

Bi-directional feedbacks of interactive vegetation dynamics and the atmospheric aerosol burden in the Earth system

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Declaration

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Acknowledgment

Abstract

The Earth system is governed by complex interactions among its various *spheres*, with the biosphere and atmosphere playing critical roles in regulating key processes. The biosphere-atmosphere interface regulates important biophysical and biogeochemical cycles through the exchange of energy and material. These cycles include the carbon and water cycles, surface energy balance, and various other essential processes. Millions of years of evolution have led plants to emit biogenic volatile organic compounds (BVOCs) as a mechanism of protection. This includes, for example, the protection of photosynthetic activity from damage caused by reactive oxygen species synthesised in leaves at high temperatures, warning through inter-plant communication, and defence against herbivory. These BVOCs, primarily isoprene (C_5H_8) and monoterpenes ($C_{10}H_{16}$), account for 90% of total global BVOC emissions, with annual global emissions of around 600 Tg.

BVOCs are highly reactive species and, given their high abundance, play a significant role in the global atmospheric gaseous and aerosol composition. Their reactivity with atmospheric oxidants, such as the hydroxyl radical (OH), influences the atmosphere's oxidation capacity and affects the lifetimes of atmospheric pollutants and greenhouse gases, such as methane (CH_4) and tropospheric ozone (O_3). These oxidation processes also produce intermediate species with lower vapour pressures, which tend to partition into the aerosol phase, forming biogenic secondary organic aerosols (bSOA).

Advances in computational power have enabled atmospheric numerical models to incorporate greater complexities, enhancing the representation of these processes and their interactions with the biosphere and other Earth system components. These models are known as Earth System Models (ESMs) and are among the most powerful tools available for advancing our understanding of the Earth system. This thesis explores the dynamic interplay between terrestrial vegetation and atmospheric aerosols through BVOC emissions, utilising advanced model representations within the framework of an ESM.

The first study presented in this work involves the coupling of BVOC emissions in the chemistry-climate model EMAC using dynamic vegetation states from a dynamic global vegetation model (DGVM) LPJ-GUESS. This coupling facilitates bidirectional interactions between the biosphere and atmosphere, enabling more accurate simulations of BVOC emissions based on dynamic vegetation states rather than static climatological data. It is shown that BVOC emissions are accurately represented with the new setup and are responsive to changes in vegetation states.

The second study investigates the variations in BVOC emissions caused by the El Niño-Southern Oscillation (ENSO). The study presents a comprehensive analysis of the changes in atmospheric conditions resulting from ENSO and sheds light on how these modifications extend to impact vegetation and BVOC emissions. It is shown that strong El Niño events increase global isoprene emission fluxes by 2.9% and that persistent ENSO conditions shift the vegetation to a new quasi-equilibrium state, leading to an amplification of BVOC emission changes with up to a 19% increase in isoprene fluxes over the Amazon.

The third study examines the impacts of deforestation on natural land vegetation cover and atmospheric aerosols. By implementing a deforestation routine in LPJ-GUESS, a deforestation scenario is simulated to evaluate the impact on BVOC emissions and secondary organic aerosols. The results reveal that compared to natural vegetation cover, present-day deforestation leads to a 26%

reduction in BVOC emissions, resulting in a decrease in the global organic aerosol (OA) burden by 0.16 Tg (a decrease of 29%). The assessment also includes changes in cloud condensation nuclei (CCN) and cloud droplet number concentration (CDNC) resulting from deforestation. In the present-day deforestation scenario, a positive total radiative effect (aerosol + cloud) of 87.9 mW m⁻² (indicating warming) is estimated compared to the natural vegetation scenario.

The fourth study delves into the discussion of BVOC-aerosol feedbacks. The formation of bSOA from BVOC emissions alters atmospheric states, subsequently influencing vegetation and BVOC emission rates. These feedback mechanisms have attracted recent attention in the literature, however, their relevance in the Earth system remains difficult to quantify due to inherent uncertainties in the models. The EMAC/LPJ-GUESS configuration employed in this thesis is used to investigate such feedback mechanisms. While these results warrant further investigation, preliminary findings suggest that the changes in BVOC emissions resulting from aerosol feedbacks simulated with the EMAC/LPJ-GUESS configuration appear to be small and likely fall within the internal variability of the model.

This work enhances our knowledge on the role of BVOC in the Earth system by providing insights into their complex interactions with atmospheric processes and the terrestrial biosphere. By enhancing the capabilities of the EMAC model, this research offers a comprehensive understanding of how BVOC emissions respond to dynamic environmental conditions, such as those influenced by El Niño events and deforestation. Furthermore, the investigation into BVOC-aerosol feedbacks contributes to our understanding of the interconnectedness between atmospheric chemistry, vegetation dynamics, and climate.

Zusammenfassung

Das System Erde wird durch komplexe Wechselwirkungen zwischen seinen verschiedenen Sphären bestimmt, wobei die Biosphäre und die Atmosphäre eine entscheidende Rolle bei der Regulierung von Schlüsselprozessen spielen. Die Schnittstelle zwischen Biosphäre und Atmosphäre reguliert die wichtigsten biophysikalischen und biogeochemischen Kreisläufe durch den Austausch von Energie und Stoffen. Zu diesen Zyklen gehören der Kohlenstoff- und der Wasserkreislauf, der Energiehaushalt der Oberfläche und verschiedene andere wichtige Prozesse. Jahrmillionen währende Evolution haben Pflanzen dazu geführt biogene flüchtige organische Verbindungen (BVOC) zu emittieren. Dies ist ein Mechanismus um die photosynthetische Aktivität vor Schäden durch reaktive Sauerstoffspezies zu schützen, die bei hohen Temperaturen in den Blättern entstehen, und um Signale zu senden und sich gegen Fressfeinden zu verteidigen. Diese flüchtigen organischen Verbindungen, vor allem Isopren (C_5H_8) und Monoterpene ($C_{10}H_{16}$), machen 90% der gesamten globalen BVOC-Quellen aus, mit jährlichen globalen Emissionen von etwa 600 Tg. BVOC sind hochreaktive Spezies und spielen aufgrund ihres hohen Vorkommens eine wichtige Rolle bei der globalen Zusammensetzung der Gase und Aerosole in der Atmosphäre. Ihre Reaktivität mit atmosphärischen Oxidationsmitteln, wie beispielsweise dem Hydroxylradikal (OH), beeinflusst die Oxidationskapazität der Atmosphäre und wirkt sich auf die Lebensdauer von Luftschadstoffen und Treibhausgasen, wie zum Beispiel Methan (CH_4) und troposphärischem Ozon (O_3) aus.

Bei diesen Oxidationsprozessen entstehen auch Intermediate mit niedrigeren Dampfdrücken, die dazu neigen in die Aerosolphase überzugehen, und biogene sekundäre organische Aerosole (bSOA) zu bilden. Fortschritte bei der Rechenleistung von Computern Modellen haben es ermöglicht eine größere Komplexität in numerischen Atmosphärenmodellen zu berücksichtigen und damit die Darstellung dieser Prozesse und ihrer Wechselwirkungen mit der Biosphäre und anderen Komponenten des Erdsystems zu verbessern. Diese Modelle werden als Erdsystemmodelle (ESM) bezeichnet und gehören zu den leistungsfähigsten Instrumenten, die für ein besseres Verständnis des Erdsystems zur Verfügung stehen. In dieser Arbeit wird das dynamische Zusammenspiel zwischen terrestrischer Vegetation und atmosphärischen Aerosolen durch BVOC-Emissionen unter Verwendung fortschrittlicher Modelldarstellungen im Rahmen eines ESM untersucht.

Die erste in dieser Arbeit vorgestellten Studie umfasst die Kopplung von BVOC-Emissionen im Chemie-Klima-Modell EMAC mit dynamischen Vegetationszuständen aus einem dynamischen globalen Vegetationsmodell (DGVM) LPJ-GUESS. Diese Kopplung erleichtert die bidirektionalen Wechselwirkungen zwischen Biosphäre und Atmosphäre und ermöglicht genauere Simulationen von BVOC-Emissionen auf der Grundlage dynamischer Vegetationszustände anstelle statischer klimatologischer Daten. Es wird gezeigt, dass die BVOC-Emissionen mit dem neuen System genau dargestellt werden und auf Veränderungen des Vegetationszustandes reagieren.

In der zweiten Studie werden die durch die El Niño-Southern Oscillation (ENSO) verursachten Schwankungen der BVOC-Emissionen untersucht. Die Studie enthält eine umfassende Analyse der durch ENSO verursachten Veränderungen der atmosphärischen Bedingungen und beleuchtet, wie sich diese Veränderungen auf die Vegetation und die BVOC-Emissionen auswirken. Es wird gezeigt, dass starke El-Niño-Ereignisse die globalen Isopren-Emissionsflüsse um 2,9% erhöhen und dass anhaltende ENSO-Bedingungen die Vegetation in einen neuen Quasi-Gleichgewichtszustand versetzen. Dies führt zu einer Verstärkung der BVOC-Emissionsänderungen mit einem Anstieg der

Isopren-Flüsse über dem Amazonas um bis zu 19%.

In der dritten Studie werden die Einflüsse von Entwaldung auf die natürliche Vegetationsdecke und die atmosphärischen Aerosole untersucht. Durch die Implementierung einer Entwaldungsroutine in LPJ-GUESS wird ein Entwaldungsszenario simuliert, um die Auswirkungen auf BVOC-Emissionen und sekundäre organische Aerosole zu untersuchen. Die Ergebnisse zeigen, dass die heutige Entwaldung im Vergleich zur natürlichen Vegetationsbedeckung zu einer Verringerung der BVOC-Emissionen um 26% führt. Dies hat eine Verringerung der globalen Belastung durch organisches Aerosol (OA) um 0,16 Tg (ein Rückgang um 29 Prozent) zur Folge. Die Analyse umfasst auch Änderungen der Wolkenkondensationskerne (CCN) und der Wolkentröpfchenzahlkonzentration (CDNC) infolge der Entwaldung. Für das heutige Entwaldungsszenario wird ein positiver Gesamtstrahlungseffekt (Aerosol + Wolken) von $87,9 \text{ mW m}^{-2}$ (was eine Erwärmung anzeigt) im Vergleich zum Szenario der natürlichen Vegetation identifiziert.

Die vierte Studie befasst sich mit der Analyse von BVOC-Aerosol-Rückkopplungen. Die Bildung von bSOA aus BVOC-Emissionen verändert die atmosphärischen Zustände, was wiederum die Vegetation und die BVOC-Emissionsraten beeinflusst. Diese Rückkopplungsmechanismen haben in jüngster Zeit in der Literatur Aufmerksamkeit erregt, ihre Bedeutung für das Erdsystem ist jedoch aufgrund der inhärenten Unsicherheiten in den Modellen nach wie vor schwer zu quantifizieren. Die in dieser Arbeit verwendete EMAC/LPJ-GUESS-Konfiguration wird zur Untersuchung solcher Rückkopplungsmechanismen verwendet. Obwohl diese Ergebnisse weitere Untersuchungen erfordern, deuten die vorläufigen Ergebnisse darauf hin, dass die Änderungen der BVOC-Emissionen, die sich aus den mit der EMAC/LPJ-GUESS-Konfiguration simulierten Aerosol-Rückkopplungen ergeben, gering zu sein scheinen und wahrscheinlich innerhalb der internen Variabilität des Modells liegen.

Diese Arbeit erweitert unser Wissen über die Rolle von BVOC im Erdsystem, indem sie Einblicke in ihre komplexen Wechselwirkungen mit atmosphärischen Prozessen und der terrestrischen Biosphäre gewährt. Durch die Erweiterung der Fähigkeiten des EMAC-Modells bietet diese Forschung ein umfassendes Verständnis dafür, wie BVOC-Emissionen auf dynamische Umweltbedingungen, wie zum Beispiel El-Niño-Ereignisse oder Entwaldung, reagieren. Darüber hinaus trägt die Untersuchung der Rückkopplungen zwischen BVOC und Aerosolen zu unserem Verständnis der Zusammenhänge zwischen Atmosphärenchemie, Vegetationsdynamik und Klima bei.

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1 Motivation

The Earth system includes a multitude of processes that are all interlinked, forming a complex web of interactions across the different *spheres*. Within this complex network, the biosphere and atmosphere emerge as key players, exerting significant influence on each other and shaping the dynamics of the entire system. The land biosphere serves as a primary driver of different ecosystems, regulating carbon and water cycles, and modulating surface energy balance [Bonan, 2008]. At the same time, atmospheric aerosols, ranging from natural sources such as sea spray and volcanic emissions to anthropogenic pollutants like particulate matter, influence radiative transfer, cloud formation, and air quality, thus exerting far-reaching impacts on the climate and human health.

The relationship between terrestrial vegetation and atmospheric aerosols manifests through various pathways. These include primary aerosol emissions from forest fires and the release of fungal spores from vegetation. However, the influence of terrestrial vegetation on atmospheric aerosols extends beyond direct emissions. Plants are known to emit vast amounts of biogenic volatile organic compounds (BVOCs), accounting for 90% of the total volatile organic carbon (VOC) emissions to the atmosphere [Guenther et al., 1995]. The processes driving VOC emissions from plants are complex and not yet fully understood, however, BVOCs seem to play a role in protecting photosynthetic activity in plants from damage caused by reactive oxygen species, which are synthesised in leaves at high temperatures [Harrison et al., 2013, Lantz et al., 2019, Niinemets, 2010]. BVOC emissions can also be triggered by other chemical, physical or biological stresses and processes; e.g., herbivory [Laothawornkitkul et al., 2008], signalling between organisms [Zuo et al., 2019], or also oxidative stress originating from the atmosphere [e.g., under elevated ozone concentrations, Sharkey et al., 2008]. Plants emit an array of VOCs, but different plant species emit different compounds according to their evolutionary adaptation. For example, the emission of isoprene can be considered an evolutionary trait that benefits certain plant species in hot, dry environments [Taylor et al., 2018]. Isoprene and monoterpenes are the most abundant species among the BVOCs [Guenther et al., 2012, Kesselmeier and Staudt, 1999, Lathi  re et al., 2006], and current studies estimate their emission between 412 and 682 Tg per year [Guenther et al., 2006, Sindelarova et al., 2014]. The global impact of BVOC emissions is significant on a global scale as their high reactivity alters the atmospheric composition [Atkinson, 2000].

The short lifespan of BVOCs stems from their rapid oxidation upon release from the canopy. This oxidation primarily involves the hydroxyl (OH) radical as well as other key oxidizing agents like O_3 and NO_3 radicals [Shrivastava et al., 2017]. These reactions produce various lower volatility oxidation products that tend to partition into the aerosol phase or condense onto existing particles, leading to the formation of biogenic secondary organic aerosols (bSOA) [Kavouras et al., 1998, Spracklen et al., 2011]. Despite the relatively low yield of SOA, the substantial volume of BVOC emissions constitutes a significant global source of bSOA, estimated at approximately 17–52 Tg yr^{-1} , with an associated global mean burden ranging from 0.5–0.77 Tg [Held and Spracklen, 2015]. Given their significance, considerable effort has been made to improve BVOC representations in global models. BVOC emissions are sensitive to factors such as temperature, light, water stress, CO_2 concentrations, and vegetation changes, leading to relatively high uncertainties in current global emission estimates [Arneth et al., 2007, Guenther et al., 1991, Schurgers et al., 2009]. Additionally, with the current challenges of rapid climate change and other human activities such

as land cover change, these variables are rapidly evolving, further influencing BVOC emissions. This thesis improves BVOC representations in the chemistry-climate model EMAC by enabling BVOC calculations based on interactive vegetation dynamics rather than fixed climatological vegetation states. For the first time in the EMAC model, dynamic vegetation information is used to influence atmospheric states through BVOC emissions, enabling bidirectional interactions between the biosphere and the atmosphere. With this new setup, the thesis further explores the impact on vegetation and BVOC emissions resulting from perturbations in the Earth system, namely the El Niño Southern Oscillation (ENSO) and vegetation cover change, i.e., deforestation. Moreover, this work investigates atmosphere-biosphere feedbacks via changes in global aerosol loading from BVOC oxidation, providing deeper insights into the response of vegetation states and consequently BVOC emissions.

2 Introduction

On February 14th 1990, the Voyager 1 space probe captured an image of planet Earth from a distance of around 6 billion kilometres. Earth appears as a speck suspended in a sunbeam amidst the vastness of space [NASA/JPL-Caltech, 2020]. Named by the astronomer Carl Sagan as the “Pale Blue Dot”, this image serves to underscore the delicate and invaluable nature of our home planet. Humans are an integral part of Earth, and as scientists, we get the privilege to study Earth’s complexities in detail. This knowledge serves as a tool for mitigating the impacts of human activities on the environment and promoting sustainable practices that ensure the long-term health and resilience of our planet.

2.1 Earth: A system of systems

The “Pale Blue Dot” image reminds us that Earth is part of larger systems governing the universe, such as our Solar System and the Milky Way Galaxy, but as a planet, Earth also consists of major subsystems that interact with each other. The term “Earth system”, refers to the interconnected and dynamic network of physical, chemical, and biological processes that operate on and within the planet Earth. In particular, it considers interactions and feedbacks, through the exchange of material and energy fluxes among the Earth’s subsystems, cycles, processes and *spheres*, namely the atmosphere, biosphere, hydrosphere, cryosphere, and geosphere [Jacobson, 2005]. The hydrosphere encompasses liquid water found in oceans, inland water bodies, and groundwater. The cryosphere comprises frozen water, while the geosphere refers to the solid Earth, including the core, mantle, crust, and other solid layers. This work mainly focuses on processes within the atmosphere and biosphere including their interactions. These spheres are described in more detail below.

2.1.1 The Atmosphere

The atmosphere refers to a thin layer of gaseous molecules and aerosols held by Earth’s gravitational pull. The predominant gases are nitrogen (78.08%), oxygen (20.95%), argon (0.93%), and water vapour (0-5%). Other molecules, known as trace gases, such as carbon dioxide, methane, and ozone, are present in lower concentrations and are typically measured in parts per million (ppmv) or parts per billion (ppbv) by volume of air. [Wallace and Hobbs, 2006]. The atmosphere can be subdivided into several layers generally characterised by temperature inversions, i.e., reversal of the vertical temperature gradient. The *troposphere* is the layer close to the Earth’s surface and extends up to 10 km. The troposphere accounts for ~80% of the mass of the atmosphere and is relatively well-mixed with temperature decreasing with height [Wallace and Hobbs, 2006]. The troposphere is followed by the *stratosphere* (~10-50 km), where temperature increases with altitude, inhibiting vertical mixing. The positive lapse rate in the stratosphere results from the absorption of ultraviolet radiation by ozone molecules (also known as the ozone layer). Above the stratosphere, lies the *mesosphere* (~50-80 km), where temperature decreases with height. The top layer is known as the *thermosphere* (~80-100 km). Here temperature increases again with height due to photodissociation and photoionisation of diatomic nitrogen and oxygen molecules. There is no distinct point to where the atmosphere ends, but at around 105 km, the mean free path between molecular collisions exceeds 1 m [Wallace and Hobbs, 2006].

Apart from geological processes that derive energy from Earth's core through radioactive processes, the sun stands as the primary source of energy for our planet. Therefore, the interaction between the atmosphere and Earth's surface with incoming electromagnetic radiation from the sun governs planetary temperatures, along with long-term weather patterns, i.e., the climate. The atmosphere is transparent to most of the incoming short-wave solar radiation but opaque to outgoing long-wave radiation emitted by the Earth's surface [Wallace and Hobbs, 2006]. A simple radiative balance calculation, accounting for the retained energy by Earth from solar radiation and the energy emitted back to space following the Stefan-Boltzmann Law, reveals that the Earth's average temperature should be around -21°C . However, Earth's actual average temperature is $\sim 15^{\circ}\text{C}$, due to the atmospheric absorption of outgoing radiation from *greenhouse gases* including water vapour and carbon dioxide (CO_2). CO_2 is a naturally occurring gas and it helped to modulate Earth's temperature for millions of years, keeping temperatures above freezing, thus enabling the existence of liquid water and life as we know it. Since the Industrial Revolution started in the late 1700s, anthropogenic activities involving the burning of *fossil fuels* have drastically increased CO_2 concentration in the atmosphere resulting in higher global temperatures [Solomon et al., 2007]. The warming of the planet has many knock-on effects influencing weather patterns and resulting in rapid climate change. Mitigating climate change is one of the biggest challenges humanity is facing today.

The Industrial Revolution brought further challenges with it: air pollution. In the 1900s people realised that a multitude of chemical species emitted from industrial activities were deteriorating the air quality within cities, leading to respiratory problems and premature deaths [Stone, 2002]. It came to the realisation that some of these pollutants could travel long distances and undergo significant transformations with serious environmental impacts. Another breakthrough emphasising the relevance of atmospheric chemistry was the discovery of the ozone hole. Stratospheric ozone plays a vital role in shielding life on Earth's surface from harmful UV radiation [Fahey et al., 2018]. Paul Crutzen, a Nobel laureate and former director at the Max Planck Institute for Chemistry in Mainz, significantly advanced our understanding of the ozone layer [Crutzen, 1970]. Crutzen also engaged in political efforts to curb the use of chlorofluorocarbons (CFCs), which were accelerating the depletion of stratospheric ozone. Following the Montreal Protocol in 1987, world leaders agreed to phase out ozone-depleting substances and the ozone hole has been steadily recovering since then [Solomon et al., 2016].

Another critical component of the atmosphere closely linked with pollution and climate change are aerosols. Aerosols include particles of varying sizes and compositions that are small enough to be carried aloft in the atmosphere. As discussed in more detail in Section 2.2.2, aerosols influence the climate system by interacting with solar and terrestrial radiation, influencing the planet's energy balance.

2.1.2 The Biosphere

The biosphere includes all living organisms on the planet, i.e., all ecosystems, including humans. Organisms in the biosphere that have fundamentally shaped our planet are called *photoautotrophs*, denoting organisms capable of synthesising sugar from CO_2 and H_2O using solar radiation, and releasing O_2 as a byproduct through the process of photosynthesis [Hall and Rao, 1999]. These

organisms are abundant in large quantities in both the ocean (marine biosphere) and on land (terrestrial biosphere). The marine biosphere exhibits strong activity within the top 100 meters of the oceans, where light penetrates the water, known as the euphotic zone. Within this layer, phytoplankton thrive by consuming nutrients and dissolved CO_2 , while generating O_2 .

The terrestrial biosphere includes all land vegetation, typically manifested in the form of forests, grasslands, tundra, and deserts. Plant species engage in ongoing competition for space, a process governed by factors such as water availability, insolation, and air temperatures. Consequently, climatic conditions in different regions give rise to distinct biomes characterised by unique combinations of plant species. Fig. 1 shows how climatic conditions (i.e., the mean annual temperatures and precipitation) dictate the biome present in a particular region [Whittaker and Likens \[1975\]](#).

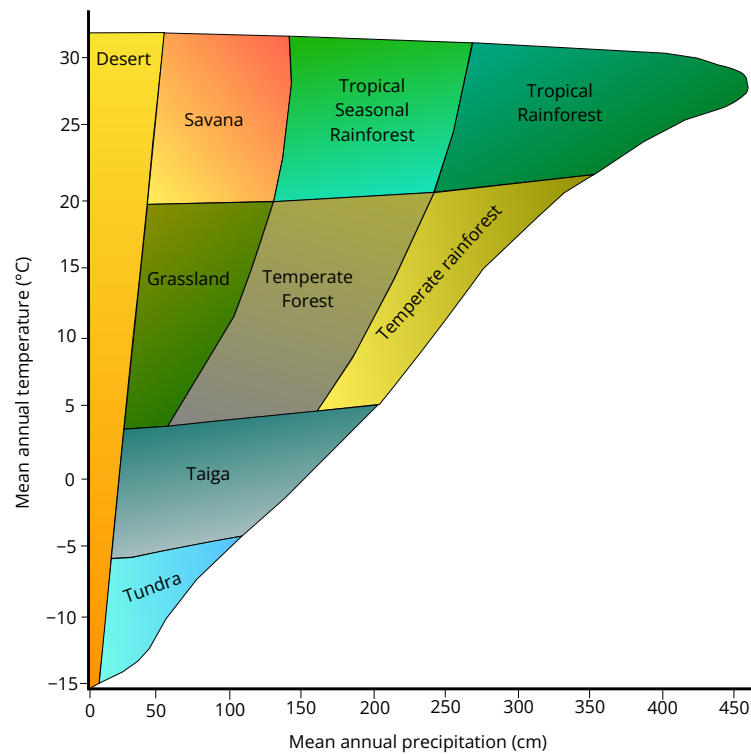


Figure 1: Classification of biomes based on temperature and precipitation. Figure by the author, based on [\[Whittaker and Likens, 1975\]](#).

2.2 The biosphere-atmosphere interface

The land-biosphere-atmosphere interface is fundamentally important in the Earth system as it involves bidirectional exchanges of energy and material between the land surface and the atmospheric boundary layer. Such interactions have a significant influence on major biogeochemical cycles of water, CO_2 , nutrients and trace gases within the Earth system with subsequent effects on the chemistry of the atmosphere and the radiation budget. The scientific investigation of land-biosphere-atmosphere interactions, therefore, contributes to a broader understanding of the Earth system as a whole.

2.2.1 Biogenic volatile organic compounds (BVOC)

The terrestrial biosphere is a primary source of volatile organic compound emissions (VOCs) such as isoprene and various terpenes, accounting for around 90% of the total VOC emissions to the atmosphere [Guenther et al., 1995]. Present annual total BVOC emissions (isoprene + monoterpenes + other reactive BVOCs) are estimated to range between 700 to 1000 Tg (C) [Guenther et al., 2006, Laothawornkitkul et al., 2009, Sindelarova et al., 2014]. The mechanisms behind BVOC emission from plants are complex and still not fully understood. However, it seems that BVOCs play a role in protecting photosynthetic activity in plants from damage caused by reactive oxygen species, which are generated in leaves at high temperatures [Harrison et al., 2013, Lantz et al., 2019, Niinemets, 2010]. The simplest and most abundant BVOC is isoprene (C_5H_8). More complex BVOCs are grouped into monoterpenes ($C_{10}H_{16}$), composed of two isoprene units, and sesquiterpenes ($C_{15}H_{24}$), composed of three isoprene units. Limonene, α -pinene, β -pinene, myrcene, sabinene, 3-carene, and ocimene are well-known examples of monoterpenes [Geron et al., 2000]. β -Caryophyllene is among the most abundant sesquiterpenes emitted [Gao et al., 2022].

Given the relative abundance of BVOC emissions and their high reactivity, BVOCs have been found to exert a significant global influence on the atmosphere. The atmospheric chemical lifetime of BVOCs ranges from minutes to hours [Atkinson, 2000], as they rapidly interact with various tropospheric species such as carbon monoxide, hydroxyl radicals (OH), methane, and ozone [Atkinson and Arey, 2003, Granier et al., 2000, Lelieveld et al., 1998, 2008, Poisson et al., 2000]. OH radicals are known as the “detergent” of the atmosphere given their ability to oxidise organic compounds in the atmosphere. Therefore, the prevalence of BVOCs influences the concentrations of OH, consequently affecting other greenhouse gases like methane, and ozone [Gligorovski et al., 2015]. BVOCs also play an important role in recycling OH through secondary reactions, thereby contributing to lower net OH consumption in a process known as OH recycling [Lelieveld et al., 2016]. Moreover, BVOC oxidation leads to aerosol formation, which has implications for planetary radiation and climate. This is discussed in more detail in Section 2.2.2.

2.2.2 Biogenic secondary organic aerosols (BSOA)

BVOCs undergo a series of stepwise degradation processes that lead to the formation of extremely low volatile organic compounds (ELVOCs). The initial oxidation of BVOCs yields first-generation products, which are organic compounds containing single or multiple oxygenated functional groups. These first-generation products, for instance, products from the isoprene+OH reaction, do not typically partition directly into the particle phase [Pandis et al., 1991]. Subsequent oxidation of these first-generation compounds results in the formation of highly volatile and soluble second-generation organic compounds, characterised by the addition of further oxidised functional groups. The volatility of these oxygenated products varies, with lower-chain compounds being more volatile than their higher or longer-chain counterparts [Mahilang et al., 2021]. ELVOC compounds exhibit extremely low volatility due to the presence of a large number of functional groups, resulting in a high oxygen-to-carbon ratio (O/C ratio > 0.7) in the intermediate species, which significantly reduces their volatility [Mutzel et al., 2015]. These intermediate species, also known as highly oxygenated molecules (HOMs), tend to partition into the aerosol phase, leading to the formation of biogenic secondary organic aerosols (BSOA) [Kavouras et al., 1998, Spracklen et al., 2011].

Isoprene-derived oxidation products primarily condense onto existing aerosols. In contrast, although the oxidation products from monoterpenes are less abundant, they play a crucial role in generating new particles and significantly influence SOA formation [Jokinen et al., 2015]. Biogenic SOA can grow sufficiently to reach a dry radius of approximately 50 nm, contributing to the aerosol fraction involved in climate interactions. In other words, bSOA participates in the absorption and scattering of radiation (direct effect) and acts as condensation nuclei, modifying cloud properties, such as albedo and lifetime (indirect effect) [Forster et al., 2007]. Once SOA is formed in the particle phase, its fate in the atmosphere includes particle growth, ageing, and eventual removal through volatilisation, i.e., the transition to the gas phase, as well as dry and wet deposition processes [Wong et al., 2019].

2.3 Perturbations in the Climate System

The Earth’s climate has remained stable over geological time scales, largely due to negative feedback mechanisms within the carbon cycle [Tajika, 2007]. Significant temperature fluctuations have been driven by sudden changes in atmospheric composition resulting from volcanic activity (affecting CO₂ concentrations) and long-term variations in insolation caused by Earth’s orbital cycles, as described by the Milankovitch theory [Holland, 1978, Milankovitch, 1941, Spiegel et al., 2010]. The present-day climate state is also governed by many feedback mechanisms, including atmospheric processes and interactions with the oceans and biosphere. Efforts have been made to study these interactions in detail and understand how they are affected by present-day perturbations in the climate system [e.g., Bonan, 2008, Cai et al., 2015]. Some of these perturbations, such as volcanic eruptions, forest fires, and the El Niño Southern Oscillation (ENSO), are natural. Other perturbations involve human influence, such as anthropogenic emissions altering atmospheric composition, and changes in the biosphere driven by human land cover changes. Additionally, the rapid increase in planetary temperatures from greenhouse gas emissions and other human activities is exacerbating the seasonality and intensity of natural disturbances like forest fires and ENSO. This work investigates the influence of ENSO and its expected intensification due to climate change on BVOC emissions (Section 3.3). Additionally, it examines the impact of human land cover changes, such as deforestation, on BVOC emissions and aerosol burden (Section 3.4).

2.3.1 The El Niño-Southern Oscillation

The El Niño–Southern Oscillation (ENSO) is a periodic oscillation (occurring every 2 to 7 years) between anomalously warm (El Niño) and cold (La Niña) sea surface temperatures (SSTs) in the tropical Pacific [McPhaden et al., 2006]. Because of the strong interactions between atmospheric and oceanic circulations, such anomalies in tropical Pacific SST have a significant impact on atmospheric processes. ENSO therefore exerts a marked influence on weather systems and global climate patterns [McPhaden et al., 2006].

Fig. 2 illustrates the ENSO cycle of warming and cooling SSTs in the tropical Pacific Ocean. Under normal conditions, the equatorial Pacific trade winds blow from east to west, pushing warmer water towards the west and causing colder water to rise from deeper in the ocean to replace it. This creates an east-west difference in SST that sustains the trade winds, driving a positive feedback loop. During a La Niña year, the east-west temperature difference intensifies, the pressure difference

increases, and the trade winds and their effects on the ocean strengthen, leading to further cooling of the east Pacific. Conversely, during an El Niño year, the east-west SST difference weakens, the pressure difference decreases, and the trade winds weaken and reverse direction. This leads to the Walker circulation convective centres re-arranging, inducing precipitation anomalies in the tropics and influencing monsoon systems via the Hadley circulation over the Pacific, Indian, and Atlantic Oceans. Teleconnections with midlatitude westerlies can also result in consistent anomaly patterns in the extratropics [Dai and Wigley, 2000]. To this end, tropical regions are often much warmer and drier than average during El Niño episodes [Dai and Wigley, 2000, Gong and Wang, 1999], but some regions tend to be cooler and wetter, e.g., western North America [Ropelewski and Halpert, 1986] and East Asia [Wu et al., 2003]. The perturbed atmospheric states from ENSO propagate to the biosphere influencing BVOC emissions. In this work, the impacts of ENSO on global BVOC budgets are investigated, as presented in Section 3.3.

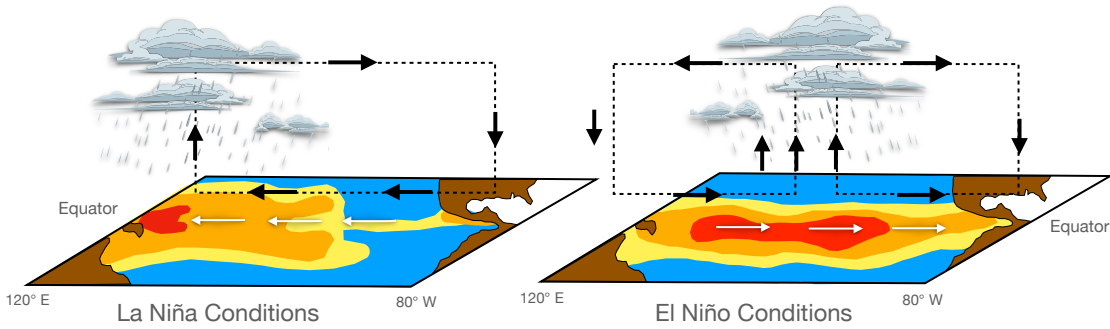


Figure 2: Sea surface temperatures, wind patterns, and areas of rising air in the tropical Pacific during La Niña and El Niño conditions. Figure by the author, based on an image by NOAA (Dr Michael J. McPhaden).

2.3.2 Human land cover change

Human activities have greatly modified the natural vegetation cover, primarily through the conversion of forests into agricultural land. It is estimated that approximately half of the Earth’s land surface has been affected by human activities [Hurt et al., 2011]. Land cover change (LCC) has a substantial impact on the Earth system as the biosphere plays a central role in major biophysical and biogeochemical cycles, as well as feedbacks with the atmosphere [Bonan, 2008]. Forests store 45% of terrestrial carbon and can sequester large amounts of carbon annually [Field and Raupach, 2004]. Furthermore, forests play a significant role in sustaining the hydrological cycle through evapotranspiration, which influences cloud formation, the onset of precipitation, and consequently surface temperatures [Betts et al., 2004, Vicente-Serrano et al., 2015]. Land cover plays a crucial role in surface albedo, with dense forests capable of absorbing up to 90% of solar radiation [Forster et al., 2007]. In high-latitude areas, forests may reduce the reflective properties of snow, ultimately contributing to planetary warming by increasing the absorption of solar energy by the land surface [Bonan, 2008]. This thesis includes an investigation (Section 3.4) into changes in BVOC emissions resulting from deforestation and their subsequent impact on aerosols.

2.4 Biosphere-atmosphere feedbacks via BVOC emissions

It has been suggested that elevated temperatures due to climate change lead to increased BVOC emissions, potentially setting off a negative climate feedback loop [Kulmala et al., 2004, 2013]. The elevated BVOC emissions would result in a higher aerosol number and mass concentration, consequently causing a cooling effect by altering cloud properties [Albrecht, 1989, Twomey, 1974]. BVOC-aerosol-climate interactions occur via three feedback loops, two negative and one positive [Sporre et al., 2019]. Increased BVOC results in higher aerosol yields which contribute to a higher aerosol optical depth (AOD) as well as more CCN. The increased AOD results in a direct surface cooling effect, thus reducing BVOC emissions (negative feedback loop). Additionally, the increase in CCN will cause clouds to have a greater CDNC and smaller droplets, ultimately resulting in a higher cloud albedo [Twomey, 1974]. An increased cloud albedo and prolonged cloud lifetime contribute to a surface temperature decrease which leads to lower BVOC emissions, thereby inducing a second negative feedback loop. The prolonged lifetime of clouds plays a crucial role in determining the onset of precipitation [Albrecht, 1989]. Alterations in precipitation patterns can significantly impact BVOC emissions, both directly through changes in soil moisture (as BVOC emissions are responsive to drought conditions [Byron et al., 2022, Šimpraga et al., 2011]), and over extended periods by influencing shifts in vegetation patterns. Nevertheless, to my knowledge, feedbacks between precipitation changes and BVOC emissions has not been assessed in the current literature.

The final feedback loop involves the impact of aerosol particle scattering on plant production. The increased aerosol mass and number concentrations not only cool the surface but also scatter more the surface radiation resulting in higher diffuse radiation relative to the global radiation. This increase in diffuse radiation boosts photosynthesis through increased photosynthetically active radiation in shaded regions of the canopy [Roderick et al., 2001]. However, in contrast to what was reported by Rap et al. [2018], the influence of diffused radiation on both plant production and BVOC emissions appears to be minimal, making this feedback loop largely insignificant [Sporre et al., 2019]. The feedbacks are summed up in Fig. 3.

2.5 Earth System Modelling

Computer models for the climate system were initially designed to simulate separate spheres in the Earth system individually. Such models include the general circulation models (GCMs) for the atmosphere and the ocean representing the physical (hydrodynamical) processes as well as other models describing different physical, chemical and biological cycles within the Earth system. Such models work with predetermined boundary conditions at the interface with other domains in the Earth system. Earth system models (ESMs), however, link all relevant components of the Earth system and thus incorporate the dynamic interactions and feedbacks between the previously separated domains. This makes ESMs highly comprehensive tools ideal for modelling past and future climate change with biogeochemical feedbacks and also for studying biosphere-atmosphere feedbacks explicitly [Flato et al., 2014]. Especially, these tools allow for a highly interdisciplinary research approach linking several fields of Earth System Sciences, e.g., in this case, vegetation dynamics (biology & ecology), atmospheric aerosols, and climate science (atmospheric chemistry & physics).

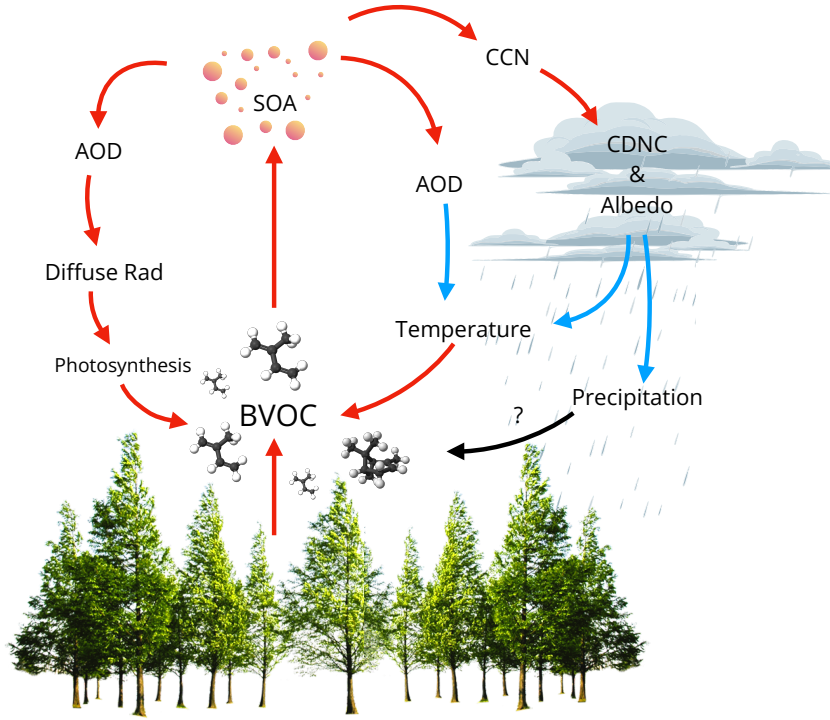


Figure 3: Biosphere-atmosphere feedbacks driven by BVOC emissions and SOA. Red arrows indicate that when the initial variable increases, it is expected that the subsequent variable will also increase. Conversely, blue arrows suggest that an increase in the initial variable will likely cause a decrease in the subsequent variable. Figure by the author, with feedbacks adopted from [Kulmala et al. \[2014\]](#), [Sporre et al. \[2019\]](#).

2.5.1 The EMAC model

The EMAC (ECHAM/MESSy Atmospheric Chemistry) model is a numerical chemistry and climate modelling system that contains submodels that represent tropospheric and middle atmospheric processes, as well as their interactions with oceans, land, and anthropogenic activities. It originally combined the ECHAM atmospheric GCM [[Roeckner et al., 2006](#)] with the Modular Earth Submodel System (MESSy) [[Jöckel et al., 2005](#)] framework and philosophy, modularising physical processes as well as most of the infrastructure into submodels that can be further developed to improve existing process representations; new submodels can also be added to represent new or alternative process representations (Fig. 4).

The MESSy structure involves 4 layers [[Jöckel et al., 2005](#)]:

- Base Model Core Layer (BMCL): This layer consists of a base model without all modular components. Currently, the general circulation model (GCM) ECHAM5 serves as the base model in EMAC.
- Base Model Interface Layer (BMIL): This layer has three primary functions: (a) enabling the base model to control the submodels (i.e., switching and calling them), (b) managing data transfer between the submodels and the base model as well as among different submodels, and (c) importing gridded initial and time-dependent boundary conditions.

- Submodel Interface Layer (SMIL): This layer facilitates the transfer of information from the BMIL to the core routines within the Submodel Core Layer (SMCL) via parameter lists and also distributes the calculated results back to the BMIL. Additionally, SMIL handles the couplings between different submodels.
- Submodel Core Layer (SMCL): This layer comprises the core routines of the submodels, representing individual processes. These routines are self-consistent, meaning their output is exclusively defined by their numerical input, and they are independent of the base model.

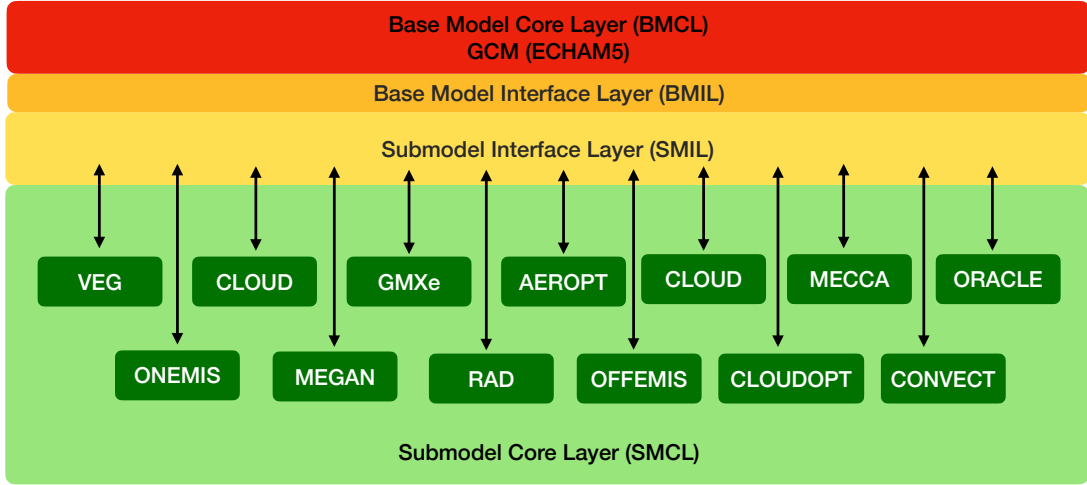


Figure 4: The MESSy framework with its four layers (BMCL, BMIL, SMIL, SMVL) and some submodels that could be used. Submodels could be switched on/off depending on the study requirements and new submodels could be developed and implemented. Figure by the author, adapted from Jöckel et al. [2005].

The combination of individual processes (from different submodels) into one simulation in MESSy is based on the operator splitting concept, where each process sequentially adds its individual contribution to a prognostic variable's total tendency [Eichinger and Jöckel, 2014, Jöckel, 2012]. The total tendency, derived from the sum of individual tendencies and the initial or previous values of the variable, is updated at each integration step. This method allows the temporal evolution of prognostic variables to be computed in sequential steps, with each step corresponding to a specific submodel. The submodels used in this work are summarised in Table 1.

LPJ-GUESS

The Lund-Potsdam-Jena General Ecosystem Simulator (LPJ-GUESS) [Smith et al., 2001, 2014] is a dynamic global vegetation model (DGVM) featuring an individual-based model of vegetation dynamics. These dynamics are simulated as the emergent outcome of growth and competition for light, space, and soil resources among woody plant individuals and a herbaceous understorey in each of a number (50 in this work) of replicate patches representing random samples of each simulated locality or grid cell. The simulated plants are classified into 12 plant functional types

Submodel	Function	Reference
AEROPT	Aerosol optical properties	Dietmüller et al. [2016]
AIRSEA	Air-Sea exchange	Pozzer et al. [2006]
BIOBURN	Biomass burning	Cabrera-Perez et al. [2016]
CLOUD	Large-scale clouds	Roeckner et al. [2004]
CLOUDOPT	Cloud optical properties	Dietmüller et al. [2016]
CONVECT	Convective clouds	Tost et al. [2006b]
CVTRANS	Convective transport of tracers	Tost et al. [2010]
DDEP	Dry deposition	Kerkweg et al. [2006]
DRADON	Emission and decay of ^{222}Rn	Jöckel et al. [2016]
E5VDIFF	ECHAM5 vertical diffusion	Roeckner et al. [2003a]
GMXe	Aerosol microphysics and gas/aerosol partitioning	Pringle et al. [2010]
GWAVE	Gravity waves	Hines [1997]
HD	Hydrological discharge	Pozzer et al. [2011]
IONS	Ionisation for the atmosphere	Ehrhart et al. [2018]
JVAL	J values	Sander et al. [2014]
LNOX	Lighting NOx	Tost et al. [2007]
MECCA	Tropospheric chemistry	Sander et al. [2011]
MEGAN	Biogenic emissions	Pozzer et al. [2022]
OFFEMIS	Offline emissions	Kerkweg et al. [2006]
ONEMIS	Online emissions	Kerkweg et al. [2006]
ORACLE	Organic aerosols	Tsimpidi et al. [2014]
ORBIT	Orbital parameters	Dietmüller et al. [2016]
OROGW	Orographic gravity wave drag	Roeckner et al. [2003a]
QBO	Quasi-biennial Oscillation nudging	Jöckel et al. [2016]
RAD	Radiative transfer	Dietmüller et al. [2016]
S4D	4D sampling	Jöckel et al. [2016]
SCAV	Scavenging	Tost et al. [2006a]
SCOUT	Selectable column output	Jöckel et al. [2016]
SEDI	Aerosol sedimentation	Kerkweg et al. [2006]
SURFACE	Surface processes	Jöckel et al. [2016]
TNUDGE	Tracer nudging	Kerkweg et al. [2006]
TROPOP	Tropopause	Jöckel et al. [2006]
VEG	Vegetation calculations (LPJ-GUESS)	Forrest et al. [2020] , Vella et al. [2023]

Table 1: Description of submodels in EMAC used in this work.

(PFTs) discriminated by growth form, phenology, photosynthetic pathway (C3 or C4), bioclimatic limits for establishment and survival and, for woody PFTs, allometry and life history strategy. In EMAC, LPJ-GUESS is employed via the submodel VEG. The vegetation in LPJ-GUESS is entirely determined by the EMAC atmospheric state, soil type, N deposition, and fluxes [[Forrest et al., 2020](#)], but there is no feedback from the vegetation to climate variables (e.g., changes in albedo and roughness length). The only feedback to the atmosphere occurs via BVOC emissions as described in Section 3.2. After each simulation day, EMAC computes the average daily values of 2-meter temperature, net downwards shortwave radiation, and total precipitation and passes these state variables to LPJ-GUESS. The current state of coupling in the EMAC/LPJ-GUESS configuration enables the investigation of the role of biogenic volatile organic compounds in aerosol production and their contribution to cloud formation and precipitation patterns. Further coupling would enable the study of the methane cycle and its lifespan, the atmospheric chemistry of various carbon compounds, the impact of fires and associated feedbacks, future nitrogen deposition rates and fertilization scenarios, and the effects of ozone on plants.

BVOC emission algorithms (ONEMIS & MEGAN)

BVOC emissions in EMAC can be represented as offline emissions (via OFFEMIS), or calculated online via ONEMIS or MEGAN. Both ONEMIS and MEGAN are based on the algorithms proposed by [Guenther et al. \[1995, 1993\]](#), where the BVOC emission flux (F) is calculated as a function of the foliar density and its vertical distribution (D [kg dry matter m⁻²]), ecosystem-specific emission factors (ϵ), and a non-dimensional activity factor (γ) that accounts for the photosynthetically active radiation (PAR) and temperature:

$$F = [D] [\epsilon] [\gamma] \quad (1)$$

The schemes mostly differ in the evaluation of the canopy process for light and temperature sensitivity on emission yields. In ONEMIS, fluxes are a function of foliar density, plant-specific emission factors and an activity factor accounting for light and temperature sensitivity. Emissions are calculated within four distinguished layers of the canopy, expressed by the leaf area density (LAD) and leaf area index (LAI). For each layer, the extinction of photosynthetically active radiation (PAR) is calculated from the direct, visible radiation and the zenith angle. The fractions of sunlit leaves and the total biomass are then used to calculate emissions from sunlit and shaded leaves within the canopy. On the other hand, fluxes in MEGAN are a function of the LAI, plant-specific emission factors, light, temperature and wind conditions within the canopy, leaf age and soil moisture. In the current MEGAN version in EMAC, the parameterised canopy environment emission activity (PCEEA) algorithm is used rather than the alternative detailed canopy environment model that calculates light and temperature at each canopy depth. The PCEEA algorithm calculates the light sensitivity within the canopy as a function of the daily average above-canopy photosynthetic photon flux density (PPFD), the solar angle and a non-dimensional factor describing the PPFD transmission through the canopy. Further technical details for canopy processes employed in ONEMIS can be found in [Ganzeveld et al. \[2002\]](#), while [Guenther et al. \[2006\]](#) provide details for MEGAN. Work from this thesis linked BVOC emissions via ONEMIS and MEGAN with interactive dynamic vegetation from LPJ-GUESS, allowing for interactive BVOC emissions that respond to changes in the biosphere (see Section 3.2).

Aerosols (GMXe & ORACLE)

In EMAC, aerosols are simulated using the Global Modal-aerosol eXtension (GMXe) submodel. GMXe is an advanced aerosol microphysics and gas/aerosol partitioning model [[Pringle et al., 2010](#)]. This two-moment aerosol model prognostically predicts both the number concentration and the mass mixing ratio of aerosol modes. It extends the M7 model [[Vignati et al., 2004](#)] to describe aerosol size distributions using seven interacting lognormal modes. Soluble aerosols are categorised into four hydrophilic modes based on their size spectrum: nucleation (NS, $r < 5$ nm), Aitken (KS, $5 \text{ nm} < r < 50$ nm), accumulation (AS, $50 \text{ nm} < r < 500$ nm), and coarse (CS, $r > 500$ nm), where r denotes the aerosol radius. Insoluble aerosols are classified into three hydrophobic modes

(KI, AI, CI) that share the same size ranges as the hydrophilic modes, except for the nucleation mode. Within each mode, the aerosol composition is uniform (internally mixed), but it varies between the different modes (externally mixed). GMXe simulates microphysical aerosol processes, including nucleation and coagulation. GMXe simulates six aerosol species, namely organic carbon (OC), black carbon (BC), water-soluble inorganic aerosols (WASO), dust (DU), sea salt (SS), and aerosol-associated water (H₂O).

Even though GMXe includes routines for partitioning of organic aerosol, and can therefore be used to treat OC explicitly, in this work, a separate submodel, ORACLE, was used for the calculations of organic carbon. The Organic Aerosol Composition and Evolution (ORACLE) submodel [Tsimpidi et al., 2014] takes into account the partitioning between aerosols and the gas phase based on the volatility basis set (VBS) framework [Donahue et al., 2006], in which OA is assumed to be semivolatile and photochemically reactive and is distributed in logarithmically spaced volatility bins. By integrating semi-volatile primary emissions, chemical aging, and SOA formation into a cohesive framework, the VBS approach is perfectly designed for use in regional and global chemical modelling. Organic emissions from various anthropogenic and natural sources are accounted for using distinct surrogate species for each source category. These surrogates are further classified into groups based on their volatility: low-volatility organic compounds (LVOCs, $C^* \leq 10^{-1} \mu\text{g}/\text{m}^3$), semi-volatile organic compounds (SVOCs, $10^{-1} < C^* \leq 10^2 \mu\text{g}/\text{m}^3$), intermediate-volatility organic compounds (IVOCs, $10^2 < C^* \leq 10^6 \mu\text{g}/\text{m}^3$), and volatile organic compounds (VOCs, $C^* > 10^6 \mu\text{g}/\text{m}^3$). These organic compounds can partition between the gas and aerosol phases, leading to the formation of organic aerosols (OA). An extended version of ORACLE (v2.0) has been used in this work, incorporating a 2-D VBS approach that tracks not only the saturation concentration but also the oxygen content of organic aerosols (OA) during atmospheric transport [Tsimpidi et al., 2018]. Representing the chemical aging of OA is important as it results in significant changes to physical and chemical properties due to the addition of oxygen atoms from reactions with OH, impacting OA hygroscopic growth and cloud condensation nuclei (CCN) activity [Chang et al., 2010], and influencing the phase state changes of SOA during its atmospheric lifetime, which can affect ice nuclei (IN) activity [Shiraiwa et al., 2017].

Aerosol optical properties (AEROPT)

The submodel AEROPT computes the optical properties from aerosol concentrations and size distributions simulated by an aerosol model, such as GMXe (in online mode), or using aerosol climatologies (in offline mode). The optical properties calculated by AEROPT include:

- **Aerosol optical thickness:** the total extinction due to scattering and absorption of aerosol particles in each grid box.
- **Single-scattering albedo:** the ratio of scattering to absorption by the aerosols.
- **Asymmetry factor:** describes the angular distribution of scattering intensity.

AEROPT obtains aerosol optical property information, i.e., complex refractive indices for the different aerosol species (OC, BC, WASO, DU, SS, H₂O), across various wavelength bands from a lookup table. The submodel treats all organic aerosols with a single set of optical properties,

meaning that while primary, secondary, natural, and anthropogenic organic aerosols are explicitly described in ORACLE, the same optical properties are applied to all these species.

Originally, the model used optical properties for OA based on burning vegetation, following [Sutherland and Khanna \[1991\]](#). While these properties may be suitable for general OA, they are not appropriate for describing secondary organic aerosols (SOA). Simulation runs from this work confirmed that using [Sutherland and Khanna \[1991\]](#) optical properties for biogenic SOA (bSOA) resulted in excessive absorption, as indicated by changes in the single-scattering albedo. Given that this work focuses on bSOA, the OA optical properties were updated according to [Ghan et al. \[2001\]](#). These optical properties are based on both primary emissions and secondary production aerosols and are used in the Community Atmosphere Model CAM5 [[Liu et al., 2012](#)].

Clouds (CLOUD & CLOUDOPT)

The standard stratiform cloud scheme in ECHAM5 addresses three prognostic variables (in kg kg^{-1}): specific humidity (q_v), cloud liquid water mixing ratio (q_w), and cloud ice mixing ratio (q_i). Cloud droplet number concentration (CDNC) and ice crystal number concentration (ICNC) are either prescribed or determined using simple empirical functions, while rain and snow are treated diagnostically [[Roeckner et al., 2003b](#)]. However, advancements in ECHAM have introduced a new prognostic equation for CDNC, enabling more realistic modelling of cloud droplet nucleation, advection, evaporation, and collision-coalescence [[Lohmann et al., 1999](#)]. Later, a prognostic equation for ICNC was also introduced [[Lohmann and Kärcher, 2002](#)]. The cloud parameterisation used in this work employs a two-moment stratiform cloud microphysics scheme [[Lohmann and Kärcher, 2002](#), [Lohmann et al., 1999, 2007](#)]. This scheme defines prognostic equations for in-cloud variables q_v , q_w , q_i , CDNC, and ICNC. The two-moment approach enhances the representation of aerosol-cloud interactions, leading to improved calculations of cloud microphysical processes and radiative transfer. CLOUDOPT calculates the optical properties of clouds, including cloud optical depth, single scattering albedo, and asymmetry factor, across various solar spectral bands. Specifically, it considers four bands in the shortwave spectrum and sixteen in the longwave spectrum to account for wavelength dependency [[Dietmüller et al., 2016](#)]. This cloud property information serves as an input for the radiation scheme.

Aerosol-cloud interactions

CLOUD incorporates a prognostic cloud droplet nucleation process to represent aerosol-cloud interactions in large-scale clouds (excluding convective warm clouds) using aerosol information provided by GMXe. This prognostic nucleation scheme is based on the aerosol activation parameterisation proposed by [Abdul-Razzak and Ghan \[2000\]](#) and employs the κ -Köhler method [[Petters and Kreidenweis, 2007](#)] to calculate aerosol hygroscopicity and critical supersaturation. This routine provides values for the parameter κ and the cloud condensation nuclei (CCN) number concentration at 0.2% and 0.4% supersaturation for each aerosol size mode [[Pringle et al., 2010](#)]. The resulting CCN concentrations are then utilised in the two-moment cloud scheme described above, effectively impacting cloud properties (i.e., CDNC).

3 Results

3.1 Overview

This thesis consists of four studies, two of which have been published in peer-reviewed journals. The third study is currently being prepared for submission to the journal *Atmospheric Chemistry & Physics*. While the fourth study requires further work before submission, preliminary results are included here as a section. I am the lead author of all studies presented in this thesis. While the research questions were conceptualised in collaboration with my supervisors, I conducted the modelling work to generate the data, analysed the model outputs, produced the figures, interpreted the results, wrote the papers, and revised the published papers according to the referees' comments during the review process. A more detailed overview of the author's contribution is included in each section. Supplementary material can be found in Section 5. All studies presented here investigate the interplay between the biosphere and atmosphere through BVOC emissions, utilising a coupled global chemistry-climate model and a dynamic global vegetation model.

Section 3.2 presents the first study of this PhD project, which has been published in the peer-reviewed journal *Geoscientific Model Development* as a model description paper. In this study, BVOC emissions in the chemistry-climate model EMAC were coupled to the global dynamic vegetation model LPJ-GUESS, allowing bidirectional interactions between vegetation and the atmosphere via BVOCs. The dynamic vegetation states were linked to the BVOC submodels ONEMIS and MEGAN. For ONEMIS, a new parameterisation was introduced in the model interface layer to calculate the leaf area density distribution across four canopy layers and the foliar density from LPJ-GUESS output. Interactive BVOC emissions from both ONEMIS and MEGAN were evaluated, with sensitivity runs including doubled CO₂ scenarios in the vegetation and radiation schemes to investigate the sensitivity of BVOC emissions to CO₂ fertilisation and increased temperatures. Section 3.3 includes a study published in the peer-reviewed journal *Biogeosciences* as a research article. In this work, the new setup with interactive vegetation and BVOC emissions was used to evaluate global changes in BVOC emissions resulting from the El Niño-Southern Oscillation (ENSO). The study includes a thorough examination of the changes in the atmospheric states from ENSO, and how these changes propagate to changes in vegetation and BVOC emissions.

Section 3.4 presents a manuscript that will be submitted to the peer-reviewed journal *Atmospheric Chemistry & Physics*. This study examines the impact of deforestation on natural land vegetation cover and atmospheric aerosols. A deforestation routine was implemented in LPJ-GUESS to simulate deforestation scenarios, and the resulting changes in BVOC emissions and secondary organic aerosols were assessed. The study includes a comprehensive analysis of changes in atmospheric states, such as organic and total aerosol burdens, aerosol-cloud interactions, and the corresponding changes in radiative forcing.

Section 3.5 presents results from an investigation focusing on BVOC feedbacks emerging from changes in aerosols. BVOC emissions can contribute to changes in the global aerosol burden. Increased SOA alter atmospheric states, such as scattering radiation, surface cooling, and changes in cloud properties. It is hypothesised that these changes can influence vegetation, thus leading to BVOC emissions feeding back on these alterations. The model setup from this work was utilised to evaluate such feedbacks.

Fig. 5 summarises the work in this thesis. The first panel depicts the model development work

undertaken to link BVOC emissions in EMAC with dynamic vegetation states from LPJ-GUESS (Section 3.2). The middle panel in Fig. 5 illustrates the study in Section 3.3, where the response of BVOC emissions to ENSO was evaluated. The figure's right panel shows results from Sections 3.4 and 3.5, where changes in atmospheric aerosols from deforestation and BVOC-aerosol feedbacks were explored.

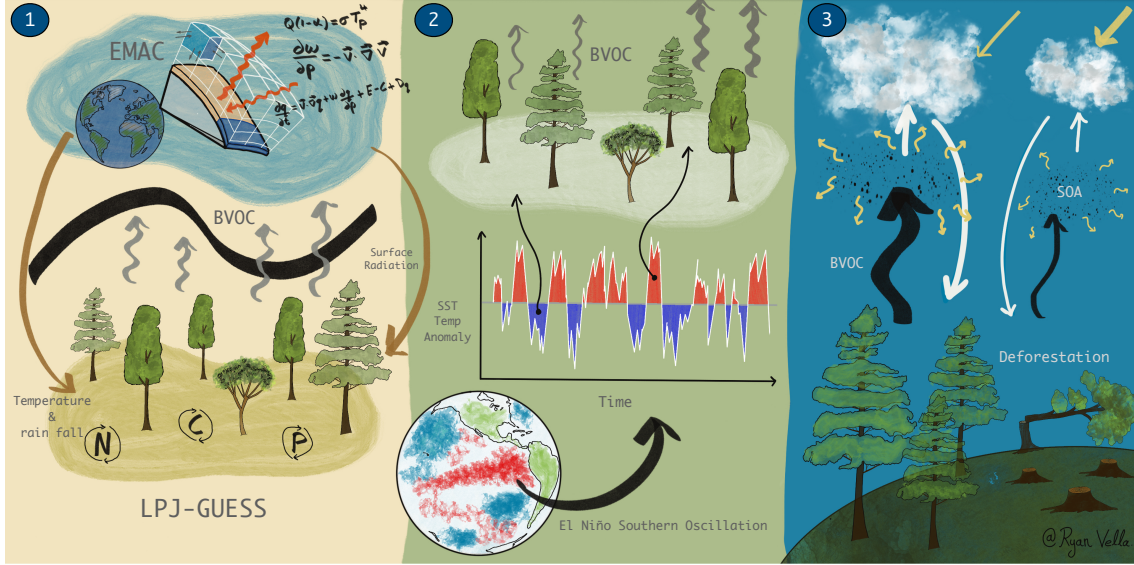


Figure 5: Summary of the results in the thesis. 1. Coupling of EAMC and LPJ-GUESS for BVOC emissions, 2. BVOC changes from ENSO, 3. Changes in aerosols from deforestation and BVOC-aerosol feedbacks.

3.2 Coupling of LPJ-GUESS/EMAC for interactive BVOC emissions

This chapter has been published in the journal *Geoscientific Model Development* as a model description paper. I am the lead author of this work, where I worked on the model setup, including code implementations for a new parameterisation. Additionally, I conducted all simulations, analysed the data, generated figures, and wrote the manuscript. The co-authors contributed to implementing the new code, interpreting the results, and proofreading the manuscript. The paper includes an *Author contributions* detailing individual contributions.

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Model Development

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Isoprene and monoterpene simulations using the chemistry–climate model EMAC (v2.55) with interactive vegetation from LPJ-GUESS (v4.0)

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Abstract. Earth system models (ESMs) integrate previously separate models of the ocean, atmosphere and vegetation into one comprehensive modelling system enabling the investigation of interactions between different components of the Earth system. Global isoprene and monoterpene emissions from terrestrial vegetation, which represent the most important source of volatile organic compounds (VOCs) in the Earth system, need to be included in global and regional chemical transport models given their major chemical impacts on the atmosphere. Due to the feedback of vegetation activity involving interactions with weather and climate, a coupled modelling system between vegetation and atmospheric chemistry is recommended to address the fate of biogenic volatile organic compounds (BVOCs). In this work, further development in linking LPJ-GUESS, a global dynamic vegetation model, to the atmospheric-chemistry-enabled atmosphere–ocean general circulation model EMAC is presented. New parameterisations are included to calculate the foliar density and leaf area density (LAD) distribution from LPJ-GUESS information. The new vegetation parameters are combined with existing LPJ-GUESS output (i.e. leaf area index and cover fractions) and used in empirically based BVOC modules in EMAC. Estimates of terrestrial BVOC emissions from EMAC’s submodels ONEMIS and MEGAN are evaluated using (1) prescribed climatological vegetation boundary conditions at the land–atmosphere interface and (2) dynamic vegetation states calculated in LPJ-GUESS (replacing the “offline” vegetation inputs). LPJ-GUESS-driven global emission estimates for isoprene and monoterpenes

from the submodel ONEMIS were 546 and 102 Tg yr^{−1}, respectively. MEGAN determines 657 and 55 Tg of isoprene and monoterpene emissions annually. The new vegetation-sensitive BVOC fluxes in EMAC are in good agreement with emissions from the semi-process-based module in LPJ-GUESS. The new coupled system is used to evaluate the temperature and vegetation sensitivity of BVOC fluxes in doubling CO₂ scenarios. This work provides evidence that the new coupled model yields suitable estimates for global BVOC emissions that are responsive to vegetation dynamics. It is concluded that the proposed model set-up is useful for studying land–biosphere–atmosphere interactions in the Earth system.

1 Introduction

The land surface of the Earth is dominated by vegetation, with forests covering $\sim 42 \times 10^6$ km² in tropical, temperate and boreal regions, making up ~ 30 % of the total land area (Bonan, 2008). The terrestrial biosphere is known to be a primary source of volatile organic compounds (VOCs) such as isoprene and various terpenes, accounting for around 90 % of the total VOC emissions to the atmosphere (Guenther et al., 1995). The processes driving VOC emissions from plants are complex and not fully understood; however, biogenic volatile organic compounds (BVOCs) seem to play a role in protecting photosynthetic activity in plants from damage caused by

reactive oxygen species, which are synthesised in leaves at high temperatures (Niinemets, 2010; Harrison et al., 2013; Lantz et al., 2019). BVOC emissions can also be triggered by other chemical, physical or biological stresses and processes, e.g. herbivory (Laothawornkitkul et al., 2008), signalling between organisms (Zuo et al., 2019) or also oxidative stress originating from the atmosphere (e.g. under elevated ozone concentrations; Sharkey et al., 2008). Plants emit an array of VOCs, but different plant species emit different compounds according to their evolutionary adaptation. For example, the emission of isoprene can be considered an evolutionary trait that benefits certain plant species in hot, dry environments (Taylor et al., 2018). Isoprene and monoterpenes are the most abundant species among the BVOCs (Kesselmeier and Staudt, 1999; Lathiere et al., 2006; Guenther et al., 2012), and their high reactivity exerts a significant influence on atmospheric composition (Atkinson, 2000). The atmospheric chemical lifetime of such BVOCs ranges from minutes to hours (Atkinson and Arey, 2003), as they quickly interact with tropospheric species including carbon monoxide, hydroxyl radicals and ozone (Lelieveld et al., 1998; Granier et al., 2000; Poisson et al., 2000; Pfister et al., 2008), thus altering the atmosphere's oxidation capacity. BVOCs are also the primary precursor for secondary organic aerosols (SOAs), which can exert a significant forcing on the radiative balance of Earth, both directly through scattering and absorption of solar radiation and indirectly through changing cloud properties (Rap et al., 2013; Scott et al., 2014). SOA also contributes to changes in the radiation balance by decreasing the solar near-surface direct radiation while at the same time increasing the diffusive radiation contribution (Wang et al., 2019).

The first BVOC models employed empirical relations describing the isoprene emission rate dependencies on temperature and light (Guenther et al., 1991, 1993; Tingey et al., 1981) and monoterpene emission rate dependency on temperature (Evans et al., 1982; Lamb et al., 1987; Tingey et al., 1980, 1981). The formulations include a species- or vegetation-type-specific emission factor characterised from field or laboratory measurements (e.g. Lamb et al., 1985; Arey et al., 1991; Guenther et al., 1993) which is defined for arbitrarily chosen environmental conditions (usually 30 °C and 1000 µmol photons per metre squared per second; Grote and Niinemets, 2007). This approach has been extensively used to study different ecosystem types all over the world, including deserts (Geron et al., 2006), grassland (Bai et al., 2006), savanna (Guenther et al., 1999; Otter et al., 2003), Mediterranean (Cortinovis et al., 2005), tropical (Harley et al., 2004), temperate (Karl et al., 2003) and boreal forests (Westberg et al., 2000). Empirical algorithms are also presently used in well-established global BVOC models such as ONEMIS (Kerkweg et al., 2006) and MEGAN (Guenther et al., 2006). These modules are presently integrated into the modelling system considered in this study (EMAC).

The land–biosphere–atmosphere interface in models is fundamentally important for studying the fate of BVOCs in the atmosphere; yet, early models were designed to simulate single components of the Earth system in isolation, prescribing simple, non-interacting boundary conditions at the interface. However, models have become increasingly coupled, with dynamic multidirectional fluxes between the different models considered. This has yielded a new category of models that we now call Earth system models (ESMs). ESMs are highly comprehensive tools ideal for modelling past and future climate change with biogeochemical feedbacks and also for studying biosphere–atmosphere interactions explicitly (Flato et al., 2014). To this end, several modelling studies have linked atmospheric-chemistry-enabled models with dynamic vegetation models to investigate the impacts of changing vegetation cover on global atmospheric emissions, atmospheric chemistry and future climate change (e.g. Levis et al., 2003; Sanderson et al., 2003; Naik et al., 2004; Lathiere et al., 2005; Arneth et al., 2007b).

Sporre et al. (2019) employed an ESM to investigate climate forcing caused by BVOC–aerosol feedbacks, where it was determined that increased BVOC emissions and subsequent SOA formation in future climate scenarios result in -0.43 W m^{-2} stronger net cloud forcing and -0.06 W m^{-2} forcing from the direct scattering of sunlight. A new ESM that integrates the chemistry–climate model EMAC (Roegner et al., 2006; Jöckel et al., 2005) with the dynamic global vegetation model (DGVM) LPJ-GUESS (Smith et al., 2001; Sitch et al., 2003; Smith et al., 2014) has recently been developed (Forrest et al., 2020). In a first study, the coupled model gave a good representation of worldwide potential natural vegetation distribution, despite some regional variations, especially at lower spatial resolutions. This study presents further model coupling of LPJ-GUESS within the EMAC modelling system with respect to vegetation-driven emissions. New vegetation parameters are computed from LPJ-GUESS variables and used as (online) input vegetation information for empirically based BVOC modules (ONEMIS and MEGAN) in EMAC. The new vegetation-sensitive isoprene and monoterpene emission fluxes in EMAC are evaluated and compared against emissions from the semi-process-based module (Niinemets et al., 2002, 1999) in LPJ-GUESS. The new model configuration is then used to examine temperature and fertilisation effects in doubling CO₂ climate scenarios.

2 Methods

2.1 EMAC modelling system (v2.55)

The EMAC (ECHAM/MESSy Atmospheric Chemistry) model is a numerical chemistry and climate modelling system that includes submodels that describe tropospheric and middle atmosphere processes, in addition to their interac-

tions with oceans, land and anthropogenic activities. It originally combined the ECHAM atmospheric general circulation model (GCM; Roeckner et al., 2006) with the Modular Earth Submodel System (MESSy; Jöckel et al., 2005) framework and philosophy, where physical processes and most of the infrastructure has been divided into “modules”, which can be further developed to improve existing process representations, and new modules can be added to represent new or alternative process representations. EMAC has been further developed to include a broader representation of atmospheric chemistry by coupling different processes such as representations for aerosols, aerosol–radiation and aerosol–cloud interactions (e.g. Tost, 2017). In this study, version 2.55 has been utilised, which is based on the well-documented version used in comprehensive model intercomparison studies (e.g. Jöckel et al., 2016).

BVOC modules in EMAC

Both ONEMIS and MEGAN are emission modules which are based on the Guenther algorithms (Guenther et al., 1993, 1995), where emissions are calculated as a function of ecosystem-specific emission factors, surface radiation, temperature, the foliar density and its vertical distribution. The schemes mostly differ in the evaluation of the canopy process for light and temperature sensitivity on emission yields. In ONEMIS, fluxes are a function of foliar density, plant-specific emission factors and an activity factor accounting for light and temperature sensitivity. Emissions are calculated within four distinguished layers of the canopy, expressed by the leaf area density (LAD) and leaf area index (LAI). For each layer, the extinction of photosynthetically active radiation (PAR) is calculated from the direct, visible radiation and the zenith angle. The fractions of sunlit leaves and the total biomass are then used to calculate emissions from sunlit and shaded leaves within the canopy. On the other hand, fluxes in MEGAN are a function of the LAI, plant-specific emission factors, light, temperature and wind conditions within the canopy, leaf age and soil moisture. In MEGAN, the parameterised canopy environment emission activity (PCEEA) algorithm is used rather than the alternative detailed canopy environment model that calculates light and temperature at each canopy depth. The PCEEA algorithm calculates the light sensitivity within the canopy as a function of the daily average above-canopy photosynthetic photon flux density (PPFD), the solar angle and a non-dimensional factor describing the PPFD transmission through the canopy. Further technical details for canopy processes employed in ONEMIS can be found in Ganzeveld et al. (2002), while Guenther et al. (2006) provide details for MEGAN.

2.2 LPJ-GUESS DGVM (v4.0)

The following section is based on the standard copyright-free LPJ-GUESS model description template ([https://web.nateko.](https://web.nateko.lu.se/lpj-guess/resources.html)

[lu.se/lpj-guess/resources.html](https://web.nateko.lu.se/lpj-guess/resources.html), last access: 14 January 2023). The Lund-Potsdam-Jena General Ecosystem Simulator (LPJ-GUESS; Smith et al., 2001; Sitch et al., 2003; Smith et al., 2014) is a DGVM featuring an individual-based model of vegetation dynamics. These dynamics are simulated as the emergent outcome of growth and competition for light, space and soil resources among woody plant individuals and a herbaceous understorey in each of a number (50 in this study) of replicate patches representing random samples of each simulated locality or grid cell. The simulated plants are classified into 1 of 12 plant functional types (PFTs) discriminated by growth form, phenology, photosynthetic pathway (C_3 or C_4), bioclimatic limits for establishment and survival and, for woody PFTs, allometry and life history strategy. LPJ-GUESS has already been implemented in global ESMs (e.g. Weiss et al., 2014; Alessandri et al., 2017), and more recently coupled with EMAC (Forrest et al., 2020). LPJ-GUESS coupled with EMAC currently provides information on potential natural vegetation rather than present-day vegetation; hence, the current configuration cannot be validated yet. However, land use configurations are currently included in the coupled EMAC/LPJ-GUESS system, allowing for a more realistic representation of the vegetation dynamics in upcoming studies.

BVOC emission routine in LPJ-GUESS

LPJ-GUESS includes a built-in BVOC emission module for the calculation of isoprene and monoterpene emission fluxes. The submodel combines the process-based leaf-level emission model (Niinemets et al., 2002, 1999), which is also based on the Guenther algorithms, with the LPJ-GUESS vegetation model for isoprene (Arneth et al., 2007b) and monoterpene (Schurgers et al., 2009) emissions. The algorithm computes BVOC production based on photosynthetic electron flux, emission factors, temperature, seasonality and also includes a CO_2 inhibition factor on leaf production of isoprene and monoterpenes relative to the ~ 370 ppmv (parts per million by volume) $[CO_2]$ in the year 2000. Further technical details on the algorithm can be found in Hantson et al. (2017).

The algorithm also needs the daily temperature range (DTR) for the calculation of BVOC emission rates, which are typically derived from climatological datasets. In this study, the DTR (defined as the difference between the maximum and minimum daily temperature) is computed in EMAC and is passed on to LPJ-GUESS on a daily basis.

Semi-process-based BVOC emissions from the LPJ-GUESS module are only calculated within the LPJ-GUESS model part and are not integrated into or transferred to EMAC at the current stage. This also means that such emissions are only available as daily averages, in contrast to the emissions provided by ONEMIS and MEGAN in EMAC, which exhibit a diurnal cycle. Thus, having LPJ-GUESS-driven emissions from ONEMIS and MEGAN in EMAC pro-

vides more consistency, including the direct link between weather, climate (change) and the impacts on vegetation and, hence, emissions. An adaptation of this LPJ-GUESS module to the shorter (a few minutes) time step of EMAC is rather complicated, especially when the current scheme uses daily average light fluxes and a daily temperature range instead of individual snapshots of radiative fluxes and temperature. This would require a complete retuning of the emission scheme, with the only benefit being the higher temporal resolution of the emission fluxes (which cannot be utilised in LPJ-GUESS but only in EMAC). Even though the scheme of Niinemets et al. (1999) is semi-process based, the processes are also highly parameterised, such that the advantages against the Guenther et al. (1993, 1995) algorithms are also small. In this work, BVOC emissions from the LPJ-GUESS routine are used for comparison only.

2.3 LPJ-GUESS-EMAC coupling for BVOC emission estimates

2.3.1 Overview of the coupling between EMAC and LPJ-GUESS

This study is part of a roadmap in which the model integration between EMAC and LPJ-GUESS is gradually tightened in well-defined, consecutive steps. To clarify the significance of this work, the roadmap from Forrest et al. (2020) is reproduced (Fig. 1), and the modelling development and evaluation efforts in this study are highlighted. This study focuses on BVOC model processes in EMAC based on interactive vegetation from LPJ-GUESS. It extends the model coupling between EMAC and LPJ-GUESS in Forrest et al. (2020) by employing new parameterisations to calculate the foliar density and leaf area density distribution from vegetation states in LPJ-GUESS. The new parameters from LPJ-GUESS are combined with existing ones (e.g. LAI and cover fractions) and are used to run empirically based BVOC modules in EMAC (i.e. ONEMIS and MEGