 2010; dry season in Bangkok is from November till April) for $PM_{2.5}$ are $52 - 61 \mu\text{g m}^{-3}$. Sresawasd et al. (2021) examined haze effects from BB on aerosol particles during the dry season in Northern Thailand. They found a haze period in March and April 2018 with $PM_{2.5}$ values of $40 - 70 \mu\text{g m}^{-3}$. These values exceed the WHO guideline for a 24 h average of $PM_{2.5}$ of $25 \mu\text{g m}^{-3}$ (valid 2005 - 2021, since 2021 even stricter at $15 \mu\text{g m}^{-3}$; World Health Organization, 2006, 2021), while the Thai National Ambient Air Quality Standard is $50 \mu\text{g m}^{-3}$ (Fold et al., 2020). Thus, our mass concentrations observed with the aircraft in a distance from the source are lower compared to ground-based stations. The areas marked in Fig. 26b are defined as a combination of flight altitude, trajectory analysis, and increased aerosol particle and trace gas values. Their colours are used to assign the data in Fig. 26a to the three areas and will appear throughout this section. From Fig. 26a, it is apparent that lower values are measured in area B while increasing concentrations and VMRs could be observed in the areas A and C.

The flight sections of higher aerosol mass concentrations are indicated with the ellipses A and C in Fig. 26b. The area B between them is characterised by lower values, mainly due to an ascent of the aircraft up to 4.8 km. The same flight segments are framed in the time series in Fig. 27. In addition to the high OA mass concentrations, the inorganic aerosol species including rBC are increased in area A and C as well (see Fig. 27 and Fig. 28a and b). The second largest aerosol component is rBC with up to $3.3 \mu\text{g m}^{-3}$, followed by

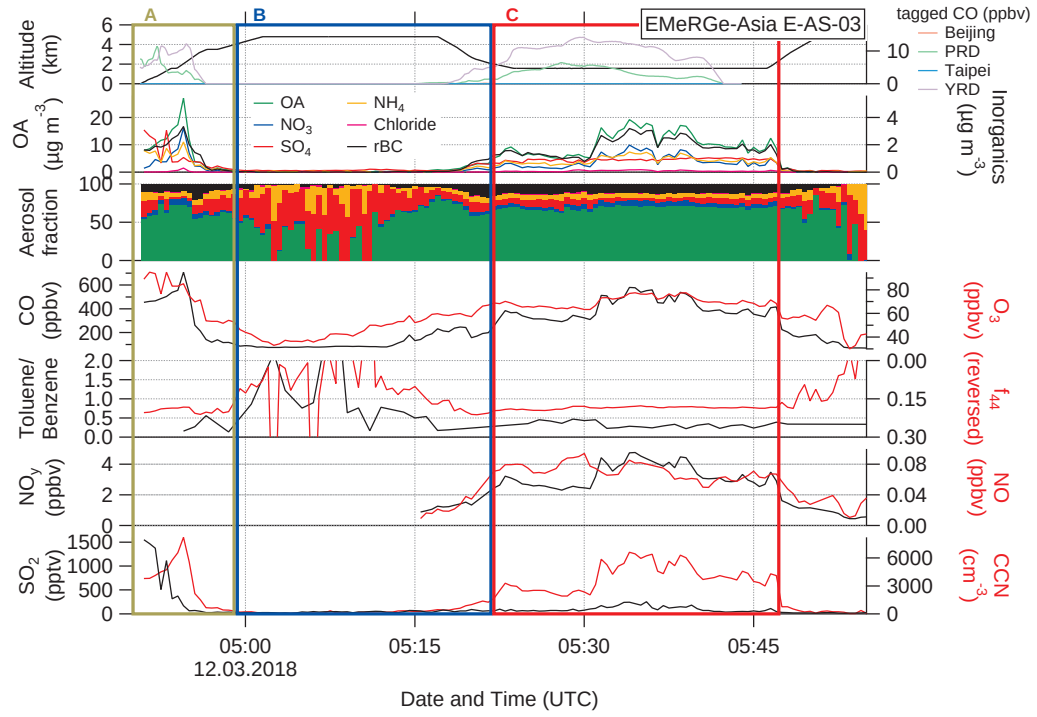


Figure 27: Excerpt of the time series of flight E-AS-03 on 12 March 2018 after take-off until 05:55 UTC. The three regions defined in Fig. 26b are marked with boxes A to C. The inorganic aerosol fragments are displayed on a separate axis to show their smaller values compared to the OA. Negative mass concentrations were set to zero before calculating the aerosol fractions which mainly concerns OA in box B. The axis of f_{44} is reversed to have the aged values on the lower end and the fresher emissions to the top as for the toluene to benzene ratio. The external data are interpolated on the C-ToF-AMS time steps which smooths out maximum values to some extent.

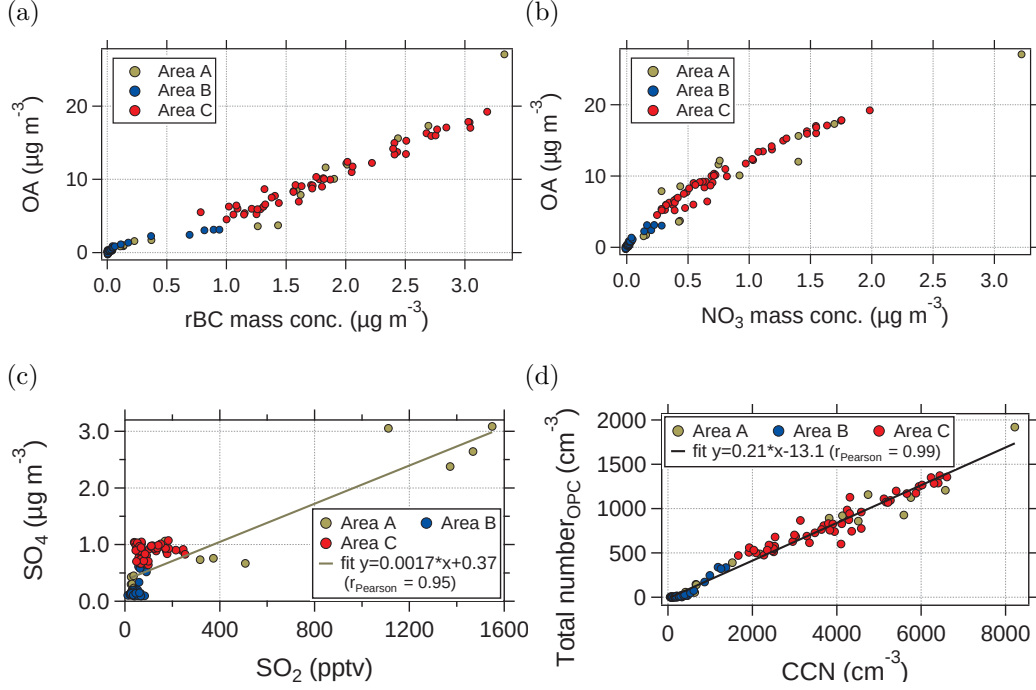


Figure 28: Scatter plots for flight E-AS-03: (a) OA mass concentration versus rBC mass concentration. (b) OA mass concentration versus NO_3 mass concentration. (c) SO_4 mass concentration versus SO_2 VMR including the least squares fit parameters for area A. (d) OPC total number concentration versus CCN number concentration including the fit parameters for all data points. The data are colour-coded to separate the three different areas.

NO_3 and NH_4 with up to $2 \mu\text{g m}^{-3}$, and the SO_4 is around $1 \mu\text{g m}^{-3}$ except for the elevated values directly after take-off (see Fig. 27). SO_4 and SO_2 data are correlating very well during area A with a Pearson correlation coefficient of 0.95 (see Fig. 28c), whereas SO_2 in area C is increasing after 05:30 UTC with rather constant SO_4 mass concentrations. The time series of the other aerosol mass species is similar besides chloride that only shows small concentrations. Moreover, the trace gases go along with the aerosol time series, having higher values for area A and C. The number of CCN is going up to 8220 cm^{-3} in area A and correlates with a Pearson r of almost one with the total number of the OPC that increased up to 1920 cm^{-3} (see Fig. 28d). The aerosol fraction in Fig. 27 explains the changing aerosol composition in the different areas: rBC is higher below 4 km and decreases with altitude. The OA accounts for around 60 % in area A and up to 72 % in area C. The relative contribution of SO_4 becomes more important with increasing altitude in area B while the influence of OA and NO_3 decreases. The composition is subject to strong fluctuations in area B other than in area C where the fractions are stable although absolute values change. The flight path in area A is defined by a continuously increasing altitude and affected by local emissions directly

after take-off. A SO_2 VMR of around 1.6 ppbv is measured in the beginning of flight E-AS-03 which starts around 170 km south-east of Bangkok. In comparison, Kim Oanh et al. (2013) investigated emissions at ground-based stations in Bangkok and observed SO_2 VMRs of 1.9 - 18 ppbv. Furthermore, they found toluene to benzene ratios of up to 2.8 indicating fresh air masses at the source. On the contrary, already aged emissions with a toluene to benzene ratio below 0.5 are encountered for area A during flight E-AS-03 (see Fig. 27) as the emissions are observed in some distance to the origin. The aerosol oxidation marker f_{44} indicates aged aerosol particles as well.

Sresawasdi et al. (2021) mentioned a haze period induced by BB in March and April 2018 over Thailand. This is consistent with the elevated rBC values in Fig. 27. The time series of the OPC volume concentration in Fig. 63a in the appendix indicates that the C-ToF-AMS misses about 1/3 of the aerosol. Herein, the separation of the OPC data at a size of 1 μm gives a non-negligible amount above the C-ToF-AMS measurement range. Dust proxies can be calculated from the OPC volume and number concentration for coarse mode particles greater/equal 0.5 μm (DeMott et al., 2010, 2015; Weger et al., 2018). Figure 63b in the appendix displays the time series of these proxies, correlating with the time series of the other aerosol parameters. Based on the C-ToF-AMS data, it is not possible to differentiate between BB and dust. Another substance not detectable by the C-ToF-AMS but possibly occurring in this size range is sea salt. Sea salt could have influenced the measurements over the Gulf of Thailand after take-off in area A and over the ocean in area C as the altitude is around 1.6 km. This assumption is supported by the fact that NO_3 is the second largest C-ToF-AMS species with up to $2 \mu\text{g m}^{-3}$ over these areas. Nitrate is related to aged sea spray aerosol particles as they can be internally mixed based on reactions with nitric acids (Laskin et al., 2012).

The upper panel of Fig. 27 contains the modelled contributions of the defined target regions (introduced in Sect. 3.5.4), showing an influence from the PRD and YRD in the areas A and C while the YRD emissions are dominating. Beijing and Taipei are also listed although their amounts in area C are too small to appear on the scale. Based on this predictions, the FLEXTRA backward trajectories are separated for the three different periods and are depicted in Fig. 29. The measurements in area A are determined by air masses circling around Bangkok for 240 h (length of the backward calculations) in about 4 km height. Furthermore, five trajectories indicate air masses coming from South Korea accumulating emissions from the YRD and PRD along their way below 2 km. The modelled mean ages are 80 - 110 h and 140 - 170 h for the PRD and YRD emissions, respectively. The lower aerosol mass concentrations and trace gas VMRs in area B are in accordance with the backward trajectories coming from east over the cleaner ocean where they started at altitudes of almost 8 km. Only a few trajectories reached altitudes below 2 km but stayed the whole 240 h over the sea. For

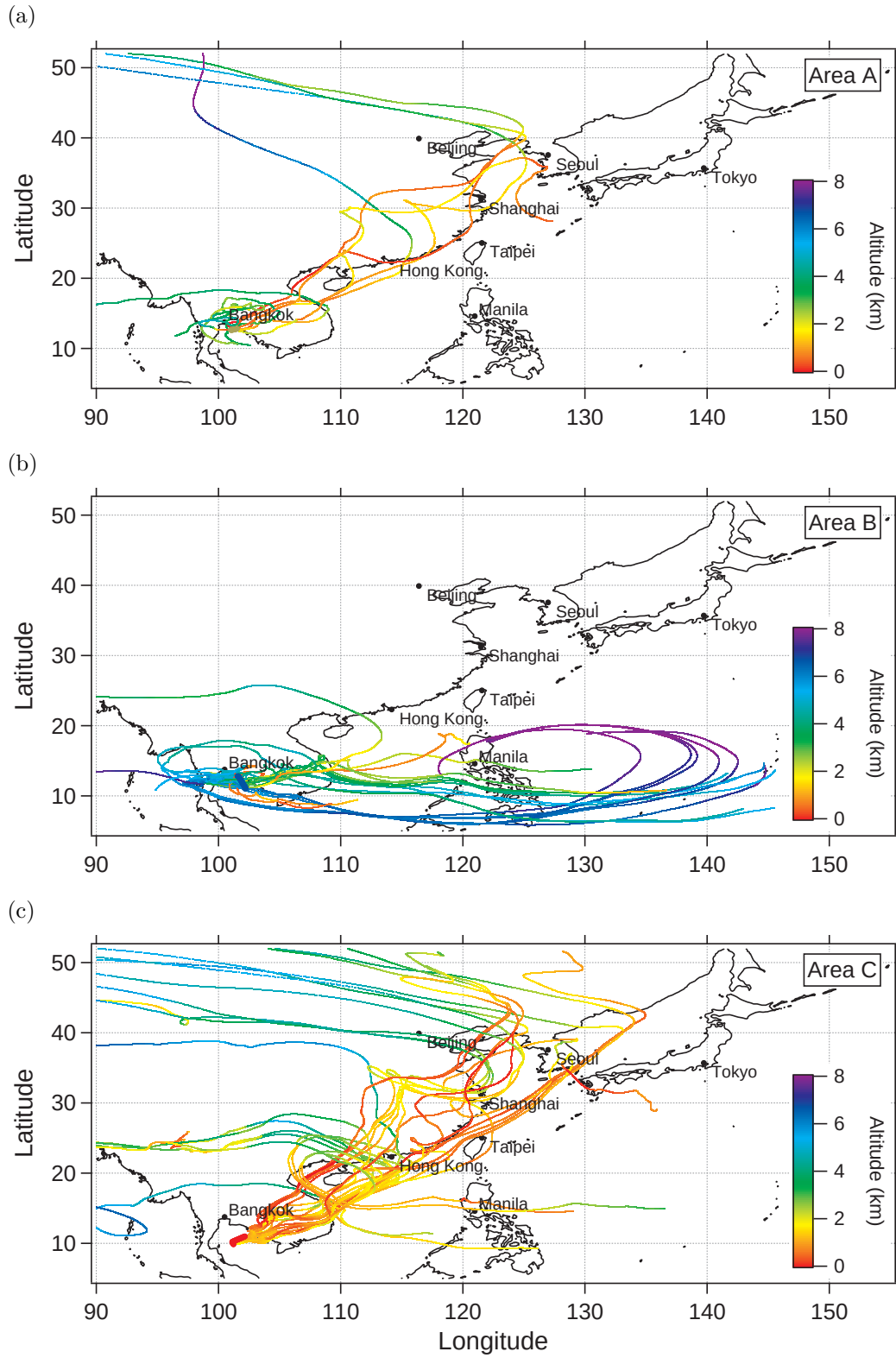


Figure 29: 240 h FLEXTRA backward trajectories starting at the in Fig. 26b defined flight segments A to C of flight E-AS-03. They are colour-coded with altitude.

area C, the main air mass history pattern consists of trajectories remaining below 2 km. They come from west and descend before turning south over the YRD and South Korea. From there the air masses stay low and accumulate emissions on their way to Thailand. Some trajectories circle in the vicinity of the PRD over South China. Furthermore, the trajectories pass Beijing and Taipei which is further indicated by the tagged CO modelling. The modelled mean ages are 80 - 100 h, 130 - 150 h, 170 h, and 100 - 140 h for the PRD, YRD, Beijing, and Taipei emissions, respectively.

Förster et al. (2023) calculated source region contributions based on the FLEXTRA backward trajectories and a CO emission data base, also described in Sect. 3.5.4. These results quantify the air mass history and reveal Southern China and South-East Asia as the main contributors in area A, followed by emissions from East China. The fraction attributed to Bangkok is only obtained during the first minute of flight E-AS-03 and is 3.85 % while the whole region of South-East Asia accounts for 27 %. In area B, half of the data points contain no signals in relation to any source region and the other half gives only minor values from South-East Asia. Area C is comprised of South-East Asian emissions, followed by East China (including the YRD) and Southern China (including the PRD). Furthermore, India is another smaller contributor in area C as some trajectories travel west around 25° N (see Fig. 29c).

Finally, the influence of BB on the selected case of flight E-AS-03 is investigated. Lin et al. (2023) examined a BB plume originating in Indochina and compared the measurements of EMeRGe with model output. The results gave an BB effect of up to 20 % for altitudes below 1 km and 30 - 80 % at 2 - 4 km in the area of Southern China, Taiwan, and the East China Sea. As introduced in Sect. 3.6.4, m/z 60 is a marker within the organic mass spectrum to quantify influence and ageing of BB. The triangle plot in Fig. 30a contrasts the ratios f_{44} and f_{60} for the three areas. The data points of area B show a wide spread and scatter around the atmospheric background indicated by the dashed line. Some data of area B are even intruding in the triangle indicating BB but they show no clear evolution and are triggered by single events. On the contrary, the data points of the areas A and C align along the same line indicating an evolution of the organic aerosol and an already aged state as they have high f_{44} values. Although the points are placed outside of the triangle, they still can be considered as BB influenced as the f_{60} values correlate well with clearly increased values of rBC and acetonitrile (see Fig. 30b and c). The acetonitrile threshold of 199 pptv is exceeded for all points in area A and C. Besides some scattered background points, the f_{60} of the whole EMeRGe data set only rarely enters the range of the triangle region. This might be due to already degraded signal at the m/z 60, the measurement of mostly aged BB plumes, or a differing instrument characteristic. Förster et al. (2023) estimated BB fractions of 2 and

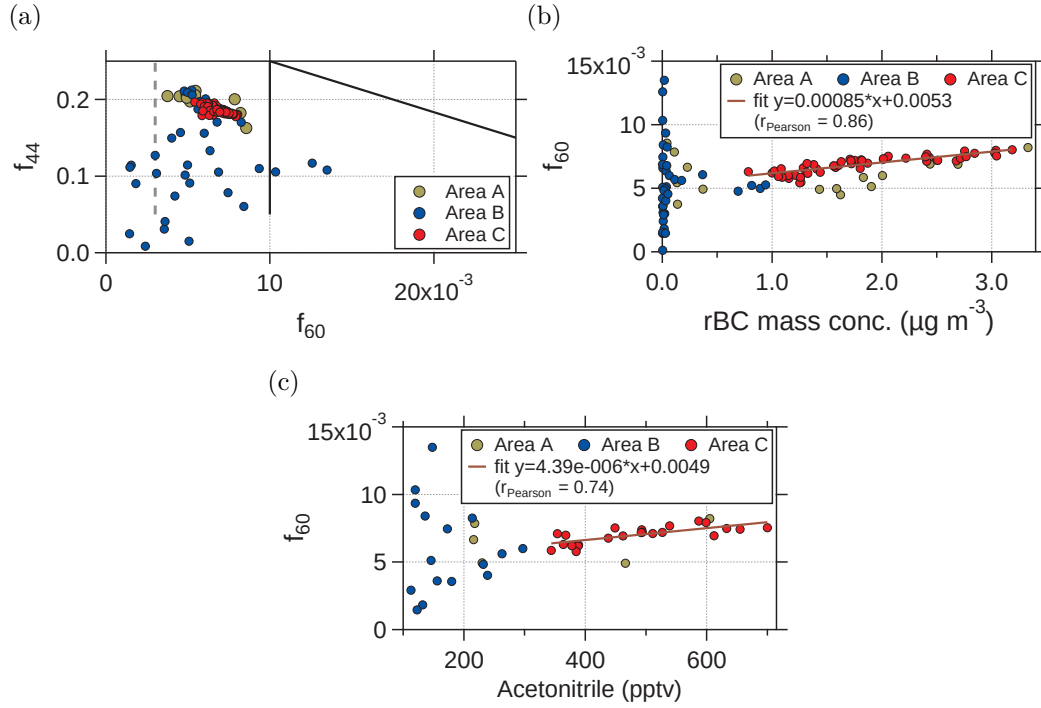


Figure 30: BB influence on flight E-AS-03: (a) Ageing graph of the C-ToF-AMS BB marker f_{60} after Cubison et al. (2011). (b) Scatter plot of f_{60} versus BC mass concentration. (c) Scatter plot of f_{60} versus the acetonitrile mass concentration containing less data points due to missing VOC data. The colour-code separates the three different areas A to C defined in Fig. 26b. (b) and (c) further include the least squares line fit parameters for area C.

16 % for EMeRGe-Europe and -Asia, respectively. They further stated the detection of aged BB in connection with long-range transport of emissions during flight E-AS-03.

In conclusion, anthropogenic emissions could be identified by high CO VMR in the Gulf of Thailand. They were generated by local sources or mixed with long-range transported air masses from East China and South Korea. The aerosol composition changed in relation to the different sources, especially in the beginning of the flight where the local contributions were pronounced but also an effect of the changing altitude was observed. Nevertheless, OA is correlated with CO as well as rBC and NO₃. Additionally, an influence of BB from Indochina could be observed in the rBC mass concentrations and the acetonitrile VMR. On the contrary, the BB marker f_{60} in the C-ToF-AMS data could only identify a weaker signal although an evolution to aged aerosol particles was detected.

4.1.5 *Aerosol composition of selected MPCs in Europe and East Asia*

This section addresses the question of whether individual MPCs show a characteristic composition. Therefore, the results from a simple emission uptake scheme according to Förster et al. (2023) are used. As explained in Sect. 3.5.4, the 10-day FLEXTRA backward trajectories are combined with the EDGAR CO emission inventory. The source region-specific CO is accumulated if the trajectory is in the BL above a specified area which could be a broad region or a single MPC. This section focuses on eight MPCs for Europe and East Asia. The CO emission uptakes are interpolated on the C-ToF-AMS time and utilised to filter the aerosol mass concentration data including rBC for each individual MPC. Furthermore, the data for each MPC are weighted by the fraction to the total modelled CO.

The influence of one MPC is observed in the immediate vicinity of the source as well as further away as the air masses are transported over greater distances. For instance, emissions originating in the MPCs BeNeRu or London are sporadically measured also in the Mediterranean (see Fig. 31a and b) or pollutants coming from Manila have been transported westwards to an area near Bangkok (see Fig. 32b). On the contrary, Munich and PoV emissions remained almost local (see Fig. 31d and Fig. 64b in the appendix). Possible explanations could be a reduced dilution of the emissions due to weaker winds in combination with the adjacent mountains which hinder the transport or strong inversions which occur frequently in the area (Caserini et al., 2017). The pollutants from Barcelona and Rome are measured mostly east downwind of their sources (see Fig. 64a and d in the appendix). Taipei emissions stayed close to their source as well and spread along the west coast of Taiwan (see Fig. 32c). Contributions from Madrid are highest over Spain but are also detected around Italy (see Fig. 31c). The most scattered distri-

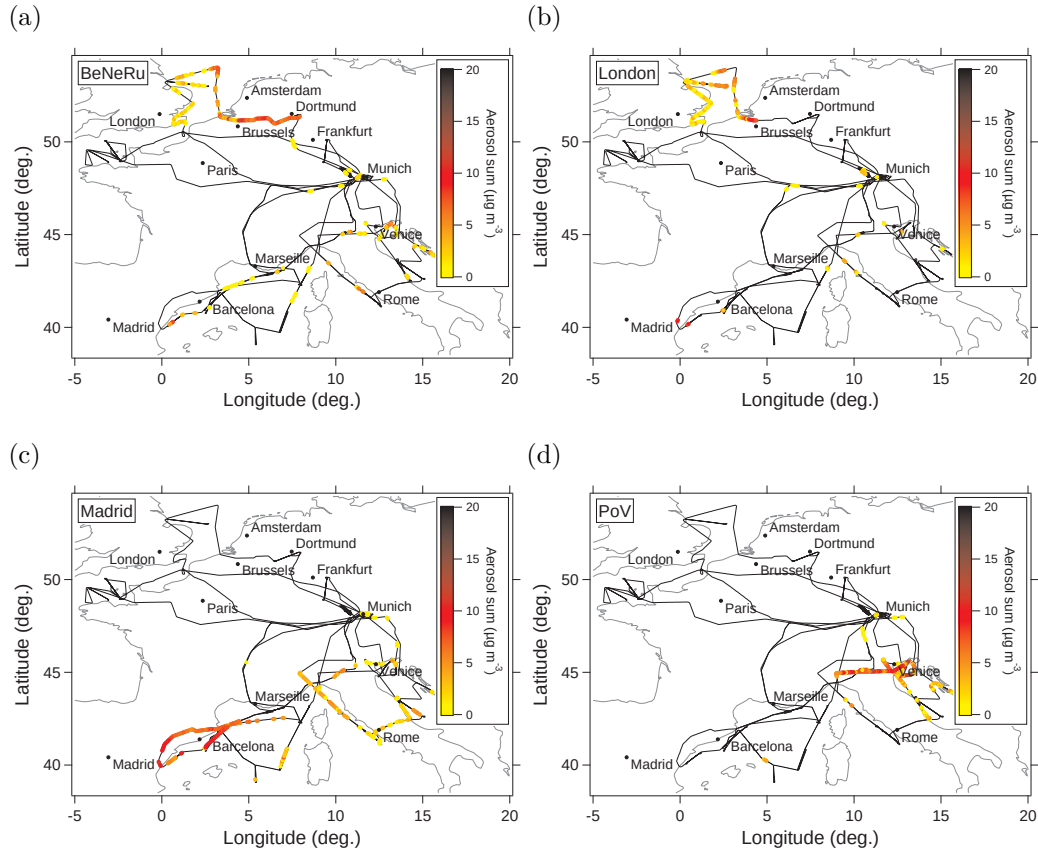


Figure 31: Flight paths of the EMerGe-Europe campaign colour-coded with the total aerosol mass concentration including rBC and filtered for emission contributions from different MPCs: BeNeRu (a), London (b), Madrid (c), and PoV (d).

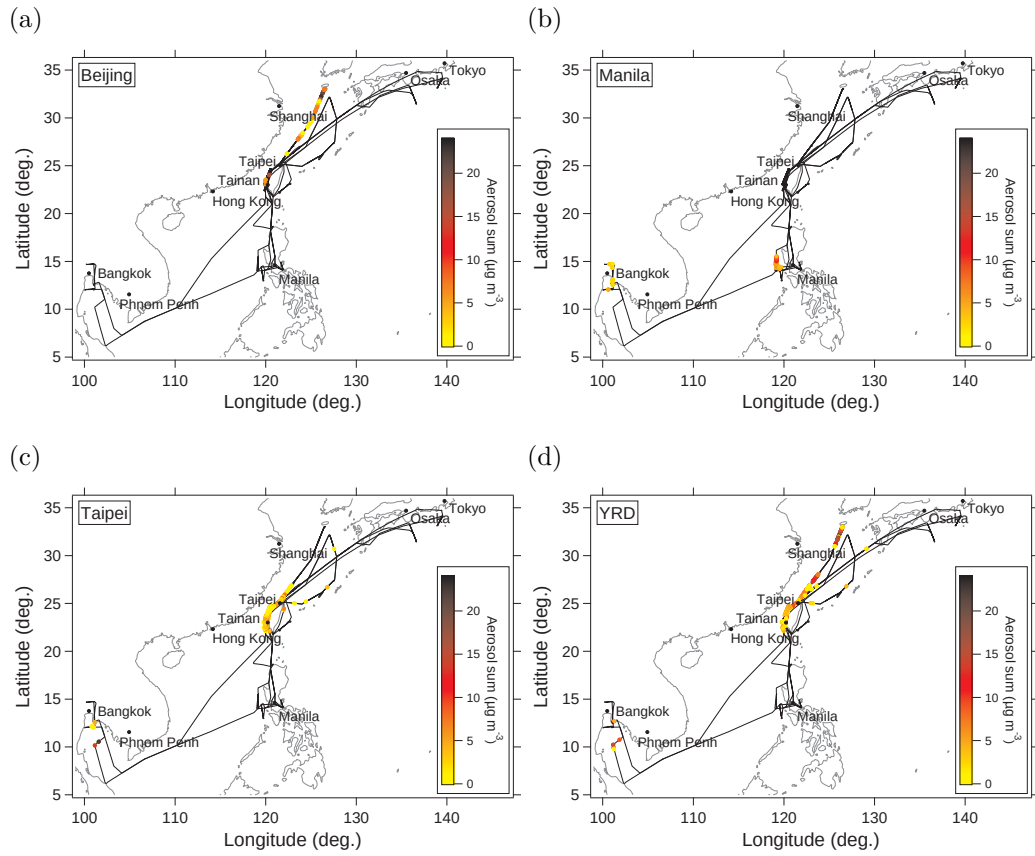


Figure 32: Flight paths of the EMerGe-Asia campaign colour-coded with the total aerosol mass concentration including rBC and filtered for emission contributions from different MPCs: Beijing (a), Manila (b), Taipei (c), and YRD (d).

bution occurred for Paris (see Fig. 64c in the appendix). However, the least contributions along the flight tracks with only a few data points arose for the East Asian MPCs Bangkok, Osaka, the PRD, and Tokyo (see Fig. 65 in the appendix). According to Förster et al. (2023), a fraction of less than 0.3 % each is attributed to those MPCs with regard to the sum of all modelled CO contribution potentials including defined source regions in Eurasia and Northern Africa. Interestingly, the emissions from Beijing are only measured during three flights (E-AS-07 to E-AS-09) which covered the same flight track closest to the east coast of China. Although there had been two other flights (E-AS-04 and E-AS-05), where the flight path is shifted further east, showing no influence from Beijing (see Fig. 32a). Some areas experience a mixture of emissions coming from different sources, e.g. London and BeNeRu pollutants mix along the flight paths north of 51° N while on the south-east coast of Great Britain also emissions from Paris interfere (see Fig. 31a and b and Fig. 64c in the appendix). Another example for different source region contributions at one place is the area west of Taiwan where emissions from Taipei and the YRD mix with smaller contributions from Beijing, Osaka, the PRD, and Tokyo (see Fig. 32 and Fig. 65 in the appendix).

The averaged aerosol compositions differ between the individual MPCs as well as between Europe and East Asia. In general, the PM_1 aerosol mass concentrations of selected MPCs are higher in East Asia with up to $7.3 \mu\text{g m}^{-3}$ for Beijing and YRD emissions whereas a maximum of $6 \mu\text{g m}^{-3}$ is measured for the PoV during EMeRGe-Europe (see Fig. 33a and c). On the contrary, the smallest concentrations are $1.5 \mu\text{g m}^{-3}$ for Paris in Europe and $2.5 \mu\text{g m}^{-3}$ for the PRD in Asia. During EMeRGe-Europe, the Southern MPCs Barcelona, Rome, Madrid, and the PoV have the greatest total mass concentrations with $3.5 - 6 \mu\text{g m}^{-3}$ while the former two which are located on the coast show lower mass concentrations (see Fig. 33a). This might be due to a reduced pollutant accumulation or faster mixing with cleaner ocean air masses. The Northern MPCs London and the BeNeRu follow in the pollution ranking with total mass concentrations between 3 and $3.5 \mu\text{g m}^{-3}$ while the Central European MPCs Munich and Paris achieve maximum 2 and $1.5 \mu\text{g m}^{-3}$ (see Fig. 33a). The meridional differentiation is also reflected in the NO_3 distribution and the corresponding NO_3 fraction which increases from south to north from around 4 % in Madrid to 22 % in the BeNeRu whereas the OA fraction decreases from 72 to 39 %, respectively (see Fig. 33b). The SO_4 proportion ranges between 12 % in Madrid and around 25 % in Barcelona while the NH_4 share is minimum with 7 % in Madrid and maximum around 15 % in the BeNeRu. Possible explanations could be different landscape characteristics as well as the extent of agricultural emissions (Rega et al., 2020; Dou et al., 2021). The amount of rBC differs slightly between the MPCs and has a maximum in Rome with around 5 % whereas chloride exceeds 1 % only for the BeNeRu and Paris (see Fig. 33b).

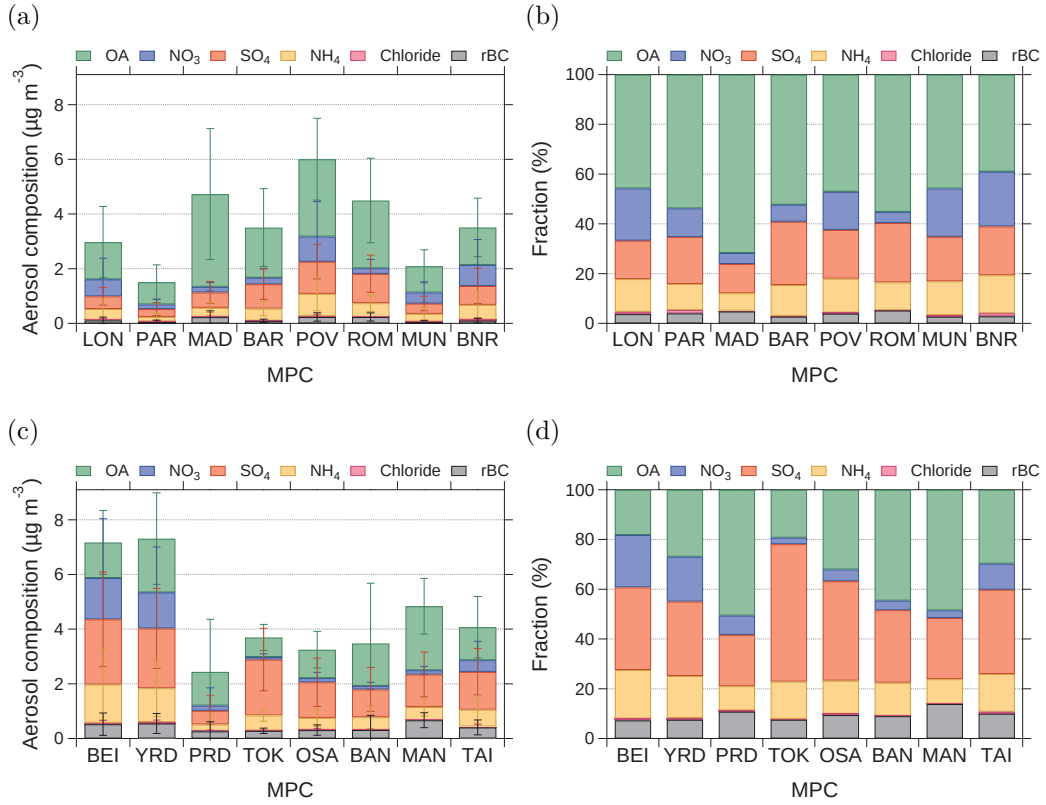


Figure 33: Average aerosol composition including rBC for eight different MPCs in Europe (a) and East Asia (c). Error bars are the standard deviation of the mean value. The y-axes are aligned for both campaigns to make the bars easily comparable. The relative compositions are depicted in (b) and (d). Abbreviations: London (LON), Paris (PAR), Madrid (MAD), Barcelona (BAR), Po Valley (POV), Rome (ROM), Munich (MUN), Belgium-Netherlands-Ruhr area (BNR), Beijing (BEI), Yangtze River Delta (YRD), Pearl River Delta (PRD), Tokyo (TOK), Osaka (OSA), Bangkok (BAN), Manila (MAN), and Taipei (TAI). The relative shares of each bar are listed in Table 24 in the appendix.

During EMeRGe-Asia, Tokyo is among the four MPCs with the lowest total mass concentrations with values smaller than $3.7 \mu\text{g m}^{-3}$ (see Fig. 33c) although it is designated as the largest MPC in the world by number of inhabitants (United Nations, 2019). The other three MPCs with lower total mass concentrations are Bangkok, Osaka, and the PRD. As stated earlier, they all are probed rarely which might lead to a sampling bias of possibly cleaner conditions than usual. Furthermore, the encountered air masses of these four MPCs are measured in a greater distance from their sources than the other East Asian MPCs which also caused enhanced dilution and mixing with cleaner air masses over the ocean (see Fig. 65 in the appendix). Moreover, wet scavenging of aerosol particles could have reduced the concentrations during transport. Beijing and YRD emissions have almost similar total mass concentrations with up to $7.3 \mu\text{g m}^{-3}$ (see Fig. 33c) although the relative contributions of the individual aerosol species differ (see Fig. 33d). More precisely, rBC and chloride values are comparable whereas NH_4 , NO_3 , and SO_4 fractions are slightly higher by around 3 % in Beijing than the YRD. In contrast, the OA proportion is higher in the YRD with around 27 % (see Fig. 33d). In general, the OA fraction increases from north to south with decreasing distance to the tropics while the SO_4 amount decreases. Compared to the European MPCs, where OA is the main aerosol component, SO_4 is the dominant species in East Asia. The relative contributions of NH_4 , NO_3 , and chloride are comparable between the two regions whereas the rBC fractions in East Asian MPCs exceed the European values by more than 2.5 (see Fig. 33b and d).

In conclusion, applying the simple CO uptake mechanism to the aerosol data, revealed characteristic differences between the individual MPCs in Europe and East Asia. The total aerosol mass concentrations vary for the single MPCs and are generally higher in East Asia. Furthermore, the aerosol compositions differ between the regions with a change of the main aerosol component as well as meridional differentiations.

4.1.6 *Comparison with literature*

In a study by Morgan et al. (2010), fractions of 20 - 50 % for OA and 10 - 30 % for SO_4 were found under both background and highly polluted conditions in Northern Europe in April, May, and September 2008. The airborne measurement campaign covered an area above Great Britain, Germany, and the Baltic Sea within an altitude range of up to 3 km. Their findings are comparable to EMeRGe-Europe in summer 2017. Their OA concentrations were typically 1.3 to 2.5 times higher than that of SO_4 with $1 - 4 \mu\text{g m}^{-3}$. During EMeRGe-Europe, the OA is on average 2.2 to 3 times higher than SO_4 . Furthermore, Morgan et al. (2010) reported spatial gradients for the NO_3 mass concentrations with elevated values in North-Western Europe compared to

further east. This is in line with the EMeRGe-Europe data except for the high values in the PoV but this area was not included in their measurements. In 2008, NO_3 contributed with 20 to 50 % to the aerosol composition depending on the location (Morgan et al., 2010) while we measured a mean of up to 27 % in polluted areas in 2017. Both studies confirm NO_3 as a contributor to highly polluted conditions as the NO_3 contribution is increasing with the total mass.

Zhou et al. (2020) reviewed the average aerosol chemical composition of ground-based measurements in Asia (i.e., China, Korea, Japan, India, and Singapore) from 2001 to 2019. The mass loadings varied regionally with the greatest concentrations in India, the North China Plain, and North-Western China ($60.2 - 81.3 \mu\text{g m}^{-3}$), while much lower values of 9.1 to $15.1 \mu\text{g m}^{-3}$ occurred in Korea, Japan, and at the background sites. The aerosol composition in East Asia was overall determined by OA accounting for 32 - 50 %. Followed by sulfate as the second largest aerosol component, dominating the inorganic aerosol in the PRD, Japan, and the Korean suburban sites with 24 - 47 %. On the contrary, sulfate and nitrate fractions were comparable with around 17 to 23 % in urban and suburban regions in Northern China. The contribution of ammonium was relatively similar over East Asia, represented by around 13 %. The chloride proportion was generally small, making up less than 2 % in most regions. Although the data reviewed by Zhou et al. (2020) included ground-based measurements, they provide insights on the possible composition close to the source. However, HALO flew in larger distance to the emission sources. Thus, the composition might have changed due to secondary aerosol formation, wet scavenging, or mixing with long-range transported air masses.

Nault et al. (2021) listed the average aerosol composition of airborne measurements in urban and urban outflow regions around the world from different campaigns between 2000 and 2016. Although the relative contributions differ by some percent for the individual locations, the results are in general in agreement with the EMeRGe data. For instance, Nault et al. (2021) found comparable compositions for Paris and London with a greater NO_3 fraction for London. Moreover, they also observed higher NO_3 shares in air masses from Beijing and the Chinese outflow than for Tokyo. In general, OA was the dominant aerosol component in Europe while SO_4 and NO_3 became more important in North-East Asia (Nault et al., 2021). In addition, NH_4 was a few percent higher in North-East Asia than in North-Western Europe which was also observed during EMeRGe.

4.2 ACIDITY OF AEROSOL PARTICLES

The acidity of atmospheric aerosol particles can be evaluated by comparing the measured ammonium ($\text{NH}_{4,\text{meas}}$) with a calculated ammonium mass con-

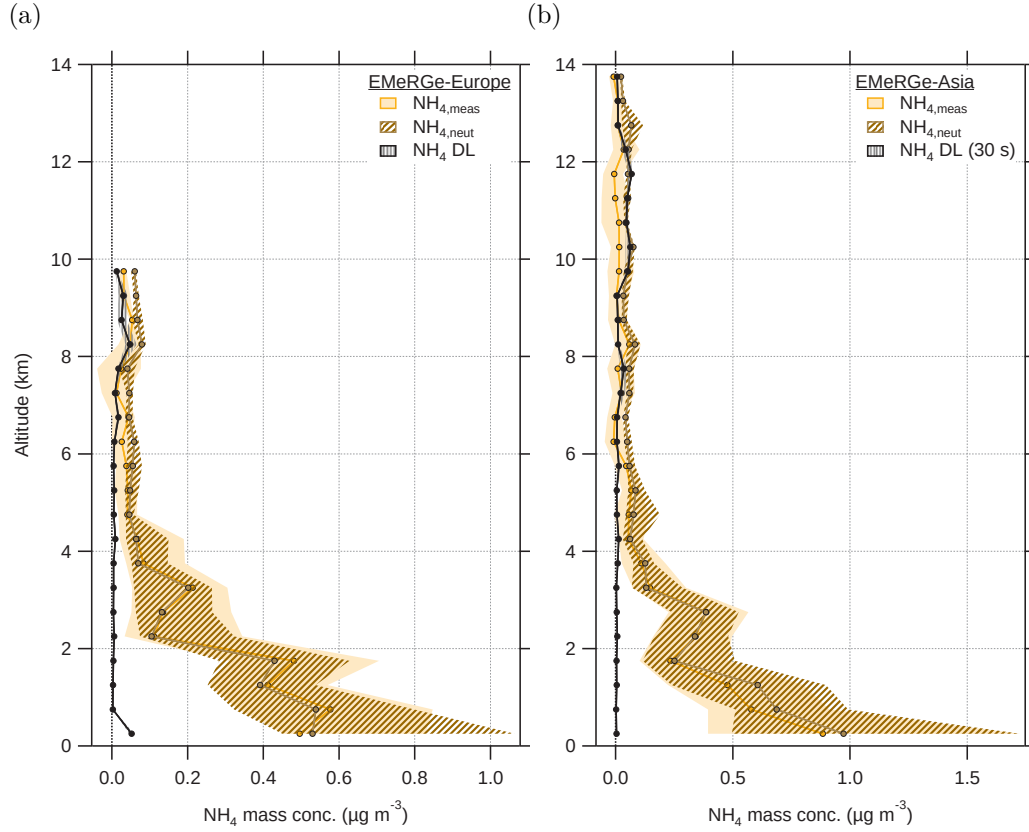


Figure 34: Averaged vertical profiles of the measured NH_4 mass concentration $\text{NH}_{4,\text{meas}}$, corresponding DL, and $\text{NH}_{4,\text{neut}}$ needed for neutralisation of the EMeRGe-Europe (a) and -Asia (b; flights E-AS-03 to E-AS-14; 30 s DL) campaigns in 500 m altitude bins. The dots represent the medians and the shaded areas the interquartile ranges.

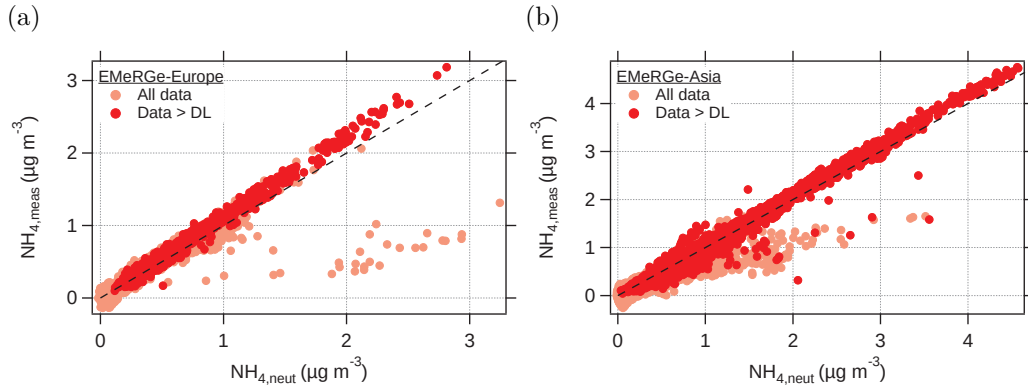


Figure 35: Scatter plots of the measured NH_4 mass concentration $\text{NH}_{4,\text{meas}}$ versus the $\text{NH}_{4,\text{neut}}$ needed for neutralisation of the EMeRGe-Europe (a) and -Asia (b; flights E-AS-03 to E-AS-14) campaigns. The dashed lines are the 1:1 lines. The light red colour shows all data while for the points coloured in darker red the data is filtered for values below the inorganic species' DLs.

centration needed for neutralisation ($\text{NH}_{4,\text{neut}}$) using the inorganic species of the C-ToF-AMS data, as already explained in Sect. 3.6.5. A decreasing amount of NH_4 with altitude is shown in the median vertical profiles for both EMeRGe campaigns (see Fig. 34). The measured NH_4 values are larger than the DL for altitudes below 7 km and 6 km for EMeRGe-Europe and -Asia, respectively. Up to these altitudes, the measured concentrations are comparable to the ammonium needed for neutralisation, showing that aerosol particles on average are neutralised. Airborne measurements by Morgan et al. (2010) found that the aerosol was neutralised in general. They measured over Northern Europe up to an altitude of 3 km in April, May and September 2008. During EMeRGe-Asia, an exception to the neutralised particles are the data below 1.75 km (see Fig. 34b) where the ratio shifts towards acidic conditions due to increased SO_4 concentrations as measured with the C-ToF-AMS. Furthermore, the aerosol particles tend to be more acidic in the upper troposphere and lower stratosphere due to small NH_4 mass concentrations and higher SO_4 partly induced by the injection of volcanic emissions. These findings are further visualised in the vertical cross sections along the latitude in the appendix (Figs. 66 and 67).

The scatter plots in Fig. 35 compare the measured with the calculated ammonium needed for neutralisation for both campaigns. The concentrations coloured in dark red are filtered for the inorganic DLs before performing the calculation and the ammonium DL is applied to $\text{NH}_{4,\text{meas}}$ as well. Chloride is the aerosol species affecting the DL filter the most because the concentrations are often close to or below the DL. However, up to 21 % of the points exceed the DL mainly in altitudes below 5.5 km, therefore, chloride is included. Most data points arrange along the 1:1 line or slightly above indi-

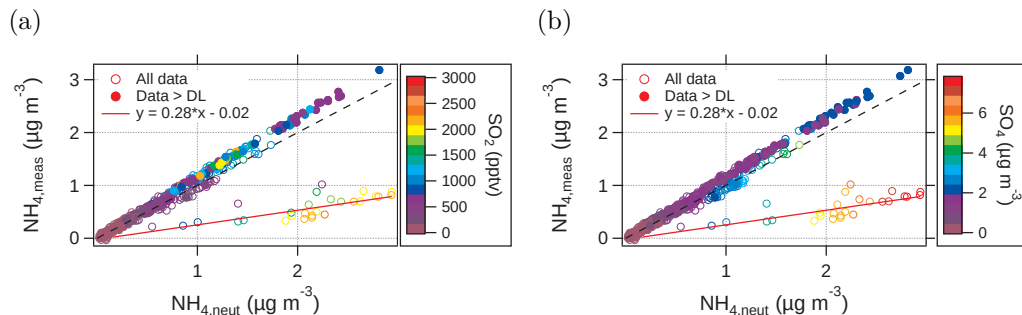


Figure 36: Scatter plots of the $\text{NH}_{4,\text{meas}}$ versus $\text{NH}_{4,\text{neut}}$ mass concentrations for flight E-EU-06 on 20 July 2017 colour-coded with SO_2 VMR (a) and SO_4 mass concentration (b). The dashed lines are the 1:1 lines indicating neutralised aerosol particles. The open circles show all data while the data of the closed circles are filtered for values below the DLs of the inorganic species. The SO_2 data points are interpolated to the C-ToF-AMS time and show less data points because of measurement gaps. The fit function is a least squares fit through the points of the event.

cating in general neutralised aerosol particles. This is in accordance with the results from Fig. 34. Furthermore, the measurements in East Asia show more acidic aerosol particles as the ratio is often below one. The acidic data for EMeRGe-Europe arise mainly from flight E-EU-06 and is further discussed in the following as well as two examples for flight E-AS-04 and E-AS-07 from EMeRGe-Asia. Table 25 in the appendix gives the linear fit values for the scatter plots of every flight, indicating that conditions favouring acidic aerosol particles occurred more often during EMeRGe-Asia.

Averaged vertical profiles and scatter plots including the whole campaign data can only give a rough overview of the conditions. Three examples from single flights are presented in the following to get a more detailed view about the acidic aerosol measurements. It becomes evident that acidity and composition of aerosol particles is not only influenced by anthropogenic MPC sources but also by other factors like volcanic eruptions, ship emissions or industrial pollution. Furthermore, a mixture of different origins prevails most of the time and more than one source needs to be considered at once.

4.2.1 Acidic aerosol particles due to volcanic emissions

During flight E-EU-06 on 20 July 2017, an event clearly deviates from the 1:1 line indicating neutralised aerosol particles with a mean acidity ratio of 0.28 (see linear fit in Fig. 36). SO_2 and SO_4 are elevated with up to 2393 pptv and $7.8 \mu\text{g m}^{-3}$, respectively. The data points of interest are marked as open circles because chloride is below the DL and also NO_3 is mostly below its DL. The flight is going over the Alps, crossing the Po Valley, turning south

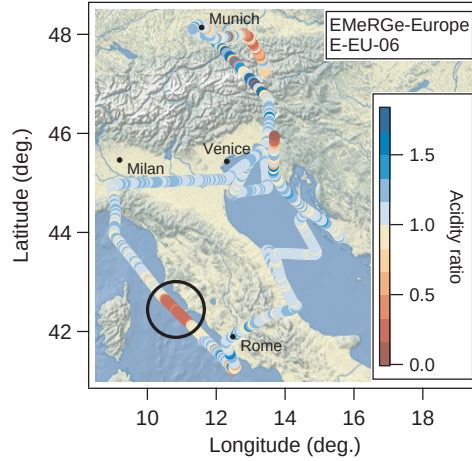


Figure 37: Map of the flight track of flight E-EU-06 colour-coded with the acidity ratio. The area of the acidic values is circled in black.

towards Rome, and heading back home over the Croatian coast (see Fig. 37). The strong acidic values are measured north-west of Rome over the Tyrrhenian Sea and occurred at an approximate altitude of 1000 m.

The acidic values are measured between 12:48:00 and 13:01:30 UTC as depicted by the black box in the time series of the measured variable (see Fig. 38). Values of OA, NO₃, rBC, CCN, and CO drop in comparison to before and after the event. The OA mass concentration is highly correlated with the CO mixing ratio, indicating that under summertime conditions in Southern Europe anthropogenic and combustion-related emissions are mainly influencing the OA mass. The concentration of SO₄ is less correlated with CO, but instead strongly to SO₂, especially around 12:55 UTC where both substances show a large increase. The f_{44} increases by 20 % to values around 0.24 during the period of acidic aerosol indicating OA oxidation (not shown). The tagged CO from the HYSPLIT dispersion model calculations, introduced in Sect. 3.5.4, shows no remarkable signals from the selected target regions, only a little influence from Rome appears. This is confirmed by one backward trajectory passing over the city as depicted in Fig. 39.

It is very likely that volcanic outgassing is the source of this event owing to several reasons. First, only particulate sulfate and SO₂ are increased during this short plume while other trace gases remained low or even decreased (see Fig. 38). Second, 31 % of the backward trajectories show a passing over Mount Etna on the east coast of Sicily in Italy (see Fig. 39). Mount Etna is one of the tallest active volcanoes in Europe with 3357 m height and showed strombolian activities (explosions, ash emissions, lava flows) occasionally from end of January till May 2017 (Global Volcanism Program, 2017). Third, SO₂ satellite data (OMPS, NASA EOSDIS Worldview) show a small eruption around 18 and 19 July 2017 and the emissions travelled to the south (not shown). Furthermore, for the trajectories filtered by their last contact

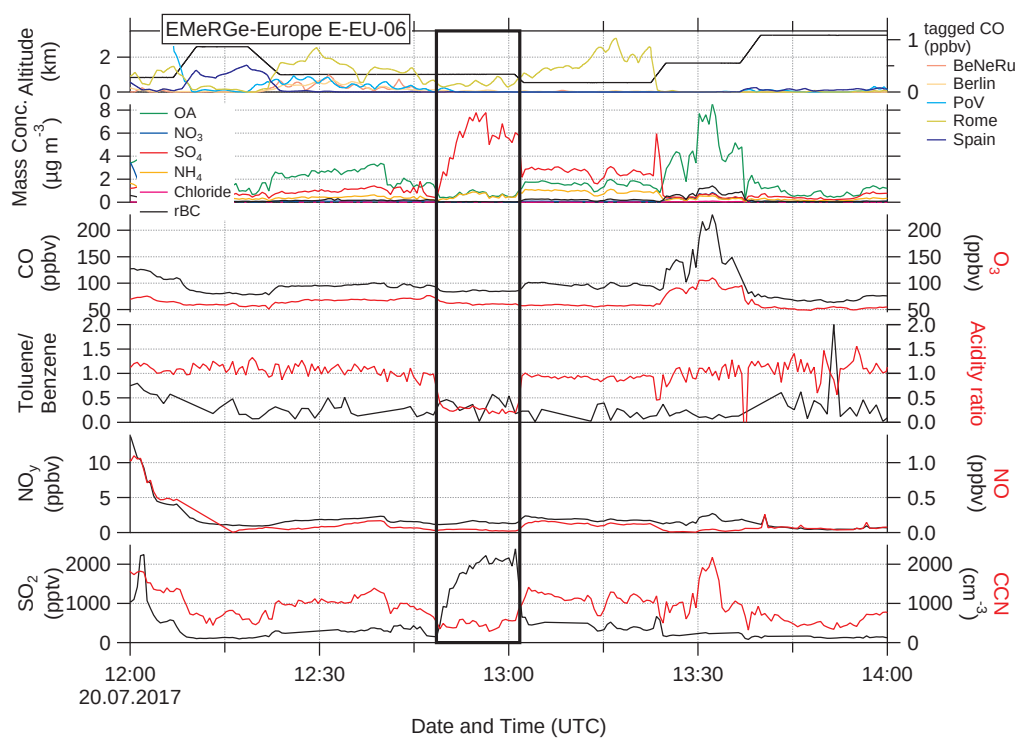


Figure 38: Excerpt of the time series of flight E-EU-06 around one hour before and after the acidic values are measured marked with the black box. The external data are interpolated on the C-ToF-AMS time which smooths out maximum values to some extent.

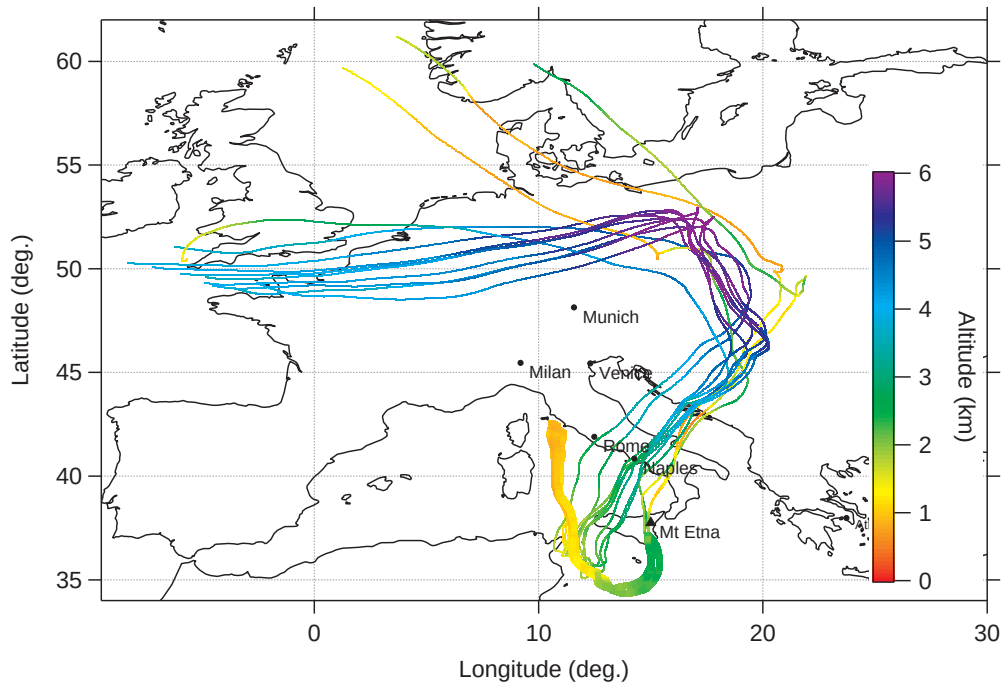


Figure 39: 192 h FLEXTRA backward trajectories starting at the flight segment comprised of acidic values during flight E-EU-06. They are colour-coded with altitude. The beginning of the thicker lines south of Italy indicate the location where the trajectories have their last BL contact. From there, they travel in the free troposphere to the measurement location.

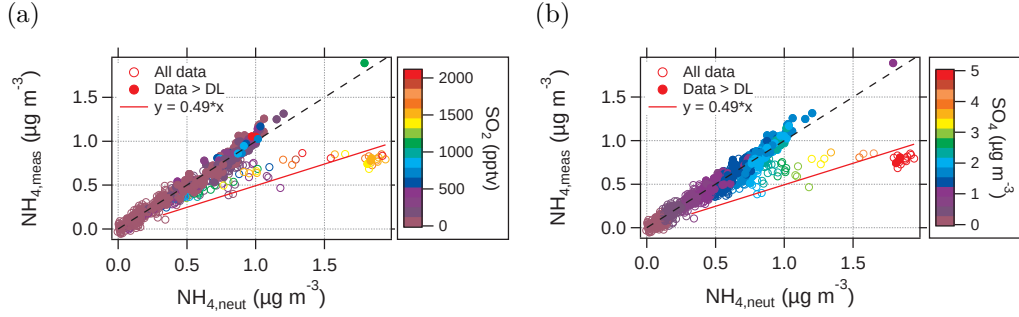


Figure 40: Scatter plots of the $\text{NH}_{4,\text{meas}}$ versus $\text{NH}_{4,\text{neut}}$ mass concentrations for flight E-AS-04 on 17 March 2018 colour-coded with SO_2 VMR (a) and SO_4 mass concentration (b). The dashed lines are the 1:1 lines indicating neutralised aerosol particles. The open circles show all data while the data of the closed circles are filtered for values below the DLs of the inorganic species. The SO_2 data points are interpolated to the C-ToF-AMS time and show less data points because of measurement gaps. The fit function is a least squares fit through the points of the event.

with the BL, just the ones going over Mt. Etna remain (see Fig. 39). They start within the BL close to Mt. Etna and travel around 78 h in the free troposphere to the measurement location. Here, the model uncertainties in calculating the BL height need to be considered. Seidel et al. (2012) reported uncertainties of 50 % for shallow BLs below 1 km, e.g. at night, and values below 20 % for deeper boundary layers at daytime. To sum up, the decreased acidity ratio in combination with increased SO_4 and SO_2 values result from the emissions of the volcano Mount Etna. The conversion of SO_2 to sulfate aerosol in the lower troposphere can be investigated using case studies like this. The method is described in Sect. 3.6.6 and the estimated timescales including also the following two examples are presented in Sect. 4.2.4.

4.2.2 Acidic aerosol particles due to mixed volcanic and anthropogenic emissions

The second example of an acidic period is taken from flight E-AS-04 on 17 March 2018. The mean acidic ratio is 0.49 given by the linear fit in Fig. 40. The SO_4 mass concentration is clearly correlated with acidity reaching values up to $5.1 \mu\text{g m}^{-3}$. The SO_2 VMR also shows an increase up to 2342 pptv for more acidic values but is more scattered than for the first example. Most of the acidic data points are indicated as below the DL because of chloride lying below its DL. The acidic values are measured in the Northern part of the flight over the East China Sea close to South Korea and Japan as depicted in Fig. 41. The flight section is part of a shuttle covering the same area in

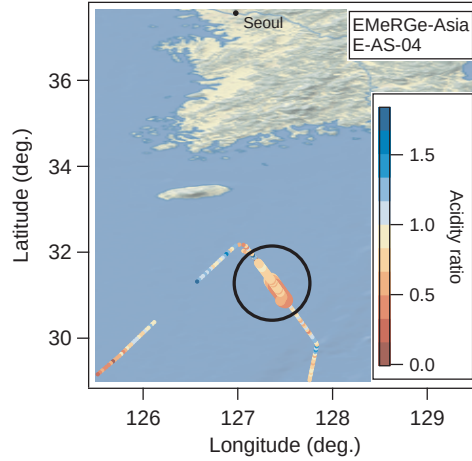


Figure 41: Map of the northern part of the flight track of flight E-AS-04 colour-coded with the acidity ratio. The area of the acidic values is circled in black. Data points of the higher flight altitude (920 m) are displayed with thicker points to be visible below the overlaying flight track in the lower altitude (315 m).

three different altitudes, namely 3200, 920, and 315 m. The former is omitted because the aerosol particles are neutralised there.

The acidic values occurred between 05:47:00 and 06:13:30 UTC as indicated by the black box in the time series (see Fig. 42). Generally, the aerosol mass concentrations, CO, and SO₂ VMR are higher at 920 m while NO, NO_y, and the CCN are increased at the lower level (315 m) showing also some smaller spikes there. The toluene to benzene ratio indicates already aged emissions with values below 0.5. The acidity ratio is lower at the higher altitude indicating more acidic particles (around 0.4 compared to around 0.7 at the lower flight level).

The measured event of acidic particles is influenced by emissions from MPCs, ships and volcanic eruptions as indicated by the following reasons. First, an influence of Beijing and the YRD is observed in the tagged CO plumes, and a very small signal from Seoul for the time before 06:00 UTC as well (see Fig. 42). Second, during the three days before the measurements, two trajectories show air masses coming from East China circling over land and the coast and two trajectories circle the volcano group Kirishimayama in Japan. In addition, three trajectories come from China and Russia and continue over North and South Korea to the measurement location. Most of the trajectories travel over Russia where they decrease in altitude to below 2 km before reaching the Sea of Japan. From there they take around 35 h to the measurement location and pass Fukuoka 10 h before the observation (see Fig. 43). The FLEXTRA trajectories explain the tagged CO signal coming from Beijing, the YRD, and Seoul, giving mean travelling times of 60, 72 and 34 h, respectively. Third, the Global Volcanism Program (2018a,b) states activities at the Shinmoedake (Shinmoe peak) with a maximum volcanic

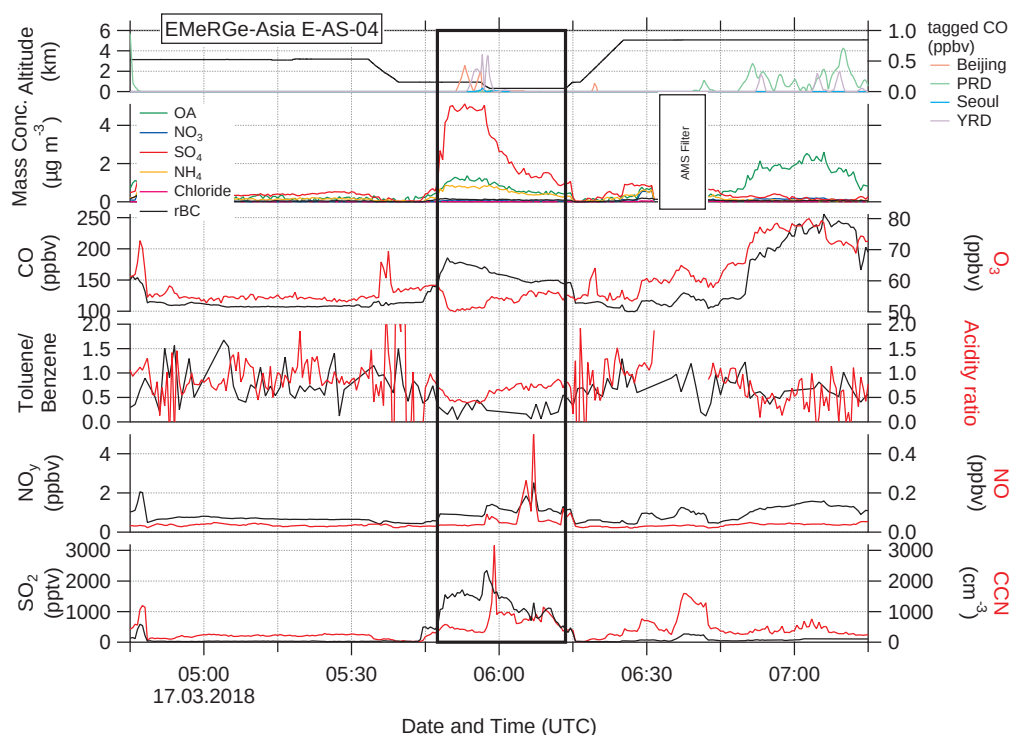


Figure 42: Excerpt of the time series of flight E-AS-04 around one hour before and after the acidic values are measured marked with the black box. The external data are interpolated on the C-ToF-AMS time which smooths out maximum values to some extent.

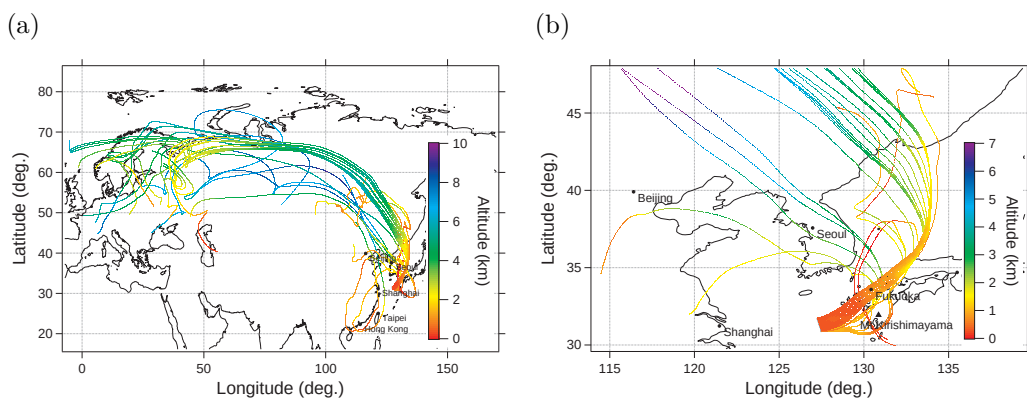


Figure 43: (a) 240 h FLEXTRA backward trajectories starting at the flight segment comprised of acidic values during flight E-AS-04. They are colour-coded with altitude and (b) is a zoom of the last 72 h before the measurements.

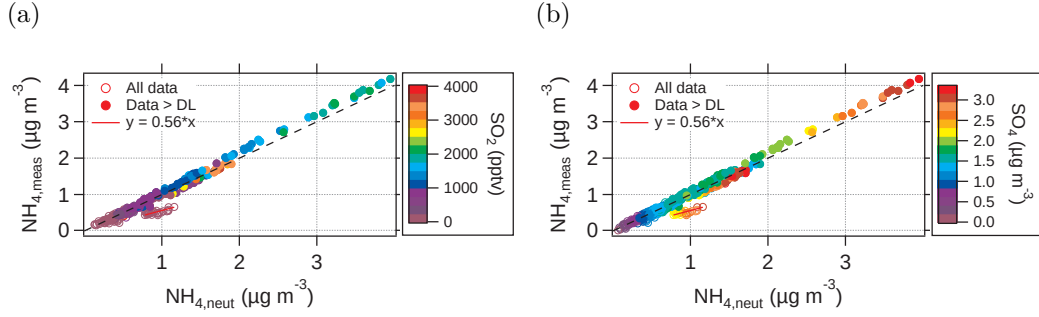


Figure 44: Scatter plots of the $\text{NH}_{4,\text{meas}}$ versus $\text{NH}_{4,\text{neut}}$ mass concentrations for flight E-AS-07 on 22 March 2018 colour-coded with SO_2 VMR (a) and SO_4 mass concentration (b). The dashed lines are the 1:1 lines indicating neutralised aerosol particles. The open circles show all data while the data of the closed circles are filtered for values below the DLs of the inorganic species. The SO_2 data points are interpolated to the C-ToF-AMS time and show less data points because of measurement gaps. The fit function is a least squares fit through the points of the event.

explosivity index (VEI) of 3. The Sinmoedake is a stratovolcano of the Kirishimayama volcano group on the island Kyushu in Japan with a height of 1421 m. Daily eruptions occurred between 06 and 13 March 2018, leading to ash plumes of 3 km above the crater drifting south-east and taking 24 h to dissipate (even 4.5 km were reached on 10 March). The explosions continued from 13 to 19 March, with emissions drifting north-east and east at 3.4 to 4.9 km altitude on 12 and 13 March and another plume reached 2.1 km above the crater after an eruption at 14:13 local time on 15 March. The backward trajectories take around 20 h close to the volcano and pass it again after the circle around 65 h later. Furthermore, while the air masses stayed low over the East China Sea and Sea of Japan, ship emissions influenced the composition and measurements as sulfur oxides (SO_x), NO_x , CO_2 (not shown), rBC (up to $0.18 \mu\text{g m}^{-3}$), and particulate matter are elevated. This results in a mixture of anthropogenic MPCs, ships, and natural volcanic emissions.

4.2.3 Acidic aerosol particles due to anthropogenic emissions

Another interesting feature in the acidity scatter plots occurred during flight E-AS-07 on 22 March 2018 with a ratio of 0.56 (see Fig. 44). It comprises non-filled circles because chloride and NO_3 are below their DLs. Averaged SO_2 values are low with 92 pptv compared to other data points with a larger VMR that arrange along the 1:1 line during periods of neutralised aerosol particles. The SO_2 might already be converted to SO_4 or removed by deposition, as it is shown later for the conversion ratio calculation. On the contrary, SO_4 mass concentrations increase with increasing NH_4 and have a mean concentration

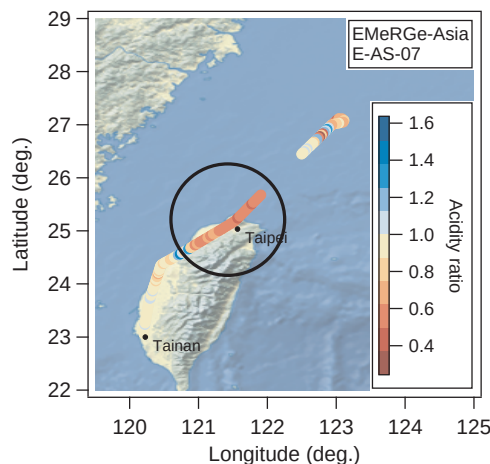


Figure 45: Map of the flight track of flight E-AS-07 colour-coded with the acidity ratio. The area of the acidic values is circled in black. The flight pattern is cut off at 05:11:30 UTC, otherwise the values on the way back would overlap the values of interest.

of $2.43 \mu\text{g m}^{-3}$ during the period of acidic aerosol particles. Looking at the flight pattern in Fig. 45, the acidic values occurred north of Taiwan at a mean altitude of 4668 m. The data gap between 25.7°N and 26.4°N is due to C-ToF-AMS filter measurements.

The time series in Fig. 46 shows the higher SO_4 mass concentrations during the measurements of acidic values between 04:20:00 and 04:34:30 UTC marked by the black box. Remarkably, all trace gas VMRs are reduced during this time in comparison to the adjacent times (mean values are $\text{CO} = 101.15 \text{ ppbv}$, $\text{O}_3 = 50.39 \text{ ppbv}$, $\text{NO} = 0.03 \text{ ppbv}$, and $\text{NO}_y = 0.34 \text{ ppbv}$). However, the CCN are around 578 cm^{-3} and also rBC shows averaged values of $0.13 \mu\text{g m}^{-3}$. The ageing markers (f_{44} and the toluene to benzene ratio) indicate a medium to old age of aerosol particles and air but are quite scattered. Furthermore, the tagged CO from the HYSPLIT dispersion model gives no signal for the selected target regions. So, the trajectories need to be considered to evaluate the air mass origin.

In contrast to the previous examples, volcanic emissions could be irrelevant this time. Based on the Global Volcanism Program (2018c,d), the most active volcano of the Philippines, the Mayon, in the Albay Gulf north-west of Legazpi City (13.257°N , 123.685°E , and 2462 m high) showed some activities with a maximum VEI of 2 in the week before the flight. During this time, dark grey ash plumes reached up to 700 m above the summit crater and the Mayon released SO_2 emissions between 969 and 2077 t d^{-1} but the steam-and-gas emissions drifted south-west on 14 March 2018. Pyroclastic activities continued until 27 March 2018. However, the backward trajectories did not cross the Philippines but show air mass origin from the west instead (see

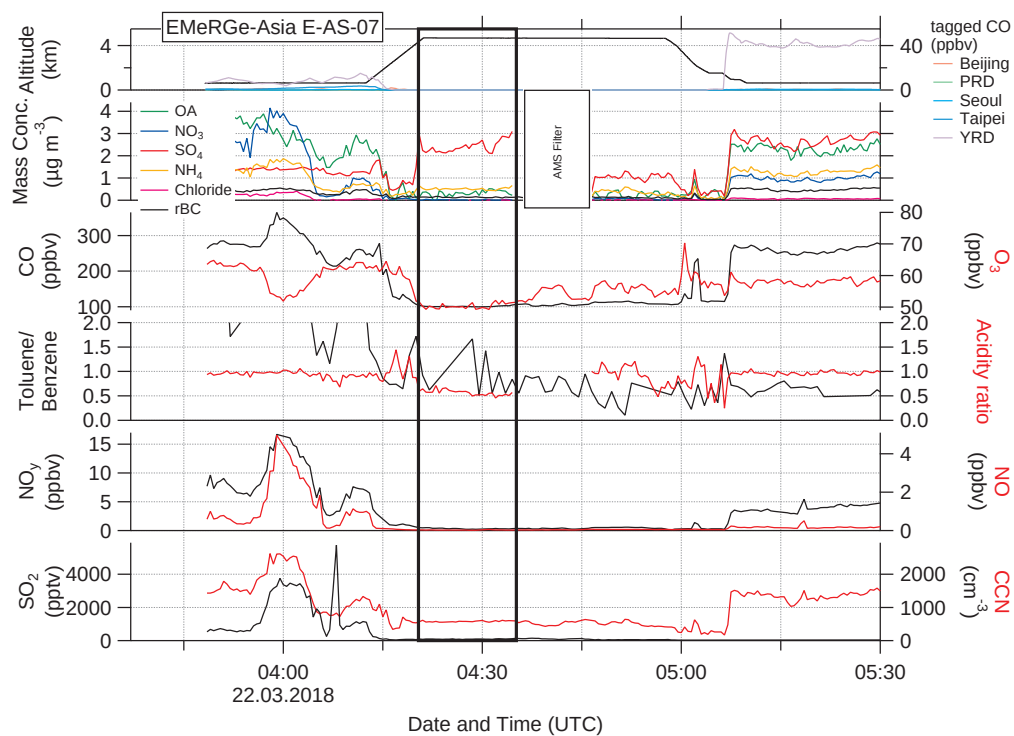


Figure 46: Excerpt of the time series of flight E-AS-07 from take-off until around one hour after the acidic values are measured marked with the black box. The external data are interpolated on the C-ToF-AMS time which smooths out maximum values to some extent.

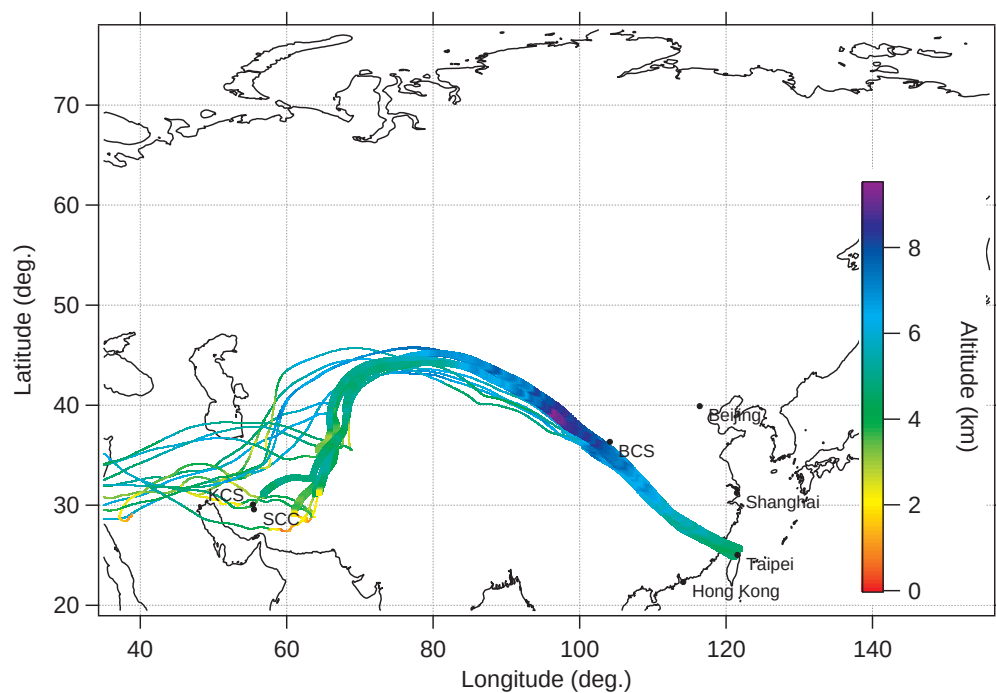


Figure 47: 180 h FLEXTRA backward trajectories starting at the flight segment comprised of acidic values during flight E-AS-07. They are colour-coded with altitude. The beginning of the thicker lines indicate the location where the trajectories have their last BL contact. From there, they travel in the free troposphere to the measurement location. Abbreviations are: Baiyin copper smelter (BCS), Khatoonabad copper smelter (KCS), and Sarcheshmeh copper complex (SCC).

Fig. 47). Thus, the volcano emissions might be insignificant for the reduced acidity.

Another explanation for the emission sources could be air masses coming from regions in Iran and China as the backward trajectories have their last contact to the BL there (see thicker lines in Fig. 47). Lelieveld et al. (2015a) and Salmabadi and Saeedi (2019) investigated emission sources over the Middle East using the Ozone Monitoring Instrument (OMI) on the Aura satellite, showing elevated SO_2 signals over the Persian Gulf and Iran close to coal-fired power plants, oil and gas industries, and smelter sites. The report by Dahiya et al. (2020) provides a comprehensive analyses of the global SO_2 emission hotspots in 2018 and 2019, quantifying the main sources in this area in 2018 as the oil and gas complexes Zagros (Iran) and Das Island (Abu Dhabi, United Arab Emirates) and the Sarcheshmeh smelter (Iran) with 550, 405, and 339 kt yr^{-1} , respectively. They were prominent sources of SO_2 already before 2018 and their evolution during the 2005–2014 period was traced by Fioletov et al. (2016) including other sources worldwide.

Sarcheshmeh copper complex (SCC) is a huge copper mine in the Kerman Province in South-Eastern Iran estimated as the second largest copper deposit worldwide containing 5 % of the world's total (DKL Engineering, Inc., 2018). The area has a mean altitude of about 2600 m. Another copper mining and smelting hub in the Kerman Province is the Khatoonabad copper smelter (KCS) which is located 180 km west of Kerman City, 35 km east of Shahr-e-Babak City and 45 km north-west of the SCC. It is operational since 2004. Pollutants originating from copper production are e.g. particulate matter, NO_x , volatile organic compounds (non-methane VOC and CH_4), CO, CO_2 , and nitrous oxide (European Environment Agency et al., 2019). Furthermore, copper smelters are a source of SO_x . Salmabadi and Saeedi (2019) identified the local transport routes of SO_2 originating from the KCS (160 kt yr^{-1}) and the potentially affected areas around the smelter throughout the seasons.

Atmospheric travelling times could be inferred from the trajectory analysis. The trajectories coming from the Iran travel 111 to 135 h until they are measured close to Taiwan. They stay in the BL over South-Eastern Iran around 2.5 d and accumulate emissions from the smelters. Some of the trajectories leave the BL close to the Baiyin copper smelter (BCS) in the Gansu Province in China which emitted 38 kt SO_2 in 2018 (Dahiya et al., 2020) and take around 42 h to reach the measurement location (see thicker lines in Fig. 47).

The trajectory analysis shows that local emissions not only influence the surrounding area but the pollutants are also transported over a long distance and can have an effect far away from their source. Compared to the previous two examples, the measured SO_2 is low resulting either from SO_2 transformation to SO_4 or SO_2 removal by deposition. The aerosol mass concentrations

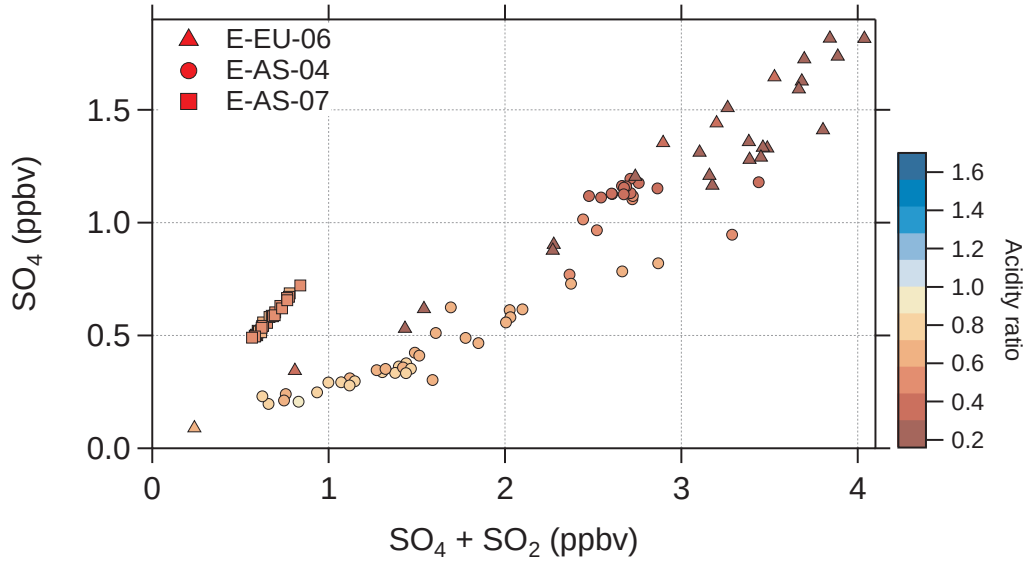


Figure 48: SO_4 production in connection with the total sulfur ($\text{SO}_4 + \text{SO}_2$) for the three examples of acidic aerosol particles colour-coded with the acidity ratio.

are also lower than for the two cases before which might also be related to the longer distance to the sources and further processing, dilution with background air, or removal along the way. The lifetime of SO_2 or in other words the conversion time to SO_4 is dependent on many factors which are reviewed in the following.

4.2.4 Conversion ratio of SO_2

The SO_4 production with respect to the total sulfur is depicted in Fig. 48 for all three flights combined. Both cases involving volcanic SO_2 emissions (E-EU-06 and E-AS-04) agree quite well. For the longer trajectory travel time of flight E-AS-07, the points are detached from the others but show an increase in SO_4 concentration, too. Their total sulfur concentration is lower and the increase more linear. Here, around 86 % of the SO_2 is already converted which is more than double as for the other two events. Dunlea et al. (2009) encountered transported Asian plumes in April and May 2006 over the Eastern Pacific Ocean, confirming the findings of Fig. 48: the older pollution layer is located along the 1:1 line and the younger data points below the 2:1 line. They assigned atmospheric processing times of 168 - 240 h to the older and 48 - 96 h to the younger measurements which are comparable to the air mass travel times of the trajectories listed in Table 11.

The conversion time of sulfur is depending on many factors which are presented in the literature comparison in the following Sect. 4.2.5. The calculation for the conversion ratio of sulfur (F_s) and the corresponding e-folding time (T_e) are described in Sect. 3.6.6. The results for the previously presented

Table 11: Overview of the conversion ratio of sulfur (F_s), estimated air mass travel times t due to volcanic eruption (Erup.) or taken from the backward trajectories (Traj.), and calculated e-folding time (T_e) for the three example flights containing periods of acidic aerosol particles described previously.

FLIGHT	F_s	t		T_e	
		Erup. (h)	Traj. (h)	Erup. (h)	Traj. (h)
E-EU-06	0.42	~48	78	~88.8	145
E-AS-04	0.32	~48	65	~122.4	165.8
E-AS-07	0.86	-	111 - 135	-	56.5 - 68.7

flight examples containing acidic aerosol particles are summarised in Table 11 and differ depending on the time since emission release. The travelling time of trajectories provides a good estimate while for volcanic emissions the time of an explicit eruption will lead to more accurate results.

Eirenschmalz (2021) estimated SO_2 lifetimes of 10 - 15 h for EMeRGe-Europe and 30 - 43 h for EMeRGe-Asia, utilising VMR of CO and organic peroxy radicals RO_2 . The shorter lifetimes in Europe result from an enhanced photochemistry in European summer compared to Asian spring. The shorter times in comparison to the results in Table 11 might arise from her focus mainly on MPCs plumes while the previously discussed examples also include natural SO_2 sources and further anthropogenic emissions not only originating from MPCs.

4.2.5 Comparison with literature

The lifetime of SO_2 varies between hours and months depending on many factors, e.g. the emission source and region, injection height of volcanic eruptions, RH, and season. Chin et al. (2000) estimated global mean tropospheric lifetimes of 2 d for DMS, 1.8 d for SO_2 , 5.8 d for SO_4 , and 7.1 d for MSA from model results. In addition, He (2012) studied trends of air pollutants and their regional transport in the free troposphere, involving SO_2 lifetimes of 38 ± 7 h during spring in China.

In connection with volcanic eruptions, the relationship between the SO_2 injection altitude and lifetime was evaluated by Carn et al. (2016) using satellite data. Furthermore, they suggested a possible dependency on the injected SO_2 amount and latitude. The analysis showed long lifetimes for large injections at high altitudes (e.g. 40 d for Mount Pinatubo which injected in around 25 km altitude) and less than one week for smaller injections near the tropopause. The Kasatochi Island plume was analysed by Jurkat et al. (2010) after they measured it over Europe at altitudes between 7 and 11.6 km

around three months later in October 2008. They found a F_s between 0.73 and 0.82 with 0.75 in the densest part of plume and calculated an upper limit of the e-folding time of 62 d for the SO_2 conversion in the lowermost stratosphere. Li and Cohen (2021) found monthly mean effective lifetimes of 43 to 61 h for the volcanic SO_2 plume between 1.10 and 2.73 km altitude for an active degassing period at Mount Kilauea in summer 2008 which matched with the 1 to 2 d from Beirle et al. (2014). They calculated sulfate lifetimes of 40 – 64 h which are lower in comparison to the global tropospheric averages of 3 to 5.5 d by Textor et al. (2006) and might be explained by rapid wet scavenging above 1.1 km.

Other factors can influence the conversion of SO_2 , namely RH, the season, and the time of day. Khoder (2002) confirmed the dependency of F_s on RH and also found a correlation with increasing O_3 while measuring in an urban area in Egypt in two different seasons (winter 1999/2000 and summer 2000). Furthermore, F_s in summer (0.09–0.28, mean 0.17) was about 2.22 times higher than in winter and increased during the day compared to the night. Kadowaki (1986) assessed similar correlations in a mixed residential/light industry area of Nagoya.

Decreasing trends of acidity were reported across North America and Europe during the last decades, mainly caused by reduced sulfate and nitrate concentrations due to diminished anthropogenic emissions of SO_2 , NO_x , and NO_y (Pye et al., 2020). Tilgner et al. (2021) added that other sources involved in acidification (e.g. DMS, volcanic SO_2 , chlorides from sea-spray-aerosol, or organic acids) could become more important in the near future depending on the amount of ammonia released from agriculture fertilisation. Many studies stated that a reduction in SO_2 emissions will also diminish isoprene-derived epoxydiols (IEPOX) and isoprene-related SOA formation, in numbers: for the South-Eastern USA a 25 – 48 % reduction in SO_2 reduced the IEPOX SOA formation by about 35 – 70 % (Pye et al., 2013; Marais et al., 2016; Budisulistiorini et al., 2017; Zhang et al., 2018) or for the PRD a sulfate aerosol cutback of 25 % lowered the IEPOX SOA by 45 % (He et al., 2018). Another affected aspect is the pH of clouds which is quite sensitive to anthropogenic emissions of sulfur, NO_x , and ammonia. Emission control had increased cloud pH in recent decades leading to a larger sulfate mass burden over polluted regions (by changing the rate of aqueous phase oxidation of SO_2 by O_3). Furthermore, effects on the aerosol radiative forcing were introduced while the related uncertainty increased locally (Turnock et al., 2019).

4.3 OXIDATION STATE OF OA

This section gives insights into the oxidation state of OA during EMeRGe. Therefore, the methods described in Sect. 3.5.4, 3.6.1, 3.6.2, and 3.6.3 are applied and combined with the photochemical ageing of VOCs and other

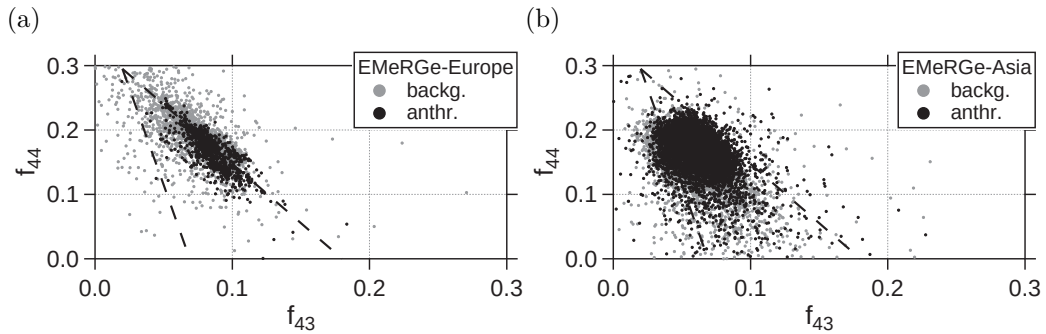


Figure 49: Triangle plots of f_{44} versus f_{43} for EMeRGe-Europe (a) and -Asia (b; flights E-AS-03 to E-AS-14). The data are divided between anthropogenic and background origin applying VOC thresholds as introduced in Sect. 3.6.2.

trace gases. First, a general overview of the data is given. Then, an explicit example of aerosol formation and processing in anthropogenically influenced air masses near Taiwan is presented including results of different air mass history modelling. In addition, the oxidation state of OA associated with individual MPCs in Europe and East Asia is investigated and related to other ageing methods. Lastly, a literature comparison is given.

4.3.1 General overview

The data of the two EMeRGe campaigns arrange differently in the triangular shape of the f_{44} versus f_{43} representation (see Fig. 49). The majority of the data points are within the triangle but some also scatter outside of it. The boundary lines of the triangle are empirical and were introduced by Ng et al. (2010) to guide the eye after performing atmospheric measurements. Thus, measurements can deviate from the original area. The differentiation into background and anthropogenic origin based on VOCs, already used in Sect. 4.1.3, is also utilised here to separate the data. For EMeRGe-Europe, the background aerosol particles cover a wider range of f_{44} values than the anthropogenically influenced aerosol (see Fig. 49a). Furthermore, the ratio f_{44} reveals older particles for the background, located in the upper corner of the triangle. On average, the measured f_{44} values in Europe are greater than 0.1 and accumulated along the right line within the f_{44} - f_{43} space, meaning younger aerosol particles are encountered only rarely. In contrast, a part of the data from EMeRGe-Asia in Fig. 49b also falls into the lower (less aged) f_{44} range while less data points can be found in the upper (most aged) corner of the triangle. However, the majority of points arrange between 0.1 and 0.22 on the f_{44} axis filling the whole space of the triangle between the boundary lines. The difference between the background and the anthropogenically influenced particles is not as pronounced as for the European data set. Though, the

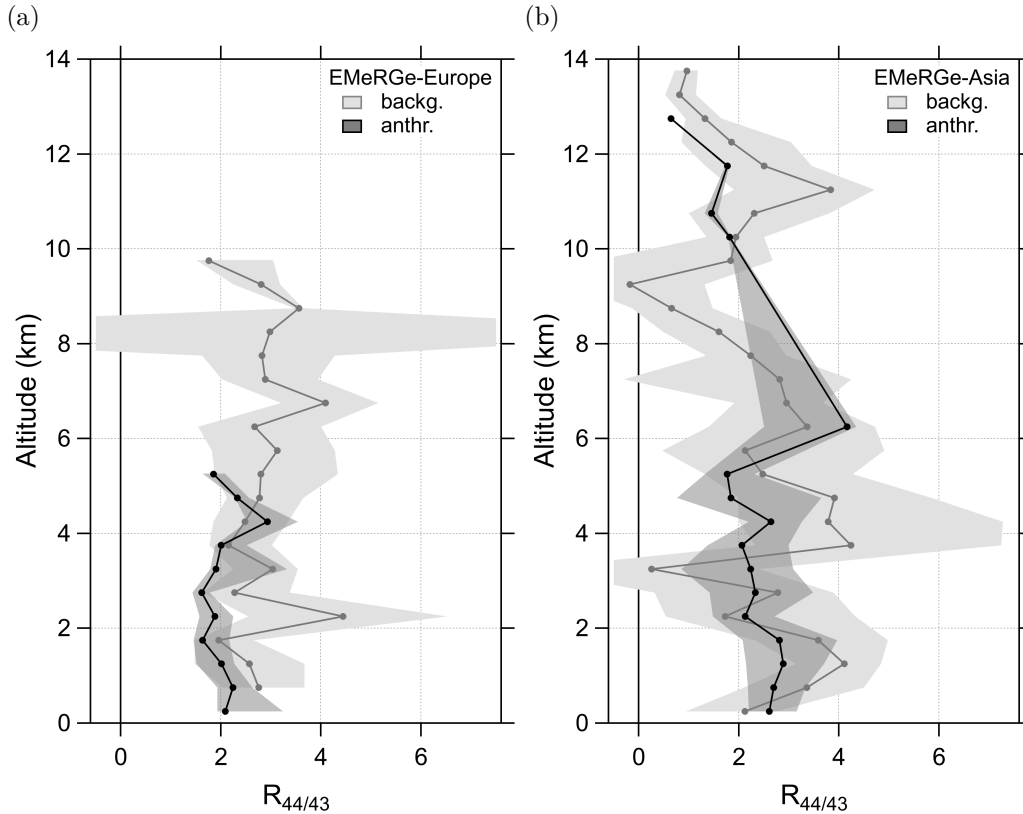


Figure 50: Averaged vertical profiles of the ageing ratio $R_{44/43} = f_{44}/f_{43}$ for the EMeRGe-Europe (a) and -Asia (b; flights E-AS-03 to E-AS-14) campaigns in 500 m altitude bins. The dots represent the medians and the shaded areas the interquartile ranges. The vertical profiles are divided between anthropogenic and background origin applying VOC thresholds. The number of points per altitude bin is the same as in Fig. 25.

background points are slightly shifted towards the left line but this is not evident from Fig. 49b because they are hidden by the anthropogenic points.

The points in the triangle are coloured with different variables in order to derive a connection or to find possible groupings, e.g. the toluene to benzene ratio is tested which is a marker for photochemical ageing of VOCs as introduced in Sect. 3.6.2. No clear correlation with the oxidation state of OA becomes obvious because too many points are involved and overlap for the total campaign data sets (see Figure 68 in the appendix). The same applies to correlations with other variables, e.g. the VMR of CO or the flight altitude. A more detailed analysis on a case by case basis is needed to establish a link between aerosol and VOC ageing, as is presented in Sect. 4.3.2.

Changes of the ageing ratio $R_{44/43}$ with altitude for both campaigns are displayed in Fig. 50 where the VOC-based separation between anthropogenically influenced and background data is applied. The ratio $R_{44/43}$ is calculated by dividing f_{44} by f_{43} . Thus, it increases with the oxidation state.

The anthropogenic profiles are almost constant with ratios around 2 up to altitudes of 4 km. This indicates a well-mixed BL and lower troposphere. An exception are the lowest 2 km in East Asia where the ratio shows values around 2.75 implying older aerosol particles than over Europe. In addition, the anthropogenic profiles for both campaigns peak above 4 km which might be induced by long-range transport of air masses and aerosol particles experiencing more photooxidation which increases the ratio. Aerosol particles originating from background air are generally less oxidised during EMeRGe-Asia with an averaged $R_{44/43}$ of 2.33 compared to 2.84 for EMeRGe-Europe. Furthermore, the background aerosol particles are more oxidised than the anthropogenically influenced particles. Surprisingly, their vertical profiles vary more than expected involving some unexplained features, e.g. the strong increases at around 2.25 and 6.75 km during EMeRGe-Europe which deviate from the otherwise rather constant profile. This might be due to long-range transport of air masses increasing the ageing ratio $R_{44/43}$. Another distinctive characteristic is the larger variability in the background profile during EMeRGe-Asia. The values are not randomly scattered, otherwise the quartile ranges would have to be larger. The ratio reaches minima at around 3.25, 9.25, and 13.25 km which are possibly caused by new particle formation involving particles growing into the size range measurable by the C-ToF-AMS. They could have formed in the outflow of convection releasing aerosol precursor gases at these altitudes (Williamson et al., 2019). Schulz et al. (2018) explained less oxidised OA in the upper troposphere over the Amazon by freshly formed isoprene-epoxydiol-derived SOA. In contrast, the background OA of the EMeRGe data set is filtered for isoprene. Thus, other precursor gases for OA like monoterpenes or sesquiterpenes could be relevant as they also have a large potential to form SOA (Kanakidou et al., 2005). An additional explanation for the minimum at 3.25 km could be convective upward transport of particles from the near BL. This is supported by the vertical profile of the background SO_4 in Fig. 25b showing an increase of mass concentrations comparable to values in the lowest altitude bin. Other indicators that could be considered to further examine the suggestions about the local minima are the vertical profiles of rBC and the aerosol number concentration as well as the particle size distributions, and trajectory origins. Moreover, the structure of the profiles could be influenced by single flights which needs additional studies.

4.3.2 *Photochemical processing of air masses encountered near Taiwan*

As presented in the previous Sect. 4.3.1, the whole campaign data set gave an overview on the oxidation state of the OA but could not attain a clear connection to other measured variables. Thus, the following analysis focuses on data from only one flight during EMeRGe-Asia. Furthermore, the aerosol

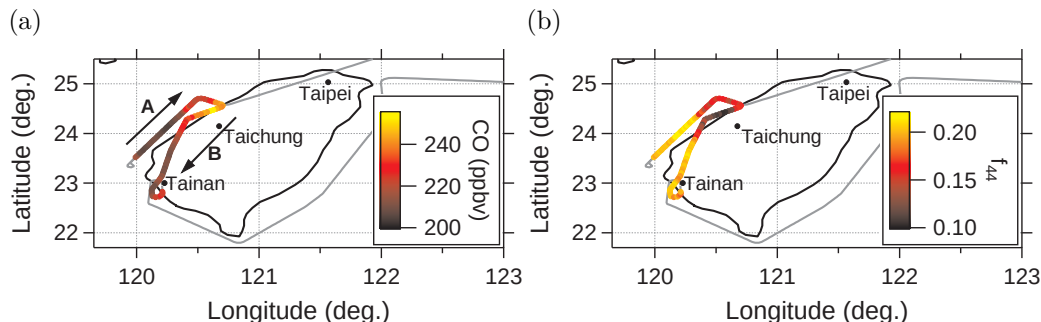


Figure 51: Map of the flight path close to Taiwan of flight E-AS-04 on 17 March 2018. The flight path is given in grey and the flight path of interest at the west coast of Taiwan is colour-coded with the CO VMR (a) and the ageing tracer f_{44} (b). The selected flight section comprises of two parts: part A (8:39:30 - 9:07:30 UTC) in 1 km altitude above the sea with flight direction to the north-east and part B (9:07:30 - 9:41:30 UTC) in 0.65 km mostly over land heading south-west.

composition is analysed and the oxidation of aerosol particles is combined with different trace gas metrics to estimate an age. Flight E-AS-04 on 17 March 2018 is chosen as a suitable example to follow the oxidative processing of OA. Here, the flight section at the west coast of Taiwan is evaluated in detail. It consists of two parts as depicted in Fig. 51a: one at approximately 1 km altitude above the sea with flight direction to the north-east and the other at around 0.65 km mostly over land with flight direction south-west. As the colouring with the CO VMR in Fig. 51a indicates, there is a emission source at the west coast of Taiwan close to Taichung enhancing the measurements in the Northern part of the flight section while CO values decrease with distance further to the south due to dilution with surrounding air masses. A strong local source in this area is the Taichung power plant which is located 21 km west of Taichung city centre. It is the second largest coal-fired power station in the world and the world's largest emitter of CO_2 with around 36 Mt in 2009 according to CARMA v3.0 (CARbon Monitoring for Action). Moreover, the power plant is responsible for 15 % of Taichung city's $\text{PM}_{2.5}$ (Chen, 2017). Lu et al. (2019) simulated the distribution of various $\text{PM}_{2.5}$ sources along the west coast of Taiwan and found an annual impact of around 2.8 % on the air quality in Tainan City in 2013 caused by the Taichung power plant. Freshly emitted aerosol particles close to the source are expected to be younger and less oxidised than transported aerosol particles at a distance to the source. This can be observed for the ageing marker f_{44} of OA in Fig. 51b. In the vicinity of Taichung, the f_{44} values are around 0.1 indicating photochemically young OA while the ratio increases to the south-west demonstrating an altered oxidation state.

The selected flight path at the west coast of Taiwan is divided into four different segments according to the preceding air mass origin as visualised

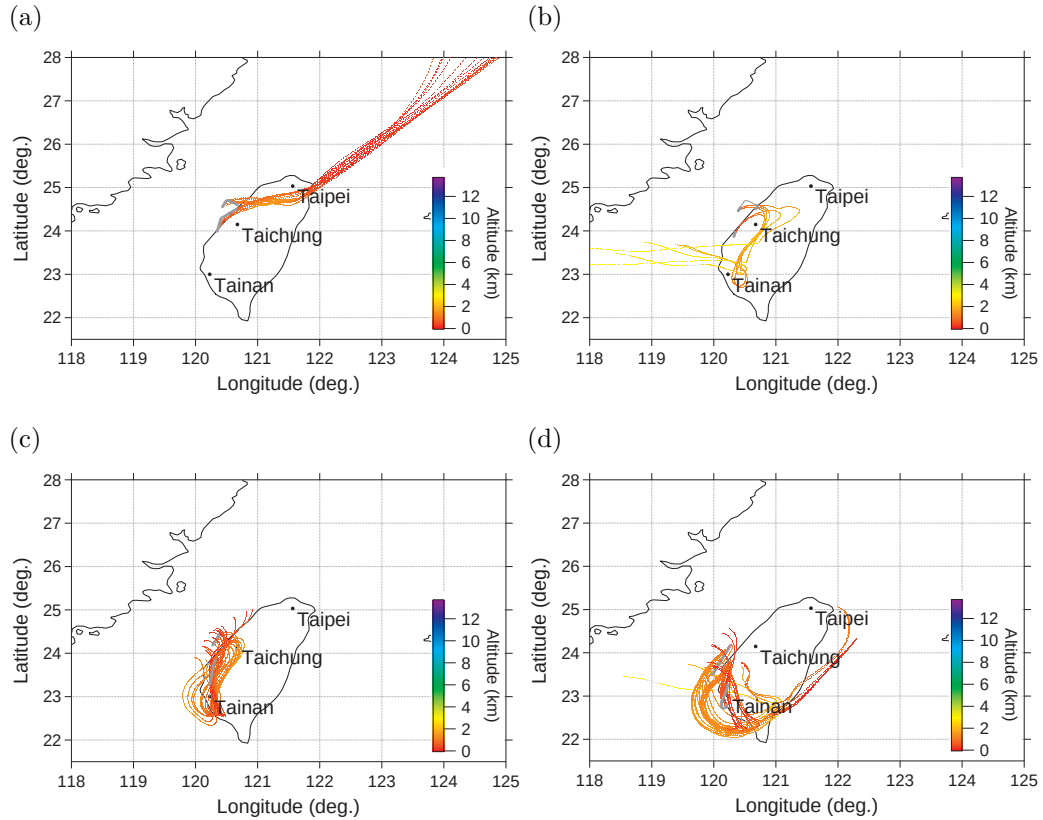


Figure 52: Flight segments west of Taiwan separated after the trajectory origin in four regions: north (a), centre (b), south (c), and south-east (d). The backward trajectories are colour-coded with altitude having a length of 24 h for "north" and 36 h for the other segments. Further information on altitude and time period for each flight segment can be found in Tab. 26 in the appendix.

by the 24 to 36 h FLEXTRA backward trajectories in Fig. 52. The segments are named accordingly as "north", "centre", "south", and "south-east". The whole trajectory lengths of 240 h for each segment are added in Fig. 69 in the appendix. To increase the number of data points in each segment and thus, to increase the statistics, the analysis of the trajectory history is performed taking both altitudes together and the averages used later are calculated including both heights. Further information on mean altitude and time period for each flight segment can be found in Tab. 26 in the appendix. The backward trajectories for the segment "north" start over Mongolia and Russia and decrease their altitude while turning to the south. They split up into two branches. One branch of trajectories goes over the Yellow Sea and the other over the Sea of Japan. After passing South Korea the branches reconvene while their altitude remains below 2 km. They cross Northern Taiwan passing Taipei before reaching the measurement location. The second segment is called "centre" because the air masses travel through the middle of Taiwan with a turn around Tainan and Taichung before their observation. The air mass origin is over the Persian Gulf and India. From there, the air masses travel east showing a maximum subsidence over Bangladesh and Myanmar (see Fig. 69b in the appendix). After that, they continue east towards Taiwan at an altitude of 3 km (see Fig. 52b). The trajectories of the segment "south" end comparable to the ones of "centre" but stay longer below 2 km before. They circle along the west coast of Taiwan and collect emissions at the same location at least twice (see Fig. 52c) which result in more oxidised aerosol particles than for the segment "centre" as shown later. The trajectories move directly over the city of Taichung and its power plant as well as the other cities and power plants at the west coast of Taiwan. As apparent from Fig. 69c in the appendix, the trajectories reach Taiwan in three branches from different directions: one from the north-west decreasing altitude immediately from over 4 km after leaving mainland China, the second arrives from the north over the Yellow and East China Sea at an altitude below 3 km, and the third one enters from the east over the Pacific Ocean after the air masses decreased their altitudes on the way from Mongolia over South Korea and Southern Japan. The ending of the trajectories for the fourth segment "south-east" is depicted in Fig. 52d. Half of the trajectories end after a wide curve over the ocean around the Southern tip of Taiwan after circling the whole island while the other half entered west with a sharp turn around Tainan. The pattern of the whole 240 h trajectories for the segment "south-east" in Fig. 69d in the appendix is comparable to the segment "south" besides more trajectories are staying low over Japan and the Pacific Ocean as well as an additional branch of trajectories comes from west over India and Indochina.

A special characteristic of the air mass movements in Taiwan is induced by the mountain ranges in the east traversing the island from north to south. The highest mountains are the Yushan in the central mountain range with

3952 m and the Xueshan in the north with 3886 m. Cheng et al. (2022) investigated the flow circulation over Taiwan and found that a sea breeze transported O_3 from the emission sources at the coast inland where it was blocked by the mountains and accumulated in the Puli Basin close to Taichung. They further encountered mountain valley circulations which were studied also by Ooi et al. (2021) in relation to long-range transport of BC originating from BB in Northern Indochina. The diurnal mountain valley breeze might trap or disperse the pollution mixed with local emissions depending on the time of day and prevailing weather situation.

Four comparison segments are added to obtain a holistic picture of the emission distribution, the air mass composition, and the ageing metric applied below: two segments east of Taiwan in two different altitudes, one segment at the same location as the flight path west of Taiwan but at a higher altitude, and the fourth segment is selected to consist of background air based on the VOC thresholds used before. Their trajectories are plotted in Figs. 70 and 71 in the appendix. The segments "east" and "east 2" in the east of Taiwan are taken to investigate the local emissions around the island in almost the same latitudinal range expecting cleaner conditions with more biogenic or marine influence than close to the emission sources on the west coast. The segments are probed in the beginning of the flight between 1:50 and 3:00 UTC while the four main segments introduced before are measured after 8:40 UTC at the end of the flight. The segment "east" has a mean altitude of about 0.5 km and the flight direction is to the north-east. The trajectory progression is comparable to the segment "north" besides the trajectories circling longer over Russia and two trajectories passing in the vicinity of the YRD and Beijing. The segment "east 2" follows the segment "east" after an altitude change to around 3.1 km including a turn to the east. A change of the wind direction with altitude is observed which also modifies the trajectory origin to air masses coming from west. Some of the trajectories even came from the Mediterranean Sea within the previous 240 h but stayed high above 6 km while others descended to accumulate emissions over Indochina and the PRD (see Fig. 71b in the appendix). The segment "high shuttle" is performed at 2.5 km as the upper level of the stacked profile at the west coast of Taiwan with a flight direction to the south-west. The corresponding trajectories started west over the Mediterranean at almost 10 km altitude and decreased the altitude towards the measurement location. Some of them descend even further to altitudes below 2 km over Pakistan, India, and Bangladesh. The "background" segment is taken from the middle of the flight in an area close to South Korea and Japan at an altitude of approximately 3.1 km. Before the observation, the air masses decreased their altitude from around 9 km. They were meandering from the Atlantic Ocean to the east over Europe, Kazakhstan, Mongolia, and Northern China within 240 h. In the beginning over the Atlantic Ocean, some of the trajectories

started below 2 km while three trajectories also reached the BL over Saudi Arabia and the Persian Gulf.

The FLEXTRA-based BL fractions of each region vary between each flight segment and also for the different backward times of 24 to 240 h (see Fig. 72 in the appendix). The trajectories coming from the north passing Beijing and the YRD are longest in the BL although the time within the BL is decreasing with increasing backward time. This effects especially the flight segments "north" and "east" having a BL fraction of 82.3 % and 98.1 % after 24 h, respectively, and still around 51 % and 77 % after 72 h. The segment "centre" has the lowest BL proportions of the four low level altitudes west of Taiwan starting with around 5 % after 24 h, followed by a decrease of time within the BL, and having its highest BL fraction of 6.4 % after 216 h according to the descended trajectories over Bangladesh and Myanmar. The BL fraction for the segments "south" and "south-east" started with around 1 % and increased up to 26 % after 96 h where the trajectories accumulated emissions while circulating on the west coast of Taiwan. For "south-east" the trajectories even did a full turn around the island after passing the PRD and coming from the Pacific Ocean where some trajectories stayed some days longer within the BL than for the "south" segment. The trajectories of the segments "high" and "east 2" have almost no BL contact within the first 72 h. Their BL fraction increases afterwards to around 10 % due to descending air masses over Indochina, India, and Bangladesh. The "background" segment has no BL contact within the first 96 h as well and reached a maximum fraction of 4.5 % after 240 h.

The summed mass concentrations of all C-ToF-AMS species, the rBC mass concentration, and the total number concentration measured by the OPC showed a remarkably similar behaviour throughout this flight with small peaks at the same times (see Fig. 53). Just around 7 UTC, there is a deviation as the C-ToF-AMS shows values greater $2 \mu\text{g m}^{-3}$ while the other two parameters remain close to zero. The previously defined flight segments are indicated by boxes of different colour and accompanied by the flight altitude in the upper panel. Mass concentrations of rBC are greater zero most of the time during this flight and the mean values are largest at the end of the flight after 8 UTC during the passage west of Taiwan with around $0.6 \mu\text{g m}^{-3}$. However, the rBC is probably not generated by BB as the acetonitrile threshold of 199 pptv has only rarely been exceeded during this flight (not shown). During the "background" segment in the middle of the flight, rBC and the total aerosol number remained low, indicating that the VOC separation is sufficient although aerosol mass concentrations of around $1 \mu\text{g m}^{-3}$ remained which are mainly comprised of SO_4 and OA. A detailed analysis of the aerosol composition in the individual segments is presented in the following paragraph. The top panel of Fig. 53 also includes the time series of the calculated CO VMR induced by pre-defined target areas as introduced in

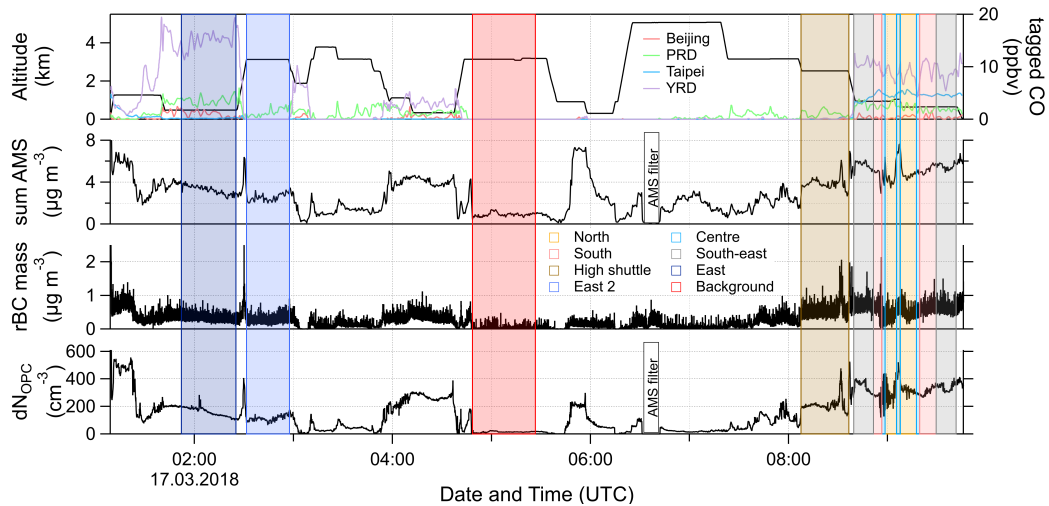


Figure 53: Time series of the whole flight E-AS-04 on 17 March 2018. Displayed are the flight altitude together with the tagged CO for the four different target regions (a), the sum of all C-ToF-AMS species (b), the rBC mass concentration (c), and the total aerosol number concentration measured with by the OPC (d). The coloured vertical areas indicate the times corresponding to the eight flight segments defined by the trajectories.

Sect. 3.5.4, namely Beijing, the PRD, Taipei, and the YRD. The averaged CO values and their relative contributions to one flight segment are listed in Table 27 in the appendix. The segments "east 2" and "high shuttle" are predominantly influenced by PRD emissions by up to 98%. The "east" segment has its main emission source in the YRD with around 76% but is also influenced by the PRD with around 18% and slightly from Beijing (5%) and Taipei (1%). The four regions west of Taiwan are influenced by the YRD with up to 58%, by Taipei with approximately 33%, and by the PRD with maximum 16%. Beijing has the least influence with only 2.6% as it is the furthest away. The calculated CO is zero during the "background" segment because the section is selected to show no anthropogenic emissions. In contrast to the tagged CO VMRs modelled with HYSPLIT, the FLEXTRA backward trajectories showed no influence of the PRD for the segments "north" and "east" as well as no direct crossing over the YRD and Beijing. Furthermore, the FLEXTRA trajectories for the segment "centre" did not show a northward component, so no contact with the YRD and Beijing. These discrepancies arise from the different trajectory models and their accuracy of around 20% with travel distance (Draxler and Hess, 1998; Stohl, 1998).

The composition for each individual segment is displayed in Fig. 54 in two different ways: first, pie charts are separating the shares of every aerosol species and second, the average values for the aerosol parameters are complemented by the trace gases CO and O₃. The pie charts of the "north" and "centre" segment show the highest NO₃ fractions with around 12% due

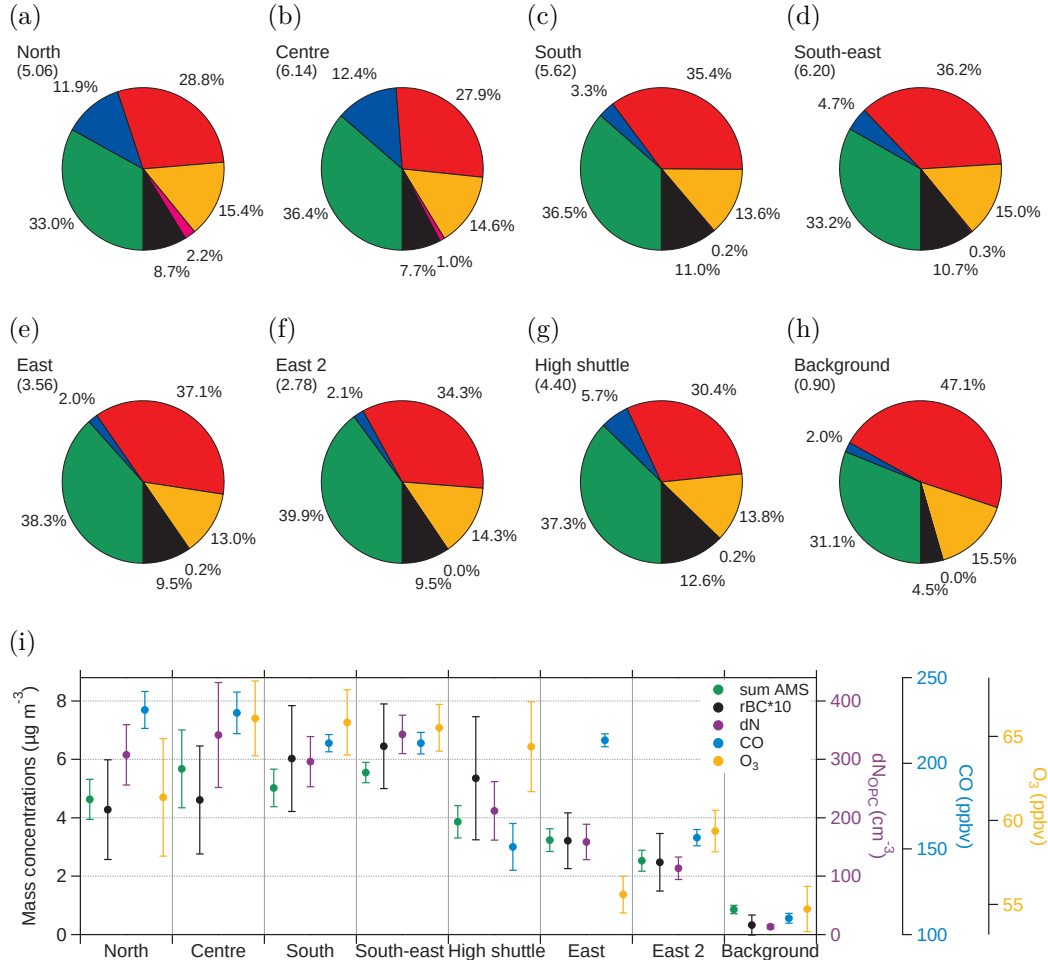


Figure 54: Overview on the average composition in the eight selected segments: (a) - (h) Pie charts containing the composition of the PM_1 aerosol particles including rBC. The absolute value of the mass concentration in $\mu\text{g m}^{-3}$ is mentioned below each segment name. Colours indicate OA (green), SO₄ (red), NO₃ (blue), NH₄ (orange), chloride (pink), and rBC (black). (i) Averaged values with standard deviation as error bars for each region for the mass concentration of the sum of all C-ToF-AMS species and rBC (times 10), the total number concentration dN_{OPC} from the OPC as well as the VMRs of CO and O₃.

to greater local pollution as CO, NO, and NO_y show maximum values of up to 256 ppbv, 0.5 ppbv, and 6.7 ppbv, respectively, around this region as well (not shown). The segments "high shuttle" and "south-east" have the second largest NO₃ shares of around 5 - 6 %. The OA shares for this four segments are between 33 and 37 %. The fraction of SO₄ is increasing around Taiwan from "north" with 29 % to "east" with 37 % implying SO₄ formation with increasing time after the emission. The aerosol compositions of the segments "south" and "south-east" are similar to each other containing less NO₃ than the "north" and "centre" segments, more rBC with around 11 %, and almost no chloride while the NH₄ fraction accounts for up to 15 %. The segments "east" and "east 2" are also comparable although the measurements are performed at different altitudes and the FLEXTRA backward trajectories showed other air mass origins. The OA fraction is greatest for these flight sections with almost 40 % compared to the others while the NO₃ shares are lowest with 2 % like within the "background". The "high shuttle" segment has the greatest rBC fraction with 12.6 % while the other anthropogenically influenced flight segments show at least 7.7 % while the "background" has the smallest fraction with 4.5 %. The relative contribution of NH₄ is most evenly distributed between the flight segments accounting for 13 to 15.5 %. The clearest difference in the aerosol composition is shown by the "background" segment containing almost 50 % SO₄ and the least OA fraction with around 31 % compared to the other regions. The higher amount of SO₄ in the background air masses has already been mentioned in the more general introduction of the data set in Sect. 4.1.3.

The total mass concentrations, as contrasted in Fig. 54i, are highest in the four segments west of Taiwan as well as the trace gases CO and O₃. Based on the VOC thresholds introduced in Sect. 3.6.2, the data of this area are marked not only as anthropogenically influenced but also as strong anthropogenic as the benzene VMRs are greater than 130 pptv. The segment "south" contains more rBC than "centre" although the trajectories of both selections travelled the same path in the end. However, the air masses of the segment "south" came closer to the Taichung power plant and also crossed the west coast of Taiwan with all the other cities and power stations at least twice collecting more emissions. The composition of the segment "high shuttle" is comprised of lower values except for O₃ but they are still higher compared to the values east of Taiwan. There, the air is cleaner besides the CO VMR in the segment "east" which is similar to the values west of Taiwan and might originate from long-range transported emissions from the north.

The VOC tagging of "background" air masses matches well to the lowest values of CO with 110 ppbv and also to the lowest O₃ (see Fig. 54i). The aerosol parameters are minimal for the "background" segment as well. The CO histogram of the whole flight is displayed in Fig. 73 in the appendix showing three maxima at 107, 151, and 212 ppbv. The threshold for the CO

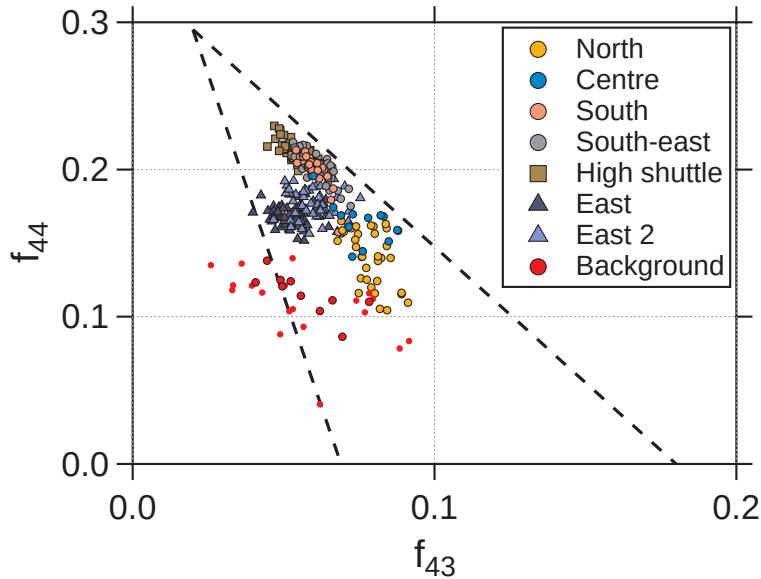


Figure 55: Data points from the eight selected flight segments in the ageing plot after Ng et al. (2010, 2011). They are separated for the different regions by the corresponding colour and comprise of values above the organic DL except for the background points, here the values below the DL are indicated in a lighter red and by smaller size.

background is set to 123 ppbv after the first maximum. The flight altitudes are determining the other two maxima, showing anthropogenically polluted air masses: the second maximum includes the segments "high shuttle" and "east 2" with altitudes above 2.5 km and the third maximum comprises the segments west of Taiwan below 1 km as well as the "east" section. The threshold defined for the CO background is in agreement with a value of 125 ppbv specified by Chen et al. (2021b) who measured in the Guangzhou urban area of China in autumn 2018.

The data for the eight different flight segments are displayed in Fig. 55 within the f_{44} versus f_{43} space. As explained in Sect. 3.6.1 and Sec. 4.3.1 above, this metric allows to visualise the degree of atmospheric processing of OA. The data from the flight segment denoted as "background" and those from the segment "north" show the lowest degree of processing while the "high shuttle" data are at the upper end of the triangle and are therefore more processed. Less oxidised aerosol particles in the "background" air masses were already discussed in relation to the averaged vertical profile of the ageing ratio $R_{44/43}$ in Fig. 50 on page 101. There, a minimum is observed in the altitude bin between 3 and 3.5 km which is consistent with the altitude of the "background" segment of this case study. Possible explanations for the lower oxidation state are new particle formation or convective upward transport of particles from the near BL. In Fig. 55, the measurements west of Taiwan form a line of proceeding oxidation starting with the segment "north",

followed by "centre" and ending with the segments "south" and "south-east" with almost similar f_{44} values. The "south" data are more processed than the "centre" ones although their trajectories cover the same area in the end but the trajectories corresponding to "south" circulated longer along the west coast of Taiwan. The air mass history based on the FLEXTRA trajectories is comparable for the segments "north" and "east" but within the triangle plot the section "north" shows less processed values extended over a wider range of f_{44} while the "east" data points are accumulated at more oxidised values and do not spread over a broad f_{44} range. This might be due to mixing with fresh emissions over land for the "north" measurements accompanied by a higher mean CO VMR and a closer distance to the strong point sources like the Taichung power plant. Although the measurements took place at low altitudes between 0.5 and 3.1 m as well as relatively close to the source regions of Taiwan, no data points with a f_{44} at the lower end of the y-axis are found. This means that even the younger air masses have already experienced significant photochemical processing.

In the following, this data set is examined more closely by means of other trace gases and photochemical markers. Figure 56 is comprised of a combination of the aerosol particle oxidation state with different trace gas metrics as an age estimate, namely the toluene to benzene ratio, the modelled and weighted mean age, and the NO_y to CO ratio. The x-axes are chosen so that the photochemical age increases to the upper right. Figure 56a displays the f_{44} against the toluene to benzene ratio (as introduced in Sect. 3.6.2) and consists of less data points because only data above the VOC DLs are considered. The smaller the VMRs, the less meaningful is the ratio. Benzene VMRs are always above the DL, only the toluene is below the DL sometimes. The combination with the toluene to benzene ratio shows that younger air masses with ratios greater than one correspond to lower f_{44} values than older air masses with a decreased ratio below one. The oxidation of anthropogenically influenced aerosol particles increases linearly with photochemical ageing of VOCs. After reaching a toluene to benzene level around one it seems like the ageing on the particle side stops at an plateau. For a toluene to benzene ratio below one only the gas phase shows further ageing.

Figure 56b compares the f_{44} with the weighted mean age calculated from the HYSPLIT model as introduced in Sect. 3.5.4. Table 27 in the appendix contains the arithmetic mean age contributions of the single target regions to the defined flight segments. No age appears for the "background" segment because the calculated CO only shows an increase for target regions and the "background" is defined to be non-anthropogenic. The "high shuttle" and "east 2" sections are the highest levels flown above 2.5 km and have the lowest calculated CO VMR values. They are mainly influenced by PRD emissions showing a mean age of around 60 h. The segment "east" is the lowest flight level with 0.5 km and clearly deviates from the other data in the graph. It

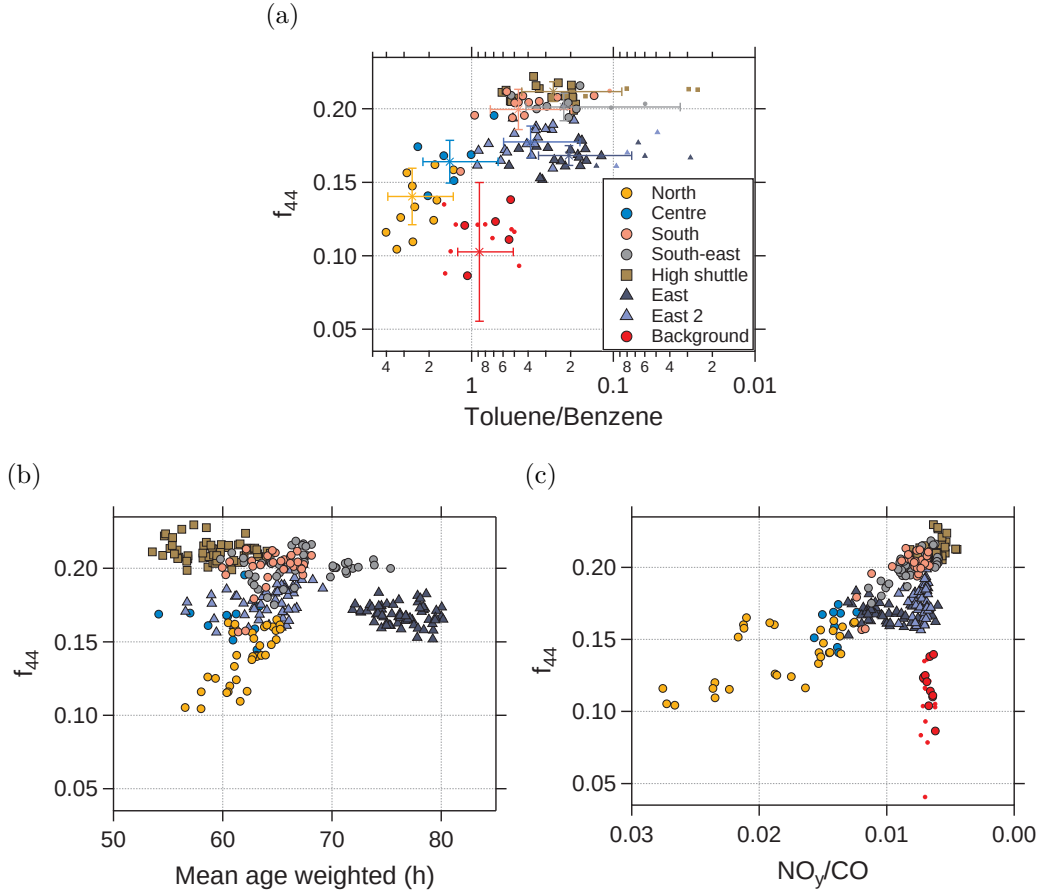


Figure 56: Combination of the aerosol particle oxidation state with different trace gas metrics as an age estimate: (a) f_{44} against the toluene to benzene ratio on a reversed logarithmic axis, so the photochemical age increases to the upper right. Black framed data points are greater than the DLs (checked for each VOC before calculating the ratio). Further displayed are the average values for each segment and the corresponding standard deviation as error bars. (b) f_{44} versus the modelled mean age weighted with the fraction of each target region. There are no data points for the "background" segment as the calculation is only performed for emitting areas. (c) f_{44} versus the NO_y to CO ratio on a reversed axis. Data points below the organic DL for the "background" segment are indicated by smaller size. The graphs are colour-coded with the defined segments and the legend in (a) is applicable to all three graphs.

has the highest modelled CO for the main source of the YRD contributing with around 76 % and showing the oldest weighted mean age with up to 80 h. Air masses from the PRD contributed with approximately 18 % to "east" and has an even older age of around 100 h. The four sections west of Taiwan fall along a line and their weighted mean age increases from "north" to "south-east" from 55 to 75 h. If the mean ages are split up for the different target regions the contributions are as follow: YRD with up to 58 % and a mean age of 79 h, Taipei with up to 33 % and a mean age below 19 h, and PRD with up to 16 % and a mean age of up to 113 h.

The last Fig. 56c within this analysis uses the NO_y to CO ratio as time axis as introduced in Sect. 3.6.3. The general findings for this metric are the same as for the other two before: a linear relation for the flight segments west of Taiwan is observed indicating a good agreement between the aerosol particle oxidation and the trace gas processing of anthropogenically influenced air masses. However, the two segments east of Taiwan as well as the "background" segment differ from the linear relation as their pollution levels are smaller than for the four segments west of Taiwan. The "background" points arrange vertically along a NO_y to CO ratio of 0.007 covering a range of less oxidised f_{44} . The CO VMRs remained almost constant at around 214 ppbv for the segment "east" and around 157 ppbv for "east 2". Furthermore, the NO_y VMR decreased during the "east" measurements while they are almost constant for "east 2" (not shown). Therefore, the data of the "east 2" segment are observed in extension of the "background" points but with higher f_{44} values. On the contrary, the "east" data are positioned more horizontally driven by the decrease of NO_y VMRs around a f_{44} of 0.17.

Although the general relation between aerosol particle and trace gas processing became obvious, a consistent picture could not be obtained and an allocation of a distinct time to a given f_{44} ratio is not possible as the situation is rather complex. Furthermore, the initial values at the source are unknown for the VOCs but would be necessary to calculate an ageing time. In addition, the emission strength is different for each city or MPC and not easily applicable to another region. Seasonal variations of the radiation intensity influence the processing time as well as passing weather systems. The short time scales on which the precursor gases transform make in situ observations difficult as the aircraft can measure city or MPC plumes usually only after longer processing times. In addition, it is complicated to follow a plume directly after emission and exactly along its processing axis which would make the results clearer and more robust. Nevertheless, it is important to enhance the basic understanding of aerosol processing as it is substantial not only in terms of air quality but could also effect cloud properties by enlarging the CCN number.

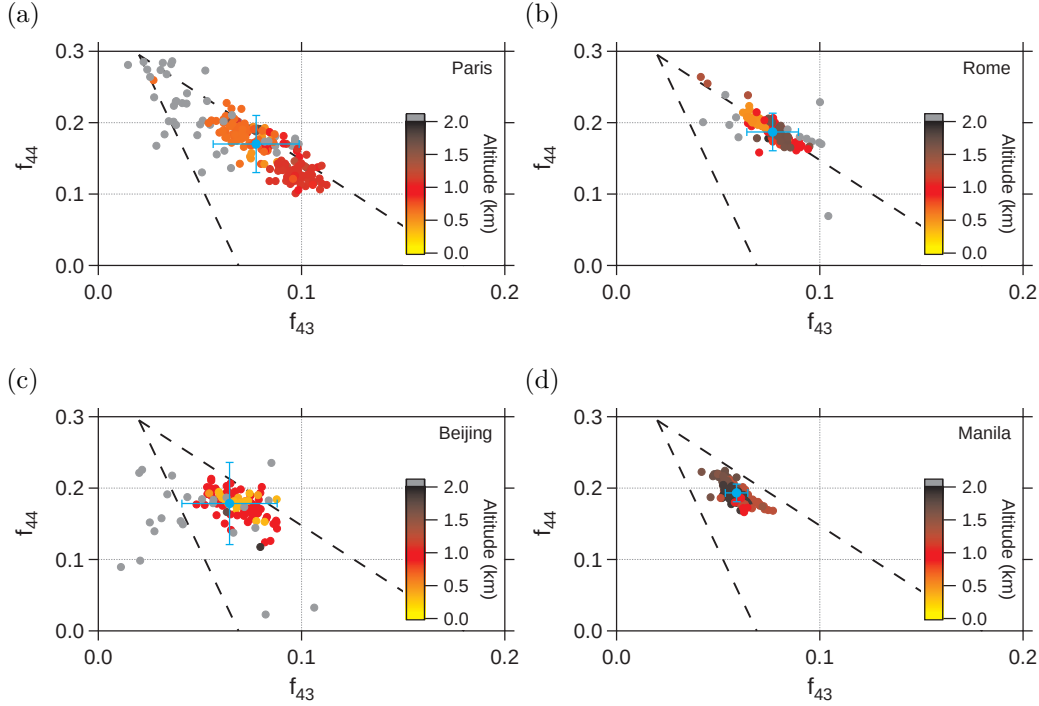


Figure 57: Arrangement of the data points in the ageing plot after Ng et al. (2010, 2011) from four selected MPCs during EMeRGe-Europe and -Asia: Paris (a), Rome (b), Beijing (c), and Manila (d). The data are colour-coded with altitude whereby the altitudes greater 2km are greyed out to highlight values close to the ground. The blue cross indicates the mean value of all data points with the corresponding standard deviations as error bars.

4.3.3 Oxidation state of OA from selected MPCs in Europe and East Asia

In this section, the oxidation state of OA associated with eight different MPCs in Europe and East Asia each is analysed. The data are filtered for the individual MPCs using the CO uptake mechanism according to Förster et al. (2023) which was already applied in Sect. 4.1.5. First, the distribution of the data from selected MPCs within the aerosol ageing triangle after Ng et al. (2010, 2011) is shown in Fig. 57. Then, the location of the MPC averages inside the triangle is compared for Europe and East Asia (see Fig. 58a and b). Lastly, the previously introduced trace gas ageing metrics are related to the aerosol oxidation state (see Fig. 58a-f).

The distribution of the selected MPC data in the ageing triangle differ (see Fig. 57), both in spread of the data points and location of the mean values as well as between the regions Europe and East Asia. For instance, the Paris and Beijing data cover a broader range inside the triangle and the points are more scattered than for Rome and Manila (see Fig. 57). This indicates that the Paris and Beijing aerosol particles include a larger mixture of different ages than Rome and Manila where the variety of aerosol ageing states is

smaller. In the case of Paris and Rome, a further explanation could be that air masses originating in Paris are encountered during six flights while air masses with contact to the BL over Rome are measured only during four flights (Förster et al., 2023). Thus, the current meteorological conditions on the measurement days may have changed more for Paris resulting in a broader f_{44} range. Data measured at altitudes greater 2 km are greyed out. Those are often the points dispersing the most, e.g. for Paris and Beijing (see Fig. 57a and c). This is in most cases related to longer transport times and mixing with potentially cleaner background air as already mentioned in Sect. 4.3.1. Furthermore, the same oxidation state of OA is observed in different altitudes, like for Manila (see Fig. 57d), due to different travel times of air masses or SOA formation followed by oxidation during transport. The f_{44} ranges of the other European MPCs are comparable to the ones of Paris (see Fig. 74 in the appendix). However, the PoV data is more similar to Rome with a narrower arrangement and the main number of points for Munich accumulate at a larger f_{43} than the other European MPCs (see Fig. 74e and f in the appendix). The Madrid emissions are lifted up and also transported above 2 km to the measurement location (Andrés Hernández et al., 2022). Therefore, many points are greyed out in the triangle plot of Madrid (see Fig. 74d in the appendix). The scatter of the other six East Asian MPCs is even higher than for Beijing as their data fill the complete space between the dashed lines of the triangle of a certain f_{44} range (see Fig. 75 in the appendix). As mentioned in Sect. 4.1.5, the MPCs Bangkok, Osaka, PRD, and Tokyo are probed rarely during the campaign which might explain why no clear signature of accumulated data developed. On the contrary, air masses with contact to the BL over Taipei are measured during every flight, in different altitudes, and varying distances to the MPC, as the base airport during the campaign is in the south of Taiwan. Therefore, the f_{44} includes a variety of values (see Fig. 75d in the appendix).

The data of the European MPCs are shifted towards the right dashed line of the triangle while the East Asian MPC data scatters more in between the two lines. This is also reflected by their averages and standard deviations (see Fig. 58a and b). More precisely, the mean values of the European MPCs gather around a f_{44} of 0.18 close to the right dashed line bordering the triangular space (see Fig. 58a). However, the mean for Munich is an exception with a less oxidised f_{44} of around 0.14 due to a closer measurement distance than for the other MPCs. The most oxidised averaged OA value during EMeRGe-Europe is encountered for the MPC Rome. In addition, the other Southern MPCs Barcelona, Madrid, and the PoV cluster around a slightly higher f_{43} than the cluster of the Northern and Central MPCs BeNeRu, London, and Paris while having almost the same f_{44} (see Fig. 58a). This implies a differing contribution of individual organic components between the areas in Europe.

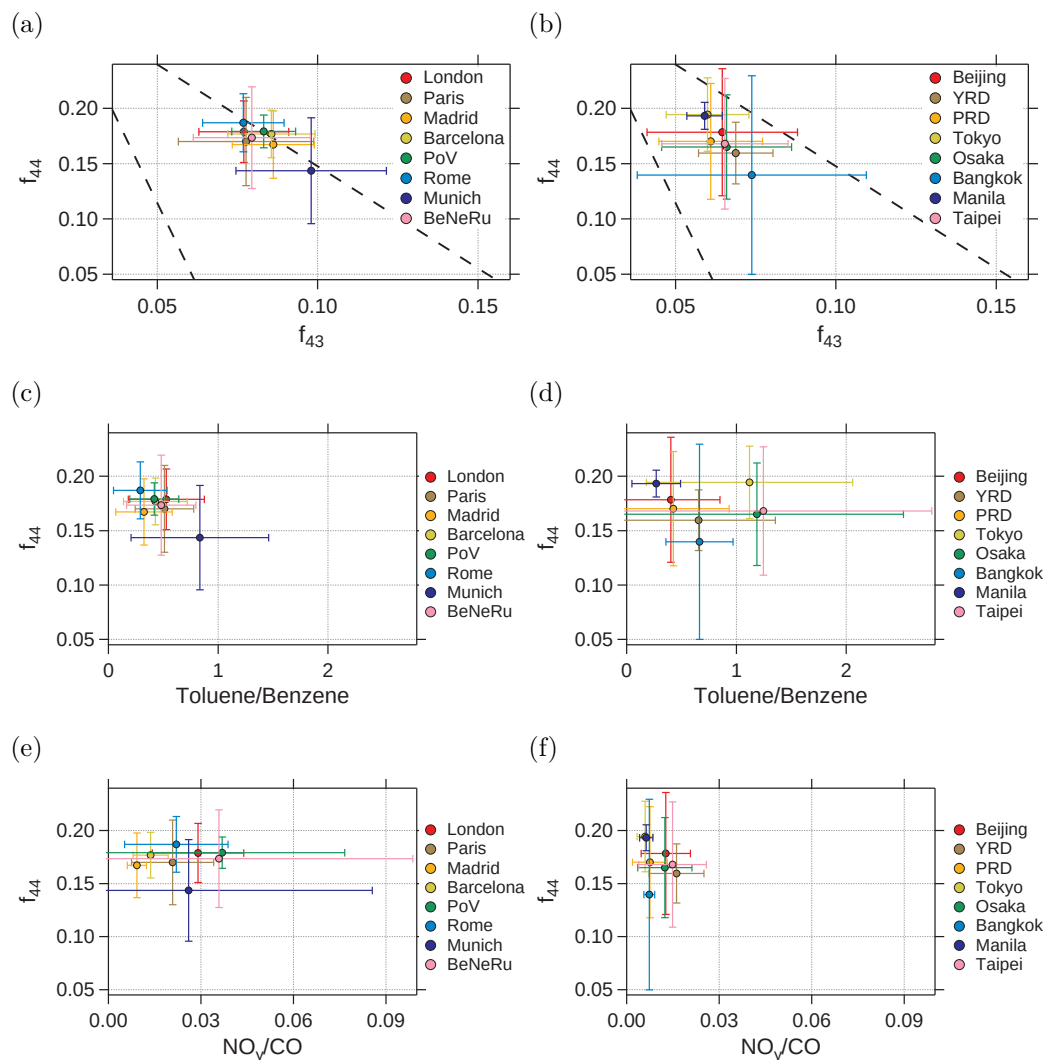


Figure 58: Arrangement of the mean values and corresponding standard deviations for the selected MPCs during EMeRGe-Europe and -Asia related to different ageing metrics: the ageing plot after Ng et al. (2010, 2011) (a) & (b), f_{44} versus the toluene to benzene ratio (c) & (d), and f_{44} versus the NO_y to CO ratio (e) & (f). The axes are aligned for both campaigns to make the plots easily comparable. Furthermore, the axes are adjusted so that the data section is better recognisable. Photochemical age increases from the bottom right to the upper left.

In contrast, the average values of the East Asian MPCs are located in the middle of the triangle (see Fig. 58b). They cover a comparable f_{44} range than the European MPCs although the distance between maximum and minimum is slightly enlarged. Furthermore, the mean values do not accumulate at one main location as for the European MPCs which indicates a more differentiated picture of the individual oxidation states. The least oxidised average is measured for Bangkok (see Fig. 58b). However, the corresponding standard deviations are large and the result might be biased by insufficient statistics. The most processed OA averages during EMeRGe-Asia are encountered for the MPCs Manila and Tokyo followed by Beijing and the PRD. The oxidation state of OA from Taipei is located in the middle of the East Asian data (see Fig. 58b).

The VOC-based ageing metric shows similarities and differences with the results of the OA oxidation (see Fig. 58c and d). For instance, the averages of Rome and Manila are indicated as the most processed air masses by the toluene to benzene ratio while the Munich mean is the least processed which is similar to the OA processing (see Fig. 58a and c). Furthermore, the mean values of the South European MPCs Barcelona and PoV as well as the North European MPCs BeNeRu, London, and Paris are also grouped together. In addition, the accumulation of MPCs is valid for the East Asian MPCs Beijing and the YRD as well as Osaka and Taipei (see Fig. 58b and d). Furthermore, the order of aerosol ageing from fresh to more aged emissions is comparable between the metrics for the MPCs means below a toluene to benzene ratio of one. On the contrary, differences occur for the processing of the Madrid mean which has the second smallest VOC ratio, indicating an old and processed air mass, while the f_{44} provides the second lowest value and thus, point to less oxidised OA (see Fig. 58a and c). However, the deviations between the two ageing metrics are larger for the East Asian data (see Fig. 58b and d). First, toluene to benzene ratios greater than one indicate fresh emissions like for Taipei, Tokyo, and Osaka while the f_{44} showed no freshly emitted aerosol particles. Second, air masses from Tokyo are indicated as fresh emissions while the aerosol particles are among the most oxidised. Lastly, air masses of Bangkok and the YRD have the same processing state related to the VOCs while their f_{44} differs. The toluene to benzene ratios further depend on the emission strength at the sources which differ between Europe and East Asia.

The third ageing metric applied to the averaged MPCs data is the ratio of NO_y to CO (see Fig. 58e and f). Based on this metric, the order of the EMeRGe-Europe data is completely different than for the OA oxidation. The MPCs BeNeRu and PoV have the highest anthropogenic signature followed by London and Munich (see Fig. 58e). In contrast, the Spanish MPCs Barcelona and Madrid show the lowest values. Except for the Bangkok average, the ageing structure of the East Asian MPC averages is similar to the OA oxidation (see Fig. 58b and f). The general difference for the NO_y to

CO ratios between the regions Europe and East Asia is remarkable. The averaged ratios are higher in Europe than in East Asia as well as the corresponding standard deviations which is mainly caused by the significantly higher CO VMRs in East Asia (see Sect. 4.1.3).

In conclusion, a variety of organic aerosol oxidation states was observed between the different MPCs as well as for each individual MPC albeit no freshly formed OA was measured. A clear difference in the oxidation characteristics of OA was determined between Europe and East Asia as the MPC averages are located in different places within the ageing triangle. On the contrary, the ratio of toluene to benzene also identified fresh emissions in East Asia while the NO_y to CO ratio had higher maximum values in Europe.

4.3.4 Comparison with literature

The aerosol composition around the world was reviewed by Zhang et al. (2007a) comparing urban and rural/remote ground-based sites. The total PM_1 mass concentration at urban sites was highest in Beijing with $72 \mu\text{g m}^{-3}$ in July 2006 while $13 \mu\text{g m}^{-3}$ were measured in Tokyo in January/February 2004 and around $13 \mu\text{g m}^{-3}$ at other remote sites in East Asia. In comparison, total mass concentrations of up to $8 \mu\text{g m}^{-3}$ were encountered during flight E-AS-04 during EMeRGe-Asia at some distance to the sources. Zhang et al. (2007a) listed higher fractions of NO_3 between 21 and 24 % at urban sites like Beijing and Tokyo while the dominant aerosol species in Beijing was OA with up to 35 %, followed by SO_4 with 26.4 %. The NH_4 showed around 17 % and chloride made the smallest proportion with 1.4 %. The aerosol composition is comparable to the anthropogenically influenced flight sections of flight E-AS-04 like the "north" segment yielding almost similar shares except for NO_3 which was lower with around 14 % and SO_4 which had been higher with up to 30.5 %. For rural/remote sites in East Asia like Jeju Island (South Korea) or Fukue Island (Japan), the composition was characterised by less NO_3 with fractions around 7 %, more OA and SO_4 with around 40 and 37 %, and almost no chloride while the NH_4 proportion remained the same (Zhang et al., 2007a). This change in composition was also observed during flight E-AS-04 for the flight sections east of Taiwan or at higher altitudes. A main contributor to increasing PM_1 mass concentration under highly polluted conditions in East Asia is NO_3 which is favoured by strong photochemical chemistry and higher precursor concentrations of nitrate (such as NO_2 , N_2O_5 , NH_3) during polluted periods (Chen et al., 2021b). Furthermore, increased oxidants of O_3 , SO_4 , aerosol liquid water content, and particle acidity enhance NO_3 formation.

Secondary aerosol formation was observed in connection with increasing photochemical ageing time in Beijing in summer 2019 (Chen et al., 2021a). The age was defined based on toluene to benzene ratios, their reaction rates,

and the OH concentration. Photo-oxidation of VOCs due to strong solar radiation lead to more oxidised OA and a formation of SOA being an important factor in summertime pollution.

Based on Stohl et al. (2002), most of the NO_y was removed already when travelling through the BL while only 3 - 5 % entered altitudes above 3 km. The removal process was slower during the first 2 d than afterwards which also applies to the data in Fig. 56c as the slope for the "north" segment is lower than for the other segments west of Taiwan. Furthermore, Stohl et al. (2002) observed NO_y to CO ratios of 0.19 and 0.23 for emissions encountered over the North Atlantic in spring 1996 and fall 1997. The ratio dropped to below 0.01 for air masses older than four days. After 5 to 6 d, the anthropogenically NO_y emitted at ground was reduced to around 50 % becoming equal to NO_y formed in situ. Anthropogenic emissions in Pennsylvania in summer 1988 and Colorado in spring/summer 1989 had NO_y to CO ratios of 0.06 to 1.2 (Parrish et al., 1991). Zellweger et al. (2003) measured at the research site Jungfraujoch in Switzerland for about 2 years. They defined a pollution event for NO_y to CO ratios greater 0.01 and found a peak of the aerosol surface area during pollution events. Under undisturbed conditions in the free troposphere, the ratio was lowest indicating advanced ageing of the air masses. Additionally, they investigated the seasonal variation of the ratio which was lower in winter due to a longer CO lifetime. To conclude, minimum NO_y to CO ratios of 0.01 were also observed in anthropogenically influenced air masses during EMeRGe-Europe in accordance with the literature. Moreover, the EMeRGe-Asia data showed an slightly smaller minimum ratio of 0.006 averaged for the MPC Tokyo. Furthermore, the dependency of air mass ageing on altitude was observed in a case study around Taiwan as measurements at higher altitudes showed smaller NO_y to CO ratios.

CONCLUSIONS AND OUTLOOK

Major population centres (MPCs) are large pollution sources affecting the air quality and human health as well as the surrounding environment and global climate. Thus, this thesis aimed for a better understanding of aerosol composition characteristics and transformation processes in the vicinity of MPCs. Therefore, two large airborne campaigns were carried out within the mission Effect of Megacities on the Transport and Transformation of Pollutants on the Regional to Global Scales (EMeRGe) using the research aircraft HALO. They were conducted in summer 2017 over Europe and in spring 2018 over East Asia. Thereby, the non-refractory components of submicron aerosol particles were measured with the compact time-of-flight aerosol mass spectrometer (C-ToF-AMS). In the following, the main findings of this thesis are summarised with respect to the questions stated in Sect. 1.3. In addition, an outlook is given providing ideas for future studies on MPC emissions.

Aerosol particle composition in Europe and East Asia

This study found larger absolute levels of submicron particulate matter in East Asia with up to $34 \mu\text{g m}^{-3}$ compared to Europe with around $18 \mu\text{g m}^{-3}$. Furthermore, the aerosol composition changed with increasing total mass concentration. For instance, nitrate aerosol (NO_3) became more important in highly polluted air masses and accounted for 27 - 30 % in the highest concentration subset. The ammonium aerosol (NH_4) fraction also increased with total mass to proportions of 15 - 18 %. In contrast, organic aerosol (OA) and sulfate aerosol (SO_4) fractions decreased with increasing pollution level. The average aerosol composition in Europe was dominated by OA with almost 55 %, followed by SO_4 with up to 22 % while NH_4 and NO_3 had comparable fractions of 11 - 12 %. In contrast, the average East Asian aerosol contained mainly SO_4 with up to 38 %, followed by around 35 % OA, and a few percent more NH_4 than NO_3 . Average chloride mass fractions had been below 1 % during both campaigns.

The measurements covered altitudes ranging from below 1 km up to 10 km in Europe and up to 14 km in East Asia. Increased aerosol mass concentrations were observed below 3 - 4 km in Europe and 4 - 5 km in East Asia although polluted layers also occurred occasionally at greater heights. The maximum mass concentrations were not always measured close to the surface but at higher altitudes, like the emission plume of Madrid which was

measured over the Mediterranean Sea or the Pearl River Delta (PRD) and Yangtze River Delta (YRD) outflows which were observed in the Gulf of Thailand. However, emissions from e.g. the Po Valley (PoV) remained local and decreased the air quality in Northern Italy. The averaged vertical profiles showed differences between the European and East Asian data. Higher SO_4 mass concentrations and fractions were measured in the East Asian (planetary) boundary layer (BL) than over Europe. The same was observed for NH_4 mass concentrations and fractions although the differences between the regions were less pronounced. In contrast, the averaged organic mass concentrations and fractions in the BL were found to be higher in Europe than in East Asia.

The high pollution levels in East Asia were also confirmed by carbon monoxide (CO) volume mixing ratios (VMRs) which showed a maximum of almost 2000 ppbv in East Asia. This was an order of magnitude greater than the CO maximum in Europe with around 320 ppbv. However, the majority of European data accumulated below 150 ppbv. During both campaigns, the OA mass concentrations were often highly correlated with the CO VMRs indicating that anthropogenic and combustion-related emissions were driving the OA mass concentrations.

Aerosol-specific emission signatures for the selected MPCs

The emission plumes of selected MPCs were probed at various distances and altitudes during the EMeRGe campaigns. Emissions originating from those MPCs were identified by applying a simple CO uptake mechanism according to Förster et al. (2023). The attributed emissions were encountered close to the source as well as further away as the air masses were transported over greater distances. In addition, pollutants of different origins mixed at the same location, e.g. emissions from London and the Belgium-Netherlands-Ruhr area (BeNeRu) were measured simultaneously over the Southern Bight of the North Sea. The analysis showed characteristic differences between the individual MPCs in Europe and East Asia. The total aerosol mass concentrations varied for the single MPCs and were generally higher in East Asia. Moreover, a meridional differentiation was observed. The Southern MPCs Barcelona, Rome, Madrid, and the PoV had the highest total mass concentrations in Europe of $3.5 - 6 \mu\text{g m}^{-3}$, followed by the Northern MPCs London and the BeNeRu with $3 - 3.5 \mu\text{g m}^{-3}$, whereas the Central European MPCs Munich and Paris had the lowest total mass concentrations of $1.5 - 2 \mu\text{g m}^{-3}$. This difference was also reflected in the NO_3 fraction which increased from south to north while the OA fraction decreased. In East Asia, the largest total aerosol mass concentrations were observed for the MPCs Beijing and the YRD. The OA fraction also decreased from south to north while the SO_4 fraction increased. Furthermore, the relative aerosol composition differed be-

tween Europe and Asia: In Europe, OA was the main aerosol component of the MPCs aerosol composition while SO_4 dominated in East Asia. The relative contributions of NH_4 , NO_3 , and chloride were comparable between the two regions whereas refractory black carbon (rBC) fractions in East Asian MPCs were more than twice as high as in Europe.

Emission sources controlling the aerosol acidity in the vicinity of MPCs

This study found that aerosol particles were in general neutralised. However, the aerosol particles tended to be more acidic in the upper troposphere and lower stratosphere. Another exception to the neutralised particles were the data below 1.75 km during EMeRGe-Asia where acidic aerosol was observed. Three example periods were investigated and identified different sulfur dioxide (SO_2) emission sources determining the aerosol acidity. These were volcanic outgassing, ship exhaust gases as well as anthropogenic MPC emissions and emissions from copper smelters. These examples showed that local pollutants affected the surrounding area but were also transported over a long distance where they influenced aerosol particles far away from their source. In this context, different conversion states of sulfur were observed.

Ageing effect on the aerosol particle composition in the outflow of MPCs

Mass spectral marker peaks and peak ratios were used to distinguish between fresh and aged OA components. It was found that fresh plumes and background conditions could clearly be separated by this method while a large variability in the degree of oxygenation was observed in the background aerosol. In a detailed case study of emissions around Taiwan, different levels of processing could be inferred as the degree of oxidation of OA increased with distance to the source. In addition, secondary aerosol formation was observed. Furthermore, the particle number concentration, rBC and trace gases showed higher values in anthropogenically influenced air masses. The degree of oxygenation of the aerosol was related to the mean age derived from dispersion model calculations as well as to the chemical plume age derived from volatile organic compounds (VOCs) and the NO_y to CO ratio. However, the latter two did not give exact numbers, but a division into fresh and aged/diluted air masses. For the case study around Taiwan, the results of the different processing methods were comparable for the flight sections below 1 km which encountered the largest local pollution. The dispersion model gave an evolution of mean ages between 55 to 75 h for those flight sections. However, an assignment of a specific plume age to a distinct oxidation state turned out to be difficult due to the complexity of the data.

After applying the MPC filter according to Förster et al. (2023), a variety of OA oxidation states was observed for the different MPCs. For instance,

the OA in air masses from Munich was the least oxidised in Europe while pollutants from the MPC Rome were the most oxidised. This was also observed by the VOC processing results. In contrast, the VOC processing method identified fresher emissions in East Asia than Europe which might be related to higher emission strengths and different photochemical conditions. In addition, differences for European and East Asian MPC emissions were observed as the oxidation states are located in different places within the ageing triangle. More precisely, although the oxidation states of the two regions covered a comparable range, the East Asian averages were located in the middle of the ageing triangle while the European averages were shifted to the right boarder of the triangle.

In conclusion, the aerosol particle composition and different aerosol properties were examined in air masses over Europe and East Asia. It was shown that pollutants accumulated locally but were also transported over longer distances where they mixed with natural emissions. During transport, the aerosol particles aged and altered their composition which can have further effects on air quality and their ability to act as cloud condensation nuclei. It was observed that the urban emissions remained mainly in the BL. As it is predicted that cities will continue to grow, anthropogenic emissions will increase. This needs to be taken into account in air pollution reduction strategies. Therefore, further studies on the distribution of emissions as well as their transformation along transport are needed. This will help to understand the involved processes and to revise existing air pollution control strategies and to develop new policies. During EMeRGe, no freshly formed OA was observed. Therefore, measurements closer to the source would be necessary to complete the picture of aerosol particle evolution. In addition, ground-based measurements are important to quantify the emission strengths and to calculate ageing times which further requires OH measurements. Moreover, follow-up experiments should have the potential to better track an emission plume in the BL which requires an aircraft that is able to fly slowly at low altitudes. A combined experiment with similar payloads on two aircraft and ground sites would improve the capabilities to study the plume evolution in the atmosphere. Although aircraft measurements are always restricted to a certain time and location, they provide an essential input for atmospheric models which can be used to quantify the regional and hemispheric impact of MPC emissions.

Part II

APPENDIX



SUPPLEMENTARY INFORMATION TO CHAPTER 2

A.1 INSTRUMENTATION

Table 12: Overview of the scientific measurement instruments used during both EMeRGe campaigns, explaining their acronyms, measured species and parameters, and giving further references. The abbreviations for the institutions are explained in the list of acronyms in the appendix from page 218ff.

ACRONYM	INSTRUMENT	SPECIES/ PARAMETERS	INSTITUTION	REFERENCE
AENEAS	<u>A</u> tmospheric <u>N</u> itrogen <u>O</u> xides <u>M</u> easuring <u>S</u> ystem	NO, NO _y	DLR-IPA	Ziereis et al. (2004)
AMETYST	<u>A</u> erosol <u>M</u> easurement <u>S</u> ystem	Fine aerosol number concentration, size distribution, and volatility	DLR-IPA	Andreae et al. (2018)
AMTEX (with PERTRAS)	<u>A</u> tmospheric <u>T</u> race <u>E</u> xperiment (<u>P</u> erfluorocarbon <u>T</u> racer <u>S</u> ystem)	CO, O ₃ , (PFCs)	DLR-IPA	Gerbig et al. (1996), Ren et al. (2015)
BAHAMAS (with SHARC)	<u>B</u> asic <u>H</u> ALO <u>M</u> easurement and <u>S</u> ensor <u>S</u> ystem (Sophisticated <u>H</u> ygrometer for <u>A</u> tmospheric <u>R</u> esear <u>C</u> h)	Basic aircraft position and meteorological data (water vapour)	DLR-FX	Mallaun et al. (2015)
CATS	Picarro greenhouse gas analyser, SO ₂ fluorescence analyser, tedlar bag sampling system	CO ₂ , CH ₄ , H ₂ O, SO ₂ , δ ¹³ C	DLR-IPA	Chen et al. (2010), Volz- Thomas et al. (2002), Fisher et al. (2006)
CCN (with SP2)	<u>C</u> loud <u>C</u> ondensation <u>N</u> uclei Rack (<u>S</u> ingle <u>P</u> article <u>S</u> oot <u>P</u> hotometer)	Cloud condensation nuclei concentration, soot, microscopic properties	MPIC	Holanda et al. (2020), Wendisch et al. (2016)

Table 12: (continued)

ACRONYM	INSTRUMENT	SPECIES/ PARAMETERS	INSTITUTION	REFERENCE
CI-ITMS	<u>C</u> hemical <u>I</u> onization <u>I</u> on <u>T</u> rap <u>M</u> ass <u>S</u> pectrometer	SO ₂ , formic acid	DLR-IPA	Speidel et al. (2007)
C-ToF-AMS (with OPC)	<u>C</u> ompact <u>T</u> ime-of- <u>F</u> light <u>A</u> erosol <u>M</u> ass <u>S</u> pectrometer (<u>O</u> ptical <u>P</u> article <u>C</u> ounter)	Aerosol mass concentration (particle number concentration)	MPIC, Univ. Mainz	Schulz et al. (2018), GRIMM (2008)
FAIRO	<u>F</u> ast <u>A</u> irborne <u>O</u> zone <u>M</u> onitor	O ₃	KIT	Zahn et al. (2012)
HAIDI	<u>H</u> eidelberg <u>A</u> irborne <u>I</u> maging <u>D</u> ifferential <u>O</u> ptical <u>A</u> bsorption <u>S</u> pectroscopy <u>I</u> nstrument	2-D trace gas slant column densities of NO ₂ , SO ₂ , CH ₂ O, C ₂ H ₂ O ₂ , H ₂ O, BrO, O ₃	Univ. of Heidel- berg	General et al. (2014)
HALO-SR	<u>H</u> ALO <u>S</u> olar <u>R</u> adiation <u>I</u> nstrument	Spectral actinic flux density (up/down)/photolysis frequencies	FZ Jülich	Bohn and Lohse (2017)
HKMS	<u>H</u> ALO <u>K</u> arlsruhe <u>M</u> ass <u>S</u> pectrometer	VOCs	KIT	Brito and Zahn (2011)
mini-DOAS	<u>m</u> ini <u>D</u> ifferential <u>O</u> ptical <u>A</u> bsorption <u>S</u> pectroscopy <u>I</u> nstrument	NO ₂ , HONO, BrO, CH ₂ O, C ₂ H ₂ O ₂ , C ₃ H ₄ O ₂ , SO ₂ , IO	Univ. of Heidel- berg	Hüneke et al. (2017)

Table 12: (continued)

ACRONYM	INSTRUMENT	SPECIES/ PARAMETERS	INSTITUTION	REFERENCE
MIRAH	<u>M</u> ea <u>s</u> urement of <u>S</u> t <u>a</u> ble <u>I</u> sotope <u>R</u> atios in <u>A</u> tmospheric <u>T</u> race <u>G</u> ases on <u>H</u> ALO	Whole air sampler (VOCs, carbon isotope ratios)	Univ. of Wup- pertal	Wintel et al. (2013)
PAN-GC	<u>P</u> eroxyacetyl <u>N</u> itrate <u>G</u> as <u>C</u> hromatograph	PAN	DLR-IPA	
PeRCEAS	<u>P</u> eroxy <u>R</u> adical <u>C</u> hemical <u>E</u> nhancement and <u>A</u> bsorption <u>S</u> pectrometer	Sum of peroxy radicals HO ₂ and RO ₂	University of Bremen	George et al. (2020)

A.2 PARTICLE LOSS CALCULATION

Table 13: Overview of the used parameters for the particle loss calculator (after von der Weiden et al. (2009)). The flow values for the first four sections can be found in Table 14 and are mainly dependent on the flight altitude due to changing bypass flows with height. Incl.=Inclination.

TUBING SECTION	FLOW (L min^{-1})	TUBE LENGTH (m)	1. & 2. INNER DIAMETER (mm)	INCL. ANGLE ($^{\circ}$)	BEND ANGLE ($^{\circ}$)
1 (AMS, OPC, Bypass)	value	0.08	5	0	90
2 (AMS, OPC, Bypass)	value	0.165	5	75	90
3 (AMS, OPC, Bypass)	value	0.11	5	90	0
4 (AMS, OPC, Bypass)	value	0.12	5	90	0
5 (AMS, OPC)	1.3	0.54	5	70	90
6 (AMS, OPC)	1.3	0.35	5	80	0
7 (AMS, OPC)	1.3	0.33	5	50	120
8 (AMS, OPC)	1.3	0.35	5	75	150
9 (AMS, OPC)	1.3	0.26	5	50	140
10 (AMS, OPC)	1.3	0.135	5	75	90
11 (AMS, OPC)	1.3	0.23	5	0	90
12 (AMS, OPC)	1.3	0.52	5	90	90
13 (AMS, OPC)	1.3	0.1	5	80	0
14 (AMS, OPC)	1.3	0.2	5	90	90
15 (AMS, OPC)	1.3	0.04	5	0	0
16 (AMS)	0.1	0.09	5	0	0

Table 14: Minimum and maximum total flow (in L min^{-1}) at ground level and maximum altitude for both campaigns, depending on a changing bypass flow that is proportional to height.

	LOWER LEVEL		MAX. ALTITUDE	
	min.	max.	min.	max
E-EU	1.95	2.1	2.47	2.93
E-AS	1.97	2.08	2.32	2.97

A.3 FLIGHT PLANNING

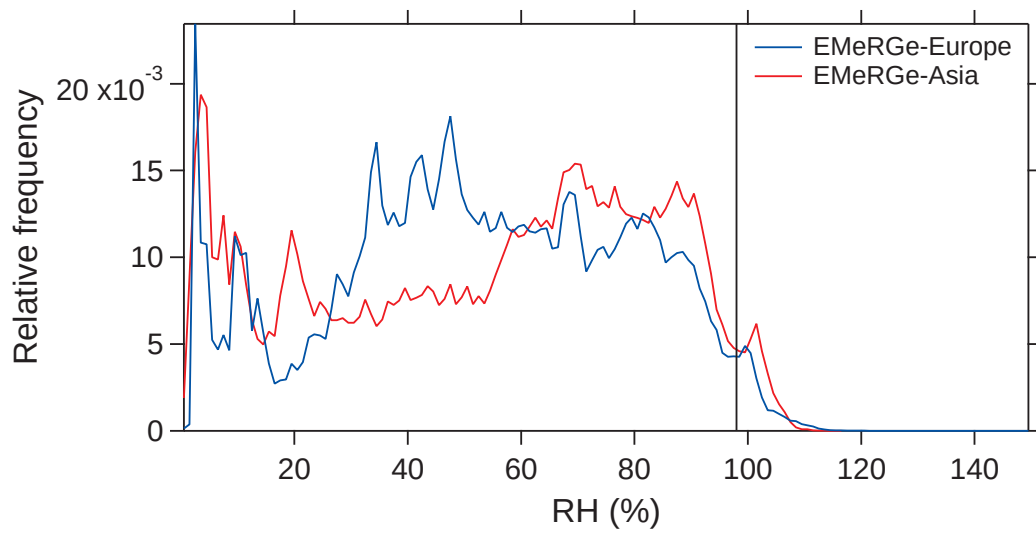


Figure 59: Relative frequency of RH values during EMeRGe-Europe and -Asia, considering all flights for the European part and flights E-AS-03 to E-AS-14 for the Asian part of the project.

SUPPLEMENTARY INFORMATION TO CHAPTER 3

B.1 CALIBRATIONS

For the flow calibration, the lens pressure was gradually adjusted with the control software of the CPI. The flowmeter measures the flow in L min^{-1} . This has to be converted to $\text{cm}^3 \text{s}^{-1}$ as follows:

$$1 \text{ cm}^3 \text{s}^{-1} = 1 \text{ L min}^{-1} * 1000/60 \quad (13)$$

The flowmeter manual (TSI, 2022) gives an error for the flow of either 2 % of the measured value or 0.005 L min^{-1} , whichever is the greater. The fixed value was converted to $0.083 \text{ cm}^3 \text{s}^{-1}$ and used for the error bars in Fig. 8.

Table 15: Flow calibration values during the EMERGe-Asia campaign.

LENS PRESSURE (TORR)	INLET FLOW, [*] FLOWMETER (L min^{-1})	CONVERTED FLOW ($\text{cm}^3 \text{s}^{-1}$)	FLOW RATE, DAQ (V)
1.9	0.079	1.317	1.81
1.8	0.068	1.133	1.72
1.7	0.058	0.967	1.62
1.6	0.048	0.800	1.52
2.0	0.088	1.467	1.91
2.1	0.100	1.667	2.01
2.2	0.113	1.883	2.11
1.9	0.078	1.300	1.81

^{*} The flowmeter offset of 0.01 L min^{-1} is already subtracted.

Table 16: Values from the ionisation efficiency (IE) calibrations used for EMeRGe-Europe in 2017. Bold text highlights the used ones. The last column contains the number of final particles left for each calibration to have some statistic.

DATE	d_{mob} (nm)	SI (bits*ns)	IE	AB (Hz)	FLOW RATE (cm ³ s ⁻¹)	FINAL PARTICLES
30.06.	200	11.7	1.29e-06	9.81e+05	1.33	29
12.07.	300	12.4	8.56e-07	8.75e+05	1.33	60
	350		6.97e-07	8.66e+05	1.33	23
	mean		7.76e-07	8.7e+05	1.33	
18.07.	350	16.6	5.38e-07	6.03e+05	1.33	200
	300		5.9e-07	6.06e+05	1.33	200
	mean		5.64e-07	6.05e+05	1.33	
04.08.	300	16.5	4.11e-07	3.35e+05	1.33	194
	350		4.25e-07	3.32e+05	1.33	198
	mean		4.18e-07	3.33e+05	1.33	
Mean (all)		14.3	7.62e-07	6.97e+05	1.33	

Table 17: Values from the *IE* calibrations used for EMeRGe-Asia in 2018. Bold text highlights the used ones. The last column contains the number of final particles left for each calibration to have some statistic.

DATE	d_{mob} (nm)	SI (bits*ns)	IE	AB (Hz)	FLOW RATE (cm ³ s ⁻¹)	FINAL PARTICLES
01.03.	350	13.8	9.84e-07	7.37e+05	1.33	30
	300		1.16e-06	7.67e+05	1.33	105
	mean		1.07e-06	7.52e+05	1.33	
18.03.	350	13.8	1.01e-06	6.97e+05	1.33	157
	300		1.07e-06	7.17e+05	1.33	268
	mean		1.04e-06	7.07e+05	1.33	
25.03.	350	14.3	9.06e-07	6.37e+05	1.33	161
	300		9.94e-07	6.2e+05	1.33	234
	mean		9.5e-07	6.28e+05	1.33	
05.04.	350	13.9	8.55e-07	5.24e+05	1.33	199
	300		9.95e-07	5.29e+05	1.33	223
	mean		9.25e-07	5.26e+05	1.33	
07.06.	350	14.3	7.52e-07	6.22e+05	1.31	241
	300		8.37e-07	6.2e+05	1.31	262
	mean		7.95e-07	6.21e+05	1.31	
Mean (all)		14.02	9.56e-07	6.47e+05	1.33	

B.2 PERFORMANCE DURING EMERGE

Table 18: Overview of the flight duration, filter measurement times, data gaps, and calculated data coverage for each flight during EMeRGe in Europe.

FLIGHT	DURATION (MIN)	FILTER (MIN)	GAPS (MIN)	COVERAGE (%)
E-EU-03	393	9	0	97.71
E-EU-04	262	12	0	95.42
E-EU-05	479	13	0	97.29
E-EU-06	508	11	0	97.83
E-EU-07	514	13	117.5	74.61
E-EU-08	457	15	0	96.72
E-EU-09	515	20	0	96.12
Sum	3128	93	117.5	93.27

Table 19: Overview of the flight duration, filter measurement times, data gaps, and calculated data coverage for each flight during EMeRGe in Asia.

FLIGHT	DURATION (MIN)	FILTER (MIN)	GAPS (MIN)	COVERAGE (%)
E-AS-01	474	18	0	96.20
E-AS-03	389	29	0	92.54
E-AS-04	516	11	0	97.87
E-AS-05	485	10	0	97.94
E-AS-06	409	11	0	97.31
E-AS-07	345	11	0	96.81
E-AS-08	532	10	0	98.12
E-AS-09	542	10	0	98.15
E-AS-10	519	13	0	97.50
E-AS-11	563	20	0	96.45
E-AS-12	360	10	0	97.22
E-AS-13	537	27	0	94.97
E-AS-14	460	32	0	93.04
E-AS-16	524	21	113	74.43
Sum	6655	233	113	94.80

B.3 BACKGROUND CORRECTION

Table 20: Overview of the applied background correction during EMeRGe in Europe. Only listing the affected flights and species.

FLIGHT	ORGANICS ($\mu\text{g m}^{-3}$)	NITRATE ($\mu\text{g m}^{-3}$)	SULFATE ($\mu\text{g m}^{-3}$)	CHLORIDE ($\mu\text{g m}^{-3}$)
E-EU-05	0.00975959	0.0118246	-	-
E-EU-08	-0.0282884	-0.0257428	-	-0.0204131
E-EU-09	-	-	-	-0.0352302

Table 21: Overview of the applied background correction during EMeRGe in Asia.

FLIGHT	ORGANICS ($\mu\text{g m}^{-3}$)	NITRATE ($\mu\text{g m}^{-3}$)	SULFATE ($\mu\text{g m}^{-3}$)	CHLORIDE ($\mu\text{g m}^{-3}$)
E-AS-01	0.105285	0.011218	-	-
E-AS-03	0.172217	0.0206014	-	-
E-AS-04	0.147421	0.156526	-	-
E-AS-05	0.160129	-	-	-
E-AS-06	0.0297836	-	-	-
E-AS-07	0.122651	0.0155836	0.0198399	-0.0302447
E-AS-08	0.137567	0.0142178	-	-
E-AS-09	0.104951	-	-	-
E-AS-10	0.0583435	-	-	-
E-AS-11	0.0601756	0.00985098	-	-
E-AS-12	0.108884	0.0130228	-	-
E-AS-13	0.178929	0.0162878	-	-
E-AS-14	0.135568	0.0144293	-	-
E-AS-16	0.241248	0.0202898	-	-

B.4 QUALITY ASSURANCE

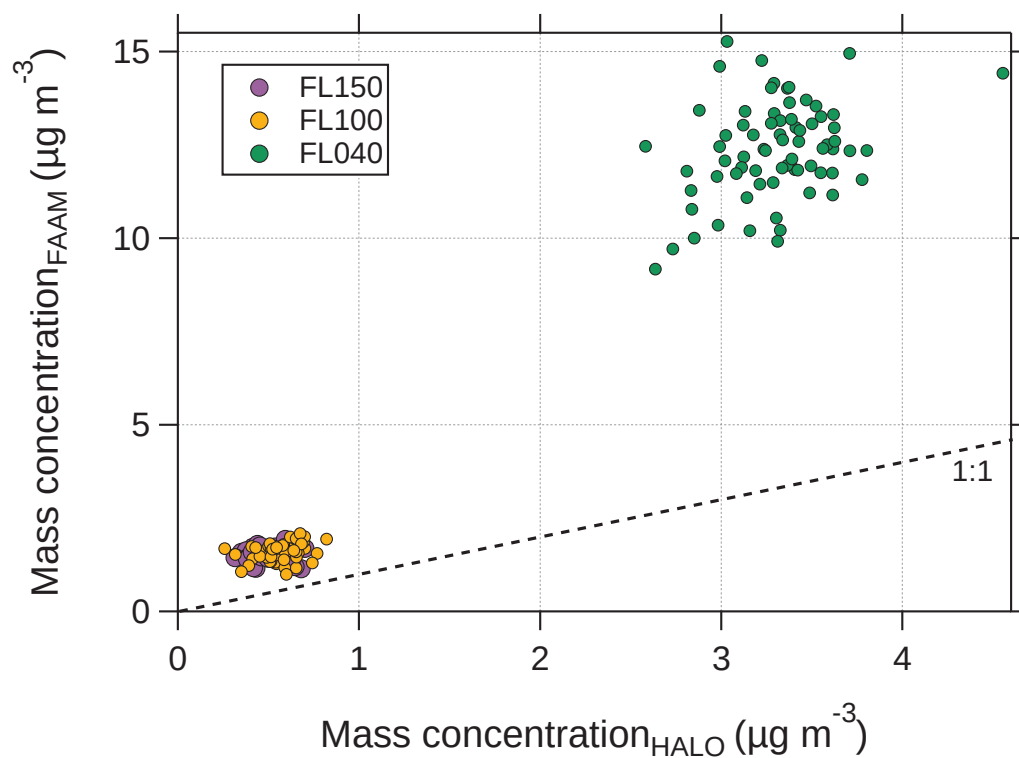


Figure 60: Correlation of the total aerosol mass concentration from the FAAM and HALO C-ToF-AMS for the three comparison altitudes. FAAM data (original, unrevised data set) were provided by University Manchester (Schumann, 2021). The data differ by a factor of 3.7, 2.7, and 2.9 on FL040, FL100, and FL150, respectively.

Table 22: Mean values of volume and mass concentrations for the three comparison altitudes. The calculated expected mass concentration is highlighted and contrasted with the measured values by the two C-ToF-AMSs.

	UNIT	FL150	FL100	FL040
SMPS volume conc.	$\mu\text{m}^3 \text{cm}^{-3}$	0.25	0.28	3.47
OPC volume conc. (up to 0.8 μm)	$\mu\text{m}^3 \text{cm}^{-3}$	0.1256	0.14	1.14
Total volume conc. dV (SMPS+OPC)	$\mu\text{m}^3 \text{cm}^{-3}$	0.375	0.42	4.61
Total mass conc. dM	$\mu\text{g m}^{-3}$	0.56	0.63	6.92
BC mass conc.	$\mu\text{g m}^{-3}$	0.006	0.009	0.07
Expected mass conc. (see Eq. 6)	$\mu\text{g m}^{-3}$	0.56	0.62	6.85
AMS (FAAM) mass conc.	$\mu\text{g m}^{-3}$	0.96	0.99	7.76
AMS (HALO) mass conc.	$\mu\text{g m}^{-3}$	0.51	0.56	3.3
AMS (FAAM) / expected mass		1.72	1.6	1.13
AMS (HALO) / expected mass		0.92	0.91	0.48

SUPPLEMENTARY INFORMATION TO CHAPTER 4

C.1 AEROSOL PARTICLE COMPOSITION IN THE VICINITY OF MPCs

Table 23: Relative shares (in %) of the average aerosol composition from C-ToF-AMS data as a function of total concentration for EMeRGe-Europe (a) and -Asia (b).

(a) EMeRGe-Europe

SPECIES	0-5 $\mu\text{g m}^{-3}$	5-10 $\mu\text{g m}^{-3}$	10-20 $\mu\text{g m}^{-3}$	20-35 $\mu\text{g m}^{-3}$	All
OA	54.45	57.48	42.66	-	54.84
NO ₃	8.65	11.78	26.96	-	11.31
SO ₄	24.58	18.84	14.64	-	21.43
NH ₄	11.97	11.33	15.15	-	11.94
Chloride	0.35	0.57	0.60	-	0.48

(b) EMeRGe-Asia

SPECIES	0-5 $\mu\text{g m}^{-3}$	5-10 $\mu\text{g m}^{-3}$	10-20 $\mu\text{g m}^{-3}$	20-35 $\mu\text{g m}^{-3}$	All
OA	39.67	30.42	29.51	32.08	35.30
NO ₃	4.67	13.71	23.06	29.38	10.66
SO ₄	41.30	37.33	28.14	19.84	37.62
NH ₄	14.03	17.75	18.35	17.56	15.84
Chloride	0.34	0.79	0.94	1.16	0.57

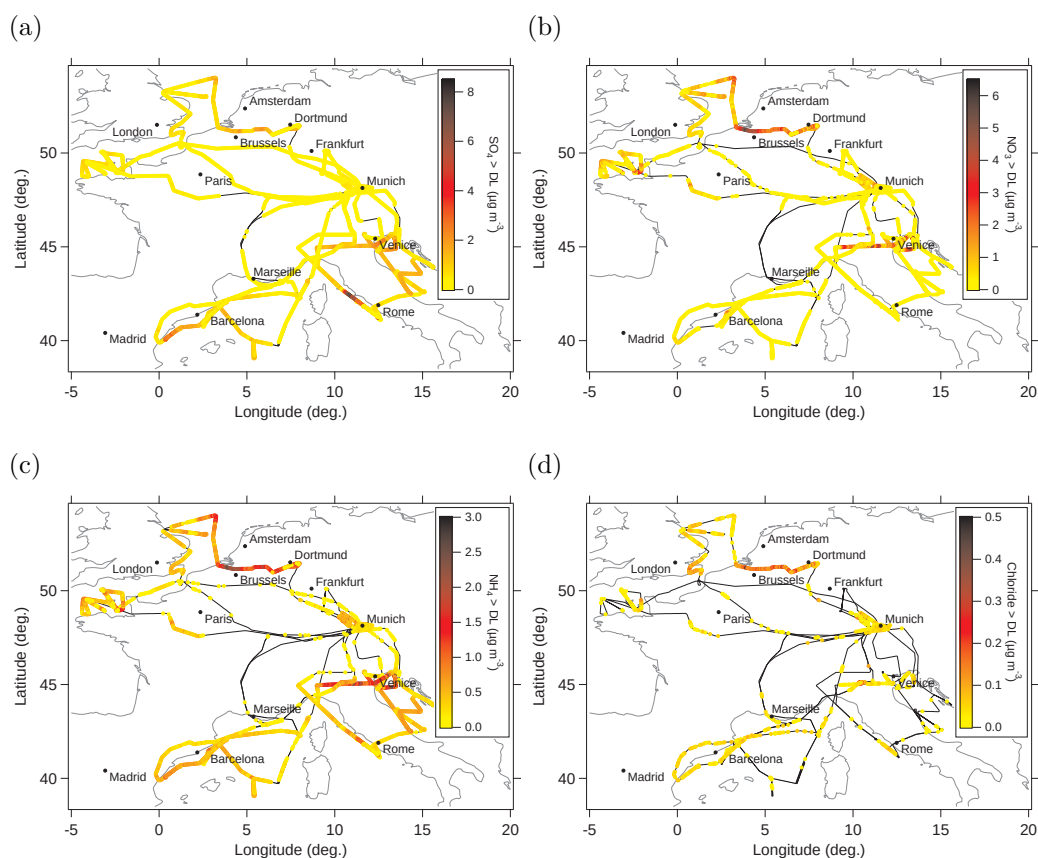


Figure 61: Flight paths of the EMeRGe-Europe campaign in summer 2017 colour-coded with the mass concentrations of SO_4 (a), NO_3 (b), NH_4 (c), and chloride (d). The data is filtered for values below the DLs to increase clarity and filter measurements are left out, too. The colour scales go up to 8.5, 6.5, 3, and 0.5 $\mu\text{g m}^{-3}$, although the maxima are 8.6, 6.7, 3.2, and 0.5 $\mu\text{g m}^{-3}$ for SO_4 , NO_3 , NH_4 , and chloride, respectively.

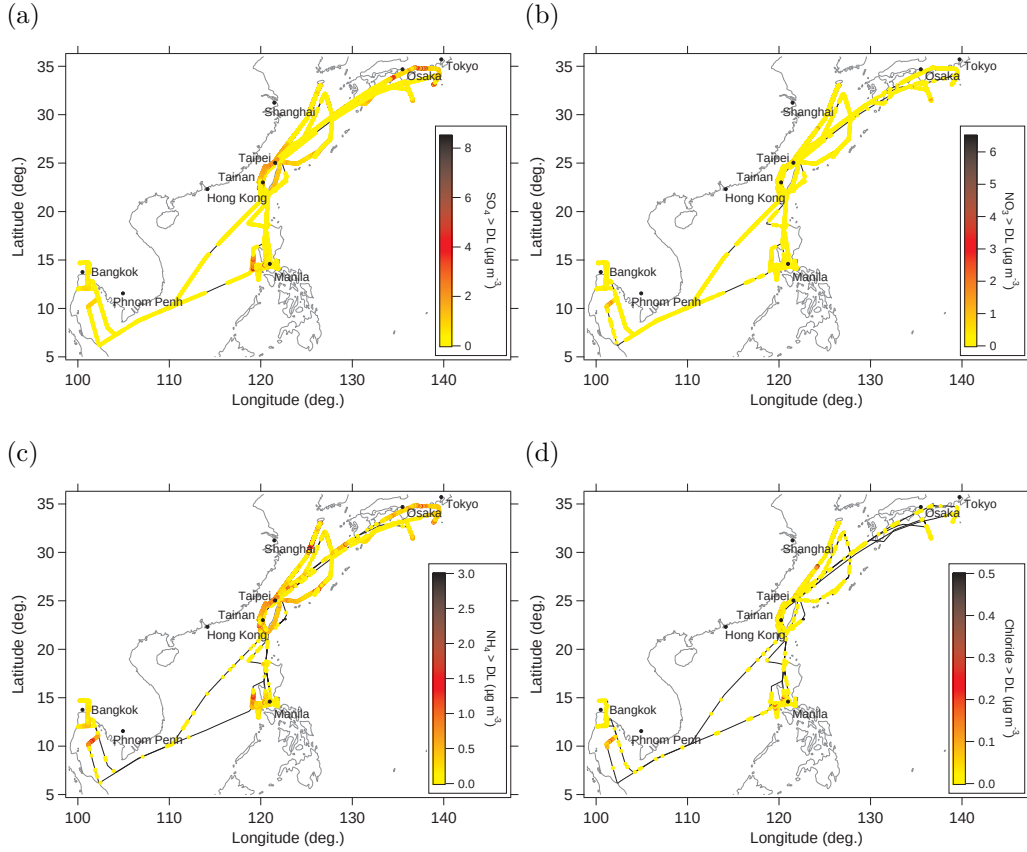


Figure 62: Flight paths of the EMERGE-Asia campaign flights E-AS-03 to E-AS-14 in spring 2018 colour-coded with the mass concentrations of SO_4 (a), NO_3 (b), NH_4 (c), and chloride (d). The data is filtered for values below the DLs to increase clarity and filter measurements are left out, too. The colour scales go up to 8.5, 6.5, 3, and $0.5 \mu\text{g m}^{-3}$ (to make it comparable to the European values), although the maxima are 9.4, 8.7, 4.8, and $0.6 \mu\text{g m}^{-3}$ for SO_4 , NO_3 , NH_4 , and chloride, respectively.

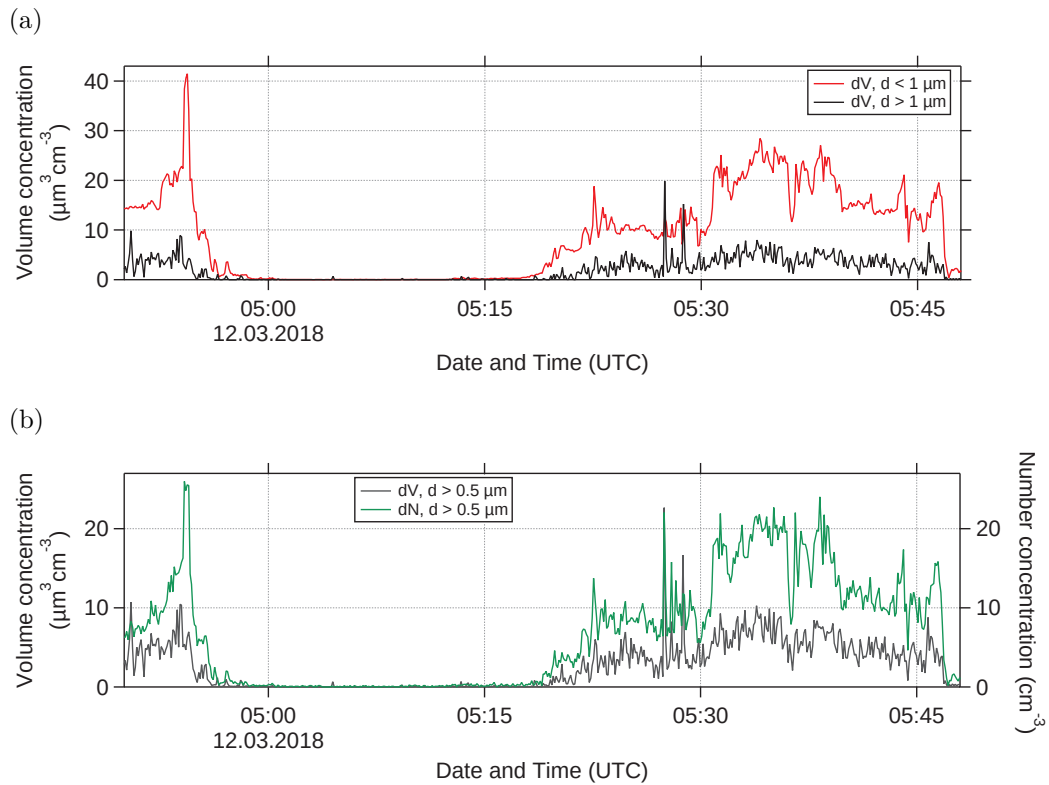


Figure 63: Excerpt of the OPC time series of flight E-AS-03: (a) volume concentrations divided at a particle diameter of $1 \mu\text{m}$. (b) Volume and number concentrations above a particle diameter of $0.5 \mu\text{m}$ used as dust proxies.

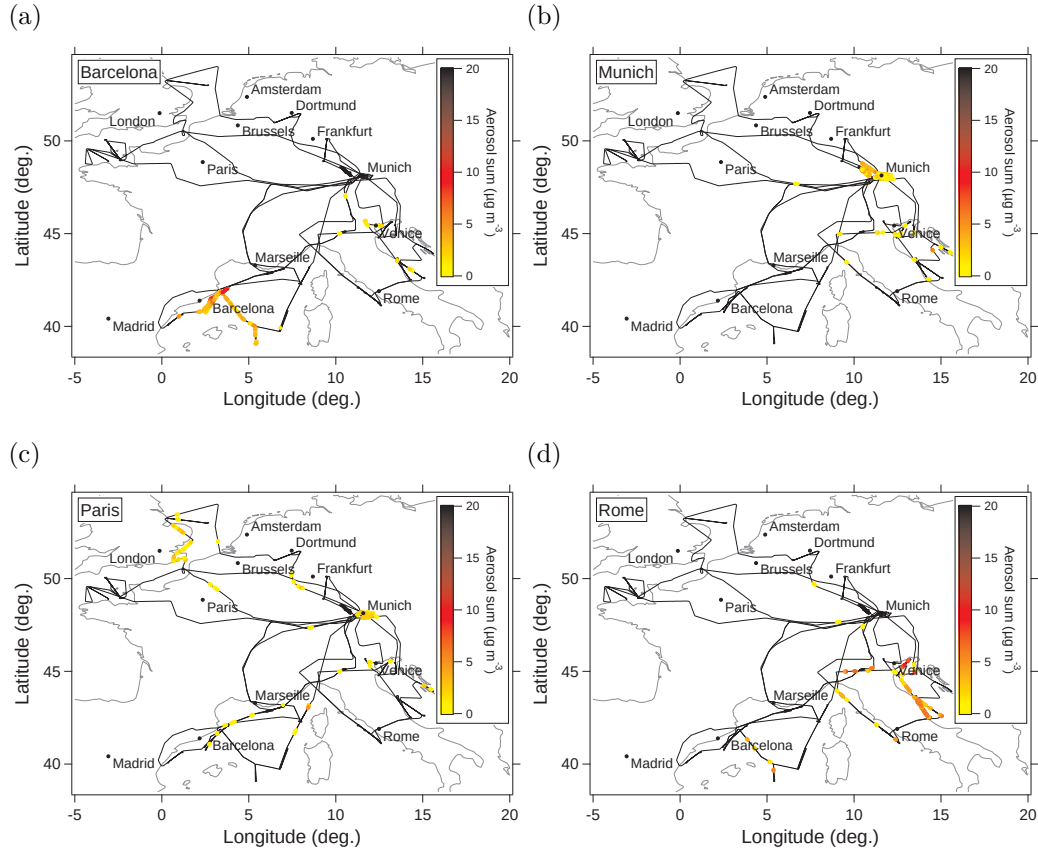


Figure 64: Flight paths of the EMerGe-Europe campaign colour-coded with the total aerosol mass concentration including rBC and filtered for emission contributions from different MPCs: Barcelona (a), Munich (b), Paris (c), and Rome (d).

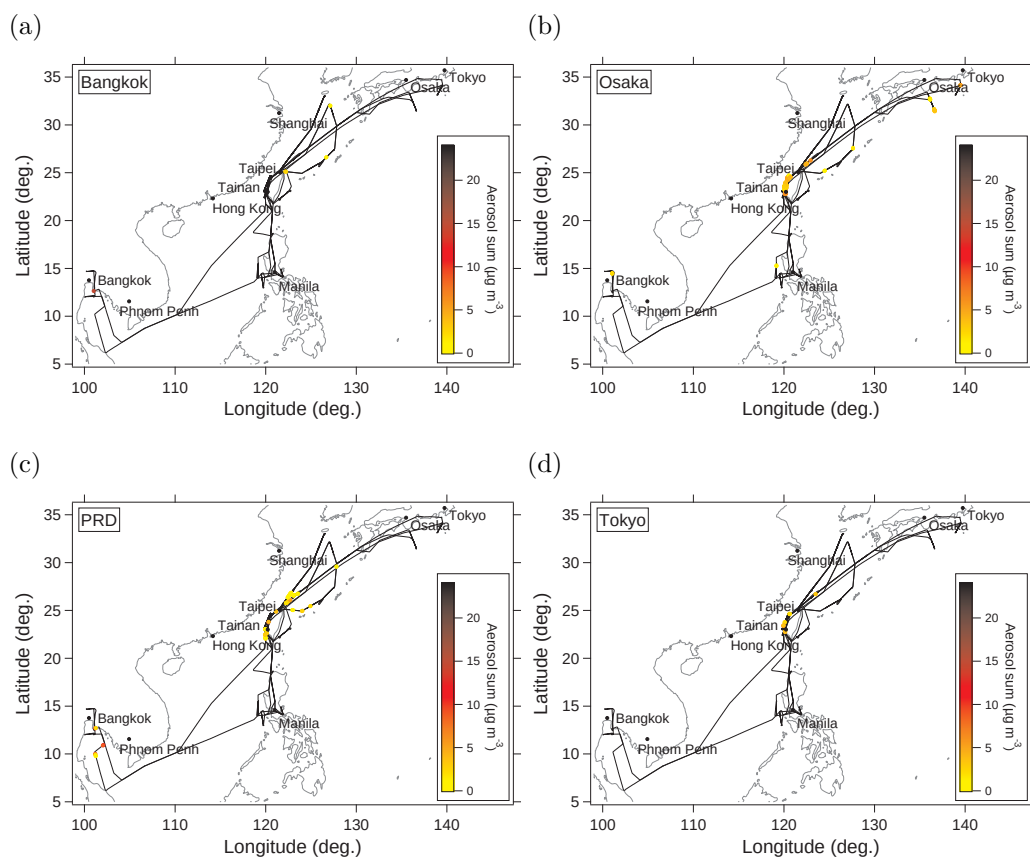


Figure 65: Flight paths of the EMerGe-Asia campaign colour-coded with the total aerosol mass concentration including rBC and filtered for emission contributions from different MPCs: Bangkok (a), Osaka (b), PRD (c), and Tokyo (d).

Table 24: Relative shares (in %) of the average aerosol composition from individual MPCs for EMeRGe-Europe (a) and -Asia (b). Abbreviations: London (LON), Paris (PAR), Madrid (MAD), Barcelona (BAR), Po Valley (POV), Rome (ROM), Munich (MUN), Belgium-Netherlands-Ruhr area (BNR), Beijing (BEI), Yangtze River Delta (YRD), Pearl River Delta (PRD), Tokyo (TOK), Osaka (OSA), Bangkok (BAN), Manila (MAN), and Taipei (TAI).

(a) EMeRGe-Europe

MPC	OA	NO ₃	SO ₄	NH ₄	Chloride	rBC
LON	45.73	20.98	15.47	13.18	0.92	3.71
PAR	53.74	11.56	18.86	10.47	1.36	4.02
MAD	71.69	4.40	11.81	6.96	0.33	4.81
BAR	52.26	6.89	25.44	12.48	0.24	2.70
POV	47.08	15.35	19.58	13.64	0.45	3.92
ROM	55.17	4.47	23.86	11.24	0.08	5.19
MUN	45.77	19.39	17.91	13.57	0.66	2.70
BNR	38.97	22.04	19.65	15.30	1.18	2.86

(b) EMeRGe-Asia

MPC	OA	NO ₃	SO ₄	NH ₄	Chloride	rBC
BEI	18.12	21.13	33.18	19.54	0.74	7.29
YRD	26.90	18.03	29.91	16.90	0.74	7.53
PRD	50.57	7.71	20.70	9.73	0.52	10.78
TOK	19.27	2.61	55.26	14.94	0.35	7.57
OSA	32.02	4.76	39.94	13.25	0.61	9.43
BAN	44.55	3.86	29.15	13.25	0.19	8.99
MAN	48.46	3.12	24.56	9.83	0.17	13.87
TAI	29.70	10.46	33.92	15.16	0.80	9.97

C.2 ACIDITY OF AEROSOL PARTICLES

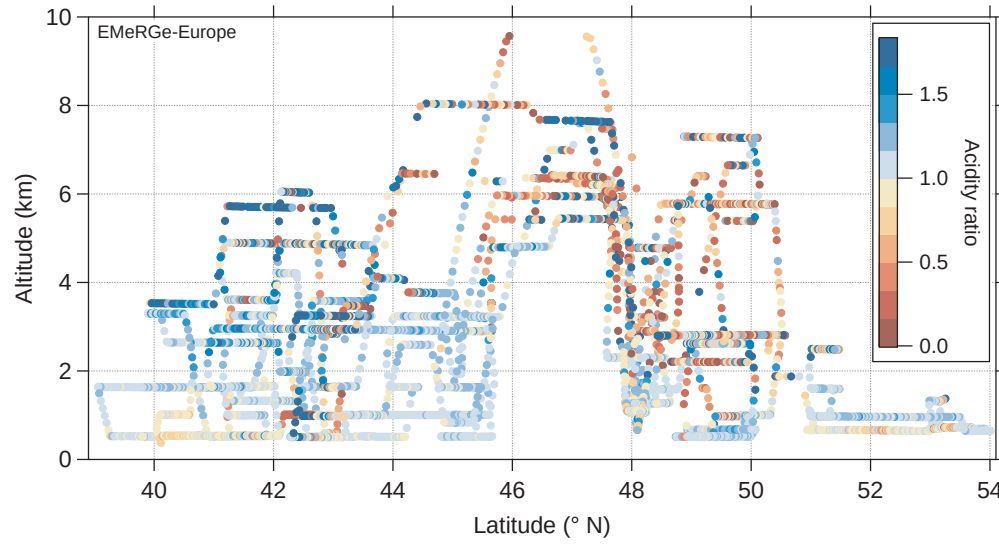


Figure 66: Vertical cross section of all EMeRGe-Europe flights colour-coded with the acidity ratio $\frac{\text{NH}_{4,\text{meas}}}{\text{NH}_{4,\text{neut}}}$. Gaps along the flight track indicate filter measurements. Acidity ratios below zero were also left out.

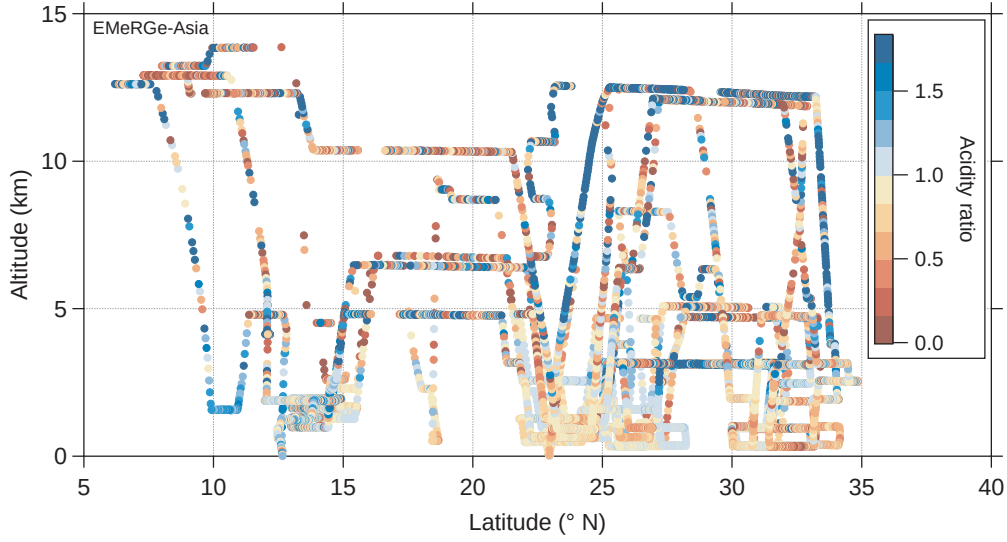


Figure 67: Same as Fig. 66 but for EMeRGe-Asia flights E-AS-03 to E-AS-14.

Table 25: Linear fit values of the acidity calculations ($\text{NH}_{4,\text{meas}}$ versus $\text{NH}_{4,\text{neut}}$) for every flight during EMeRGe-Europe and -Asia.

FLIGHT NO.	EMERGE-EUROPE	EMERGE-ASIA
01	n/a	$0.78 \cdot x - 0.02$
03	$1.14 \cdot x - 0.01$	$1.13 \cdot x - 0.02$
04	$1.13 \cdot x - 0.03$	$0.76 \cdot x + 0.07$
05	$1.06 \cdot x - 0.003$	$1.04 \cdot x - 0.002$
06	$0.84 \cdot x + 0.10$	$0.86 \cdot x - 0.02$
07	$1.10 \cdot x - 0.003$	$1.06 \cdot x - 0.07$
08	$1.13 \cdot x - 0.05$	$1.03 \cdot x + 0.03$
09	$0.94 \cdot x + 0.03$	$1.01 \cdot x - 0.04$
10	n/a	$0.85 \cdot x - 0.003$
11	n/a	$0.87 \cdot x - 0.06$
12	n/a	$0.81 \cdot x - 0.06$
13	n/a	$0.65 \cdot x + 0.06$
14	n/a	$0.93 \cdot x - 0.03$
16	n/a	$1.05 \cdot x + 0.02$

C.3 OXIDATION STATE OF OA

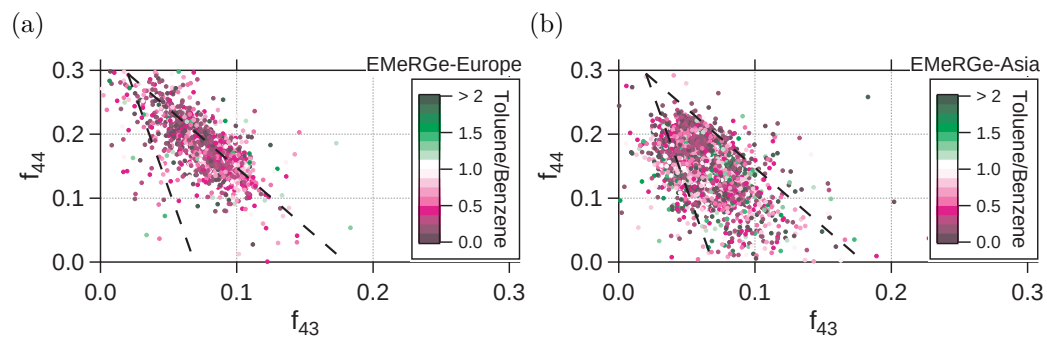


Figure 68: Triangle plots of f_{44} versus f_{43} for EMeRGe-Europe (a) and -Asia (b; flights E-AS-03 to E-AS-14). The data are colour-coded with the toluene to benzene ratio, a marker for photochemical ageing of VOCs as introduced in Sect. 3.6.2.

Table 26: Averaged heights and time ranges for the selected regions during flight E-AS-04.

REGION	HEIGHT (m)	TIME (UTC)
North	980.83	8:58:30 - 9:05:30
	651.98	9:07:30 - 9:17:30
Centre	947.21	8:56:30 - 8:58:30
	1048.07	9:05:30 - 9:07:30
	645.4	9:17:30 - 9:19:30
South	942.06	8:51:30 - 8:56:30
	651.68	9:19:30 - 9:29:30
South-east	956.3	8:39:30 - 8:51:30
	651.58	9:29:30 - 9:41:30
High shuttle	2534.47	8:07:30 - 8:36:30
East	483.54	1:53:30 - 2:24:30
East 2	3134.08	2:31:30 - 2:57:30
Background*	3157.53	4:48:30 - 5:26:30

* For the "background" segment the whole period is given here. Although, based on the VOC tagging, only background (means non-anthropogenic and non-biogenic) values within this period were taken into account for averaging which excludes about 66 % of the points (including data gaps due to the instrument specific measurement cycle and instrumental background measurements).

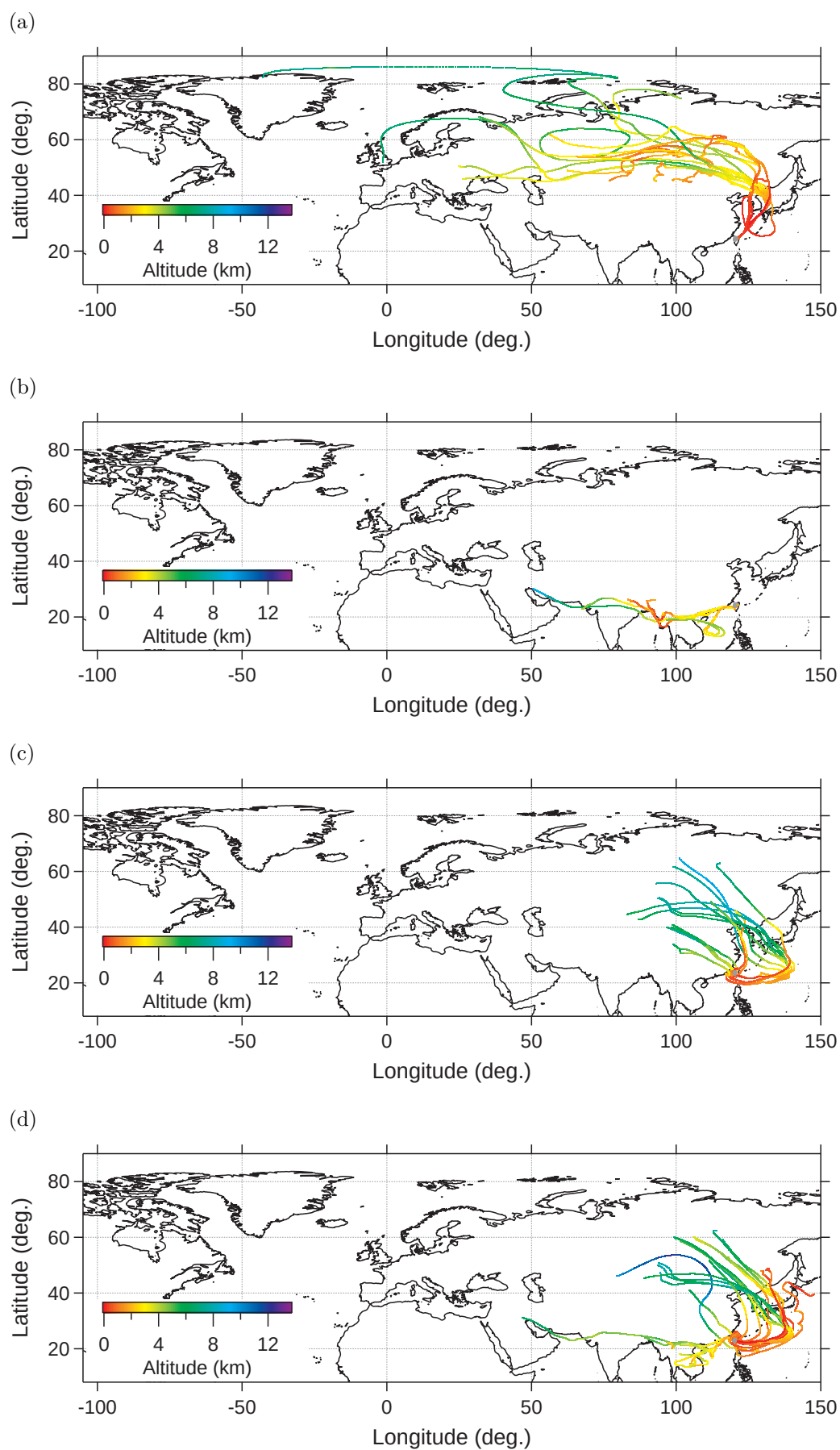


Figure 69: 240 h backward FLEXTRA trajectories for four different flight sections during flight E-AS-04: north (a), centre (b), south (c), and south-east (d). The trajectories are colour-coded with altitude.

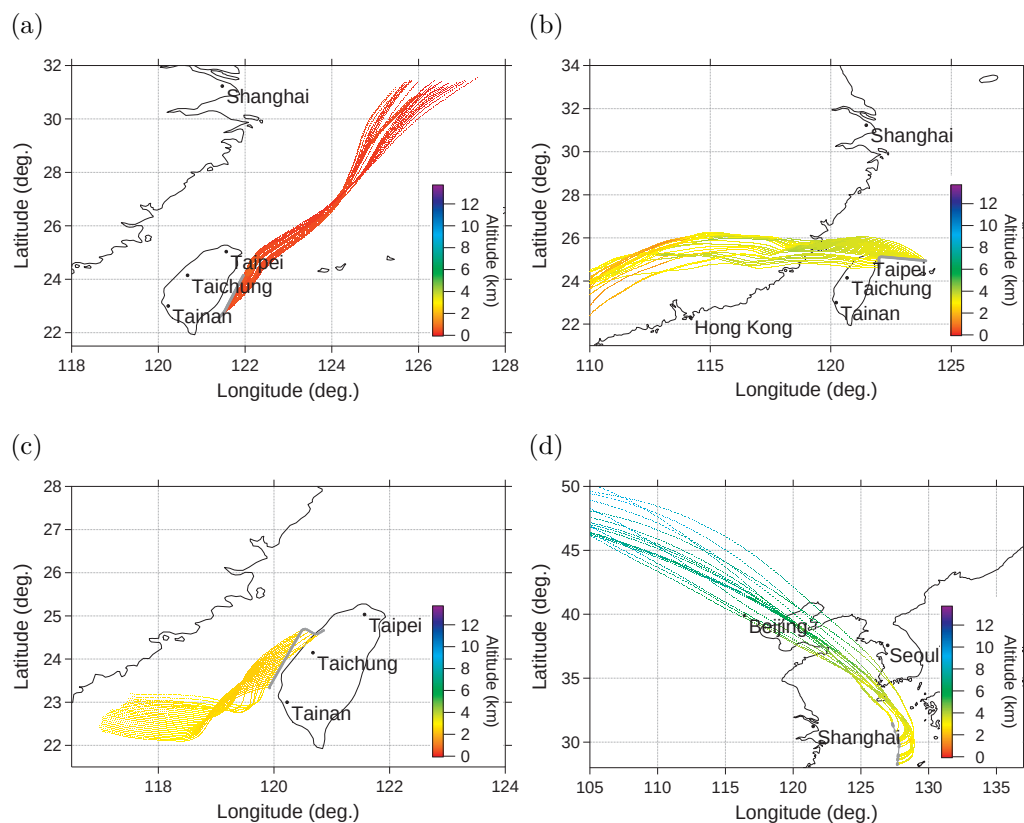


Figure 70: Four comparison flight segments in grey from flight E-AS-04: east (a), east 2 (b), high shuttle (c), and background (d). The backward trajectories are colour-coded with altitude having a length of 24 h for "east" and "high shuttle", 60 h for "east 2", and 36 h for "background". Further information on altitude and time period for each flight segment can be found in Tab. 26.

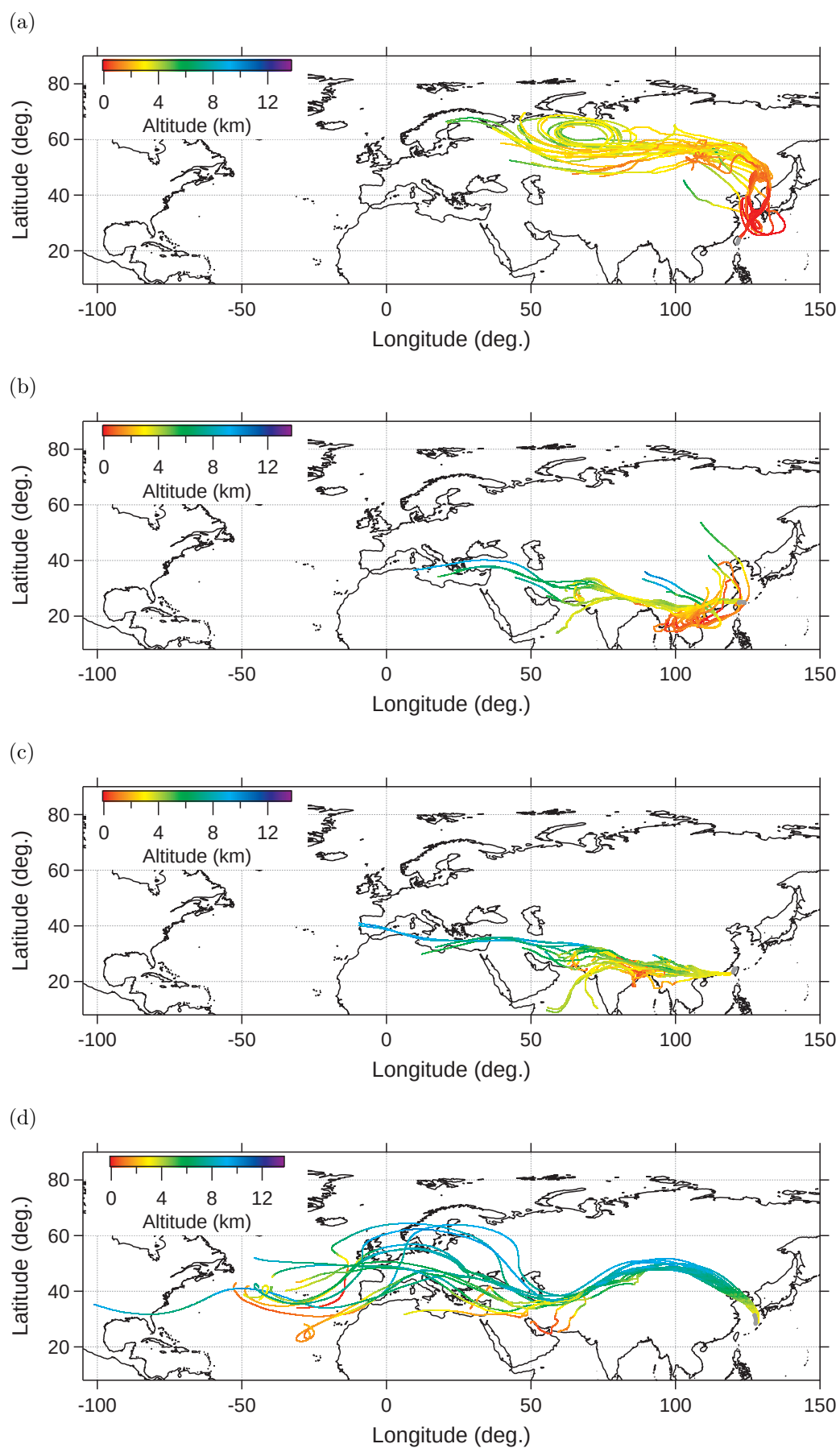


Figure 71: 240 h backward FLEXTRA trajectories for four different flight sections during flight E-AS-04: east (a), east 2 (b), high shuttle (c), and background (d). The trajectories are colour-coded with altitude.

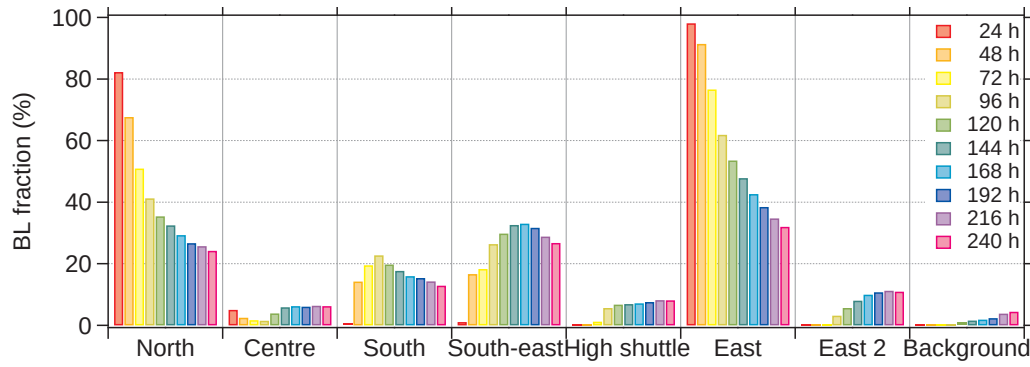


Figure 72: BL fractions for the eight defined flight segments. Their calculation is based on the FLEXTRA backward trajectories and the BL height within the model is defined as BL thickness plus surface altitude. It is checked whether each trajectory altitude is smaller than the BL height and the fraction to the total number of points is formed. The time within the BL is divided between 24 to 240 h.

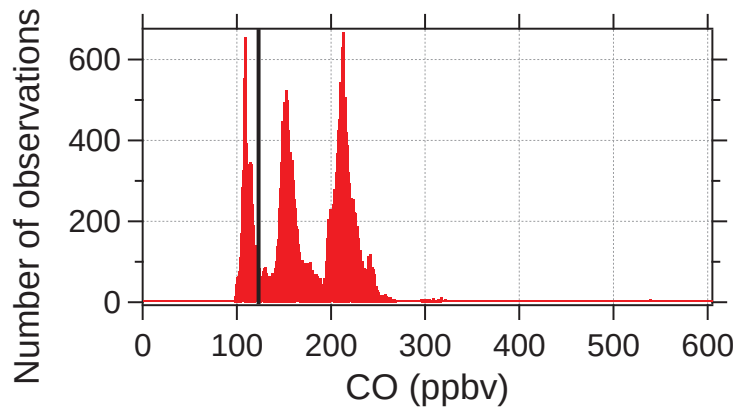


Figure 73: Histogram of the CO VMRs of flight E-AS-04 highlighting the background value set to 123 ppbv after the first maximum.

Table 27: Mean CO VMR enhancements in ppbv (simulated as explained in ect. 3.5.4) for the four target regions, their relative contribution to one segment in %, and the corresponding mean age in h. VMR values below 0.1 ppbv are excluded for all target regions. Therefore, the influence from Seoul and Osaka is neglected completely and the numbers might not add up to exactly 100 % per line. Geographical extents of the regions are given in Tab. 8.

SEGMENT	VARIABLE	BEIJING	PRD	TAIPEI	YRD
North	CO VMR	0.41	2.53	4.81	8.5
	Rel. contribution	2.5	15.58	29.7	52.04
	Mean age	88.99	103.17	8.72	79.08
Centre	CO VMR	0.42	2.18	4.82	7.62
	Rel. contribution	2.58	14.37	32.78	50.0
	Mean age	84.18	107.19	10.53	78.59
South	CO VMR	0.43	1.97	4.79	10.12
	Rel. contribution	2.51	11.24	27.65	58.14
	Mean age	87.15	112.29	14.23	78.48
South-east	CO VMR	0.33	2.34	4.09	9.48
	Rel. contribution	1.97	14.27	25.63	57.92
	Mean age	89.84	112.87	18.61	76.39
High shuttle	CO VMR	-	1.08	-	-
	Rel. contribution	-	86.27	-	-
	Mean age	-	58.6	-	-
East	CO VMR	0.98	3.5	0.19	15.13
	Rel. contribution	4.93	17.74	0.95	76.33
	Mean age	85.7	100.48	90.36	70.02
East 2	CO VMR	-	1.3	-	-
	Rel. contribution	-	98.26	-	-
	Mean age	-	64.12	-	-
Background		-	-	-	-

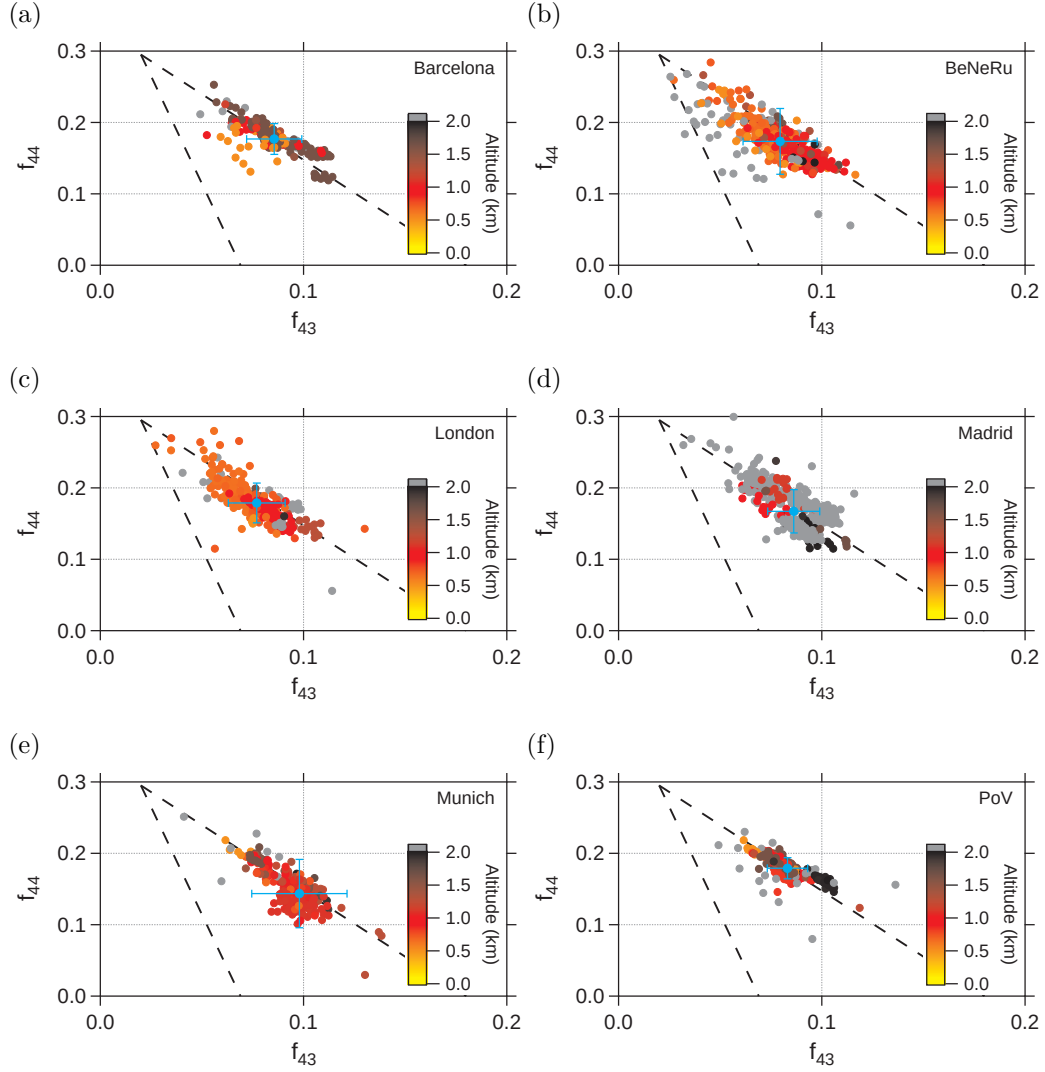


Figure 74: Arrangement of the data points in the ageing plot after Ng et al. (2010, 2011) from four selected MPCs during EMeRGe-Europe: Barcelona (a), BeNeRu (b), London (c), Madrid (d), Munich (e), and PoV (f). The data are colour-coded with altitude whereby the altitudes greater 2 km are greyed out to highlight values close to the ground. The blue cross indicates the mean value of all data points with the corresponding standard deviations as error bars.

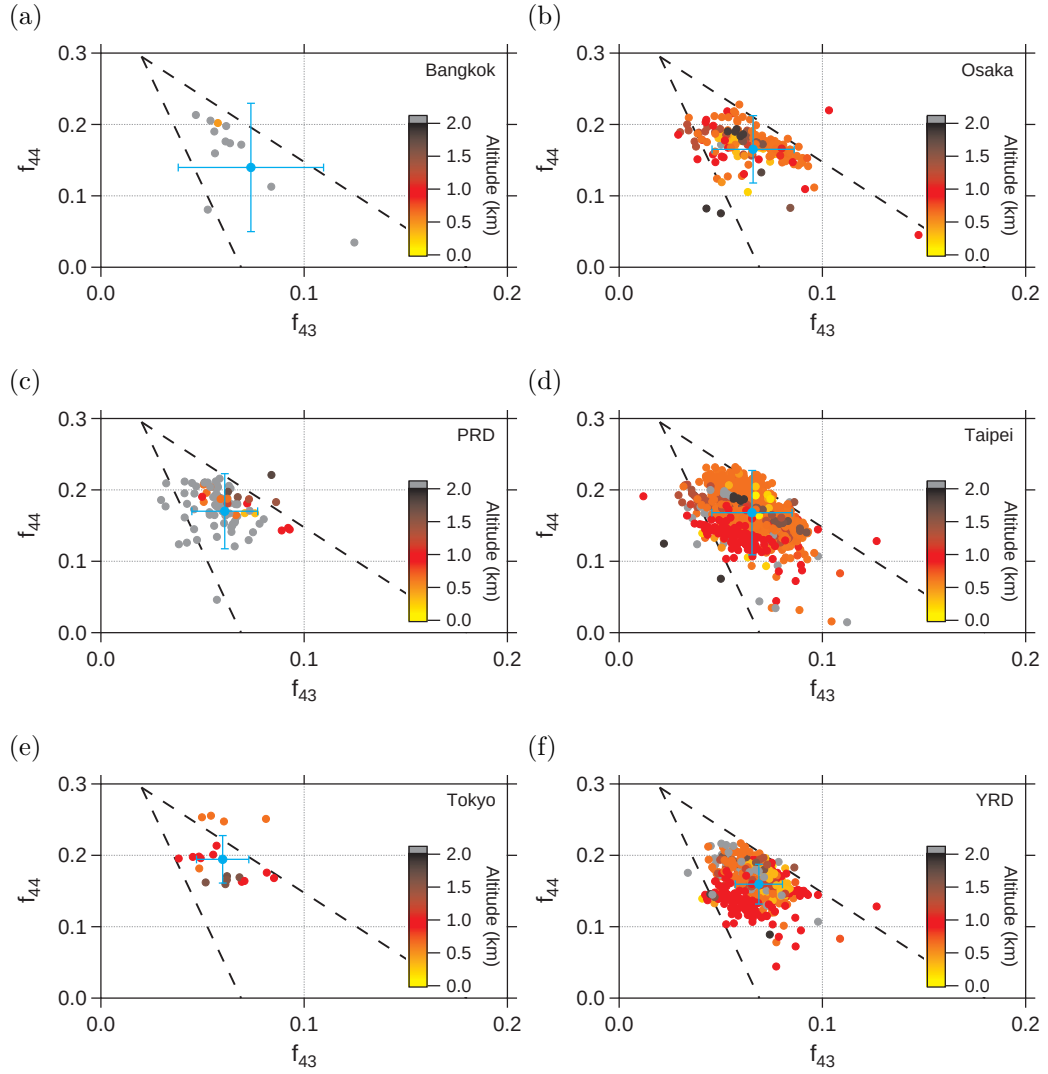


Figure 75: Arrangement of the data points in the ageing plot after Ng et al. (2010, 2011) from four selected MPCs during EMeRGe-Asia: Bangkok (a), Osaka (b), PRD (c), Taipei (d), Tokyo (e), and YRD (f). The data are colour-coded with altitude whereby the altitudes greater 2 km are greyed out to highlight values close to the ground. The blue cross indicates the mean value of all data points with the corresponding standard deviations as error bars.

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ACRONYMS

GENERAL ACRONYMS

AENEAS	AtmosphERIC Nitrogen oxides mEAsuring System
AMETYST	Aerosol MEasurement sYSTem
AMTEX	AtMospheric Tracer EXperiment
AQG	air quality guidelines
BAHAMAS	BASic HALO Measurement And sensor System
BB	biomass burning
BC	black carbon
BCS	Baiyin copper smelter
BeNeRu	Belgium-Netherlands-Ruhr area
BFSP	brute force single particle
BL	(planetary) boundary layer
CAMS	Copernicus Atmosphere Monitoring Service
CCN	cloud condensation nuclei
CCNC	CCN counter
CPC	condensation particle counter
CPI	constant pressure inlet
C-ToF-AMS	compact time-of-flight aerosol mass spectrometer
DAQ	data aquisition
DL	detection limit
DLR	Deutsches Zentrum für Luft- und Raumfahrt
DLR-FX	DLR Flight Experiments
DLR-IPA	DLR Institute of Atmospheric Physics
DMS	dimethyl sulfide
DMA	differential mobility analyser
ECMWF	European Centre for Medium-Range Weather Forecast
EDGAR	Emissions Database for Global Atmospheric Research
EMeRGe	Effect of Megacities on the Transport and Transformation of Pollutants on the Regional to Global Scales

FAAM	Facility for Airborne Atmospheric Measurements
FL	flight level
FLEXTRA	flexible trajectory model
FZ Jülich	Forschungszentrum Jülich
GPS	Global Positioning System
HALO	High Altitude and LOng range research aircraft
HASI	HALO Aerosol Submicrometer Inlet
HKMS	HALO Karlsruhe Mass Spectrometer
HOA	hydrocarbon-like organic aerosol
HTAP	Hemispheric Transport of Air Pollution
HYSPLIT	Hybrid Single-Particle Lagrangian Integrated Trajectory model
IAGOS-CARIBIC	In-service Aircraft for a Global Observing System - Civil Aircraft for the Regular Investigation of the Atmosphere Based on an Instrument Container
IEPOX	isoprene-derived epoxydiols
IUP	Institute of Environmental Physics, University of Bremen
KCS	Khatoonabad copper smelter
KIT	Karlsruhe Institute of Technology
LO-OOA	less-oxidised OOA
LV-OOA	low-volatile OOA
MCP	micro-channel plate
MO-OOA	more-oxidised OOA
MPC	major population centre
MPIC	Max Planck Institute for Chemistry
MSA	methanesulfonic acid
NCEP	National Centers for Environmental Prediction
OA	organic aerosol
OMI	Ozone Monitoring Instrument
OPC	optical particle counter
OOA	oxygenated organic aerosol
PAN	peroxyacetyl nitrate
PFC	perfluorocarbon

PM	particulate matter
PM_1	particulate matter with a diameter of 1 μm or less
$PM_{2.5}$	particulate matter with a diameter of 2.5 μm or less
PM_{10}	particulate matter with a diameter of 10 μm or less
POA	primary organic aerosol
PoV	Po Valley
PRD	Pearl River Delta
PSL	polystyrene latex
rBC	refractory black carbon
RH	relative humidity
SCC	Sarcheshmeh copper complex
SHARC	Sophisticated Hygrometer for Atmospheric Research
SMPS	Scanning Mobility Particle Sizer
SOA	secondary organic aerosol
SP2	Single Particle Soot Photometer
STP	standard temperature and pressure
SV-OOA	semi-volatile OOA
TCCON	Total Carbon Column Observing Network
TRA	temporary restricted area
TSI	Thermo-Systems Engineering Co.
UTC	coordinated universal time
VEI	volcanic explosivity index
VMR	volume mixing ratio
VOC	volatile organic compound
WHO	World Health Organization
YRD	Yangtze River Delta

PHYSICAL SYMBOLS

AB	Hz	air beam signal
CE		collection efficiency
d_{mob}	m	particle mobility diameter
d_{va}	m	particle vacuum aerodynamic diameter
d_{50}	m	cut-off diameter at 50 % transmission efficiency

F_s		conversion ratio of sulfur
f_{43}		ratio of the signal at m/z 43 (main ion: $\text{C}_2\text{H}_3\text{O}^+$) to the total organic signal
f_{44}		ratio of the signal at m/z 44 (main ion: CO_2^+) to the total organic signal
f_{60}		ratio of the signal at m/z 60 (main ion: $\text{C}_2\text{H}_4\text{O}_2^+$) to the total organic signal
IE		ionisation efficiency
m/z	m	mass to charge ratio
p	hPa	pressure
RIE		relative ionisation efficiency
SI		single ion
T	K	temperature
T_e		e-folding time
θ_v	K	virtual potential temperature

CHEMICAL SYMBOLS

CH_4	methane
CO	carbon monoxide
CO_2	carbon dioxide
H_2O	water
NH_4	ammonium aerosol
NH_4NO_3	ammonium nitrate
$(\text{NH}_4)_2\text{SO}_4$	ammonium sulfate
NO	nitric oxide
NO_x	nitrogen oxides
NO_y	total reactive nitrogen
NO_2	nitrogen dioxide
NO_3	nitrate aerosol
OH	hydroxide
O_3	ozone
SO_2	sulfur dioxide
SO_4	sulfate aerosol
SO_x	sulfur oxides

LIST OF PUBLICATIONS

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International Union of Geodesy and Geophysics General Assembly, Montreal, Canada, 8-18 July, 2019.

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In Chapter 2, maps were created from public domain GIS data found on the Natural Earth website (<http://www.naturalearthdata.com>). NCEP-NCAR Reanalysis 1 data were provided by the NOAA Physical Sciences Laboratory, Boulder Colorado from their website (<https://psl.noaa.gov/>). The basic meteorological data were provided by Andreas Giez, Volker Dreiling, Martin Zöger, and Christian Mallaun. The aerosol number concentrations were provided by Daniel Sauer and Jennifer Wolf. The measurements for the particle losses in the inlet line were carried out by myself.

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Chapter 4 presents the results of the EMeRGe campaign. This intensive field campaign consumed a lot of work for many people. All participants of the EMeRGe campaign took their effort for a successful campaign. John Burrows, Maria D. Andrés Hernández, Hans Schlager, Ralf Koppmann, Andreas Zahn, Ulrich Platt, Klaus Pfeilsticker, Johannes Schneider, Mihalis Vrekoussis, Andrea Pozzer, and Helmut Ziereis designed the research project. General logistics were handled by DLR-FX, the pilots realised the specific flight patterns. The preparation of the C-ToF-AMS was done by myself with the help of Johannes Schneider, Christiane Schulz, Christian Gurk, Philipp Schuhmann, Thomas Klimach, Sergej Molleker, Frank Drewnick, Thomas

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*May the calm be widespread
May the ocean glisten as greenstone
May the shimmer of light
Ever dance across your pathway*
— Maori proverb by Rangawhenua Tawhaki

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CURRICULUM VITAE

The curriculum vitae is not included in the electronic version.

DECLARATION

I hereby declare that I wrote the submitted dissertation without any unauthorised 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. External contributions to data and analyses presented in this study are specified in detail on p. 227.

Mainz, 18 September 2023



Katharina Kaiser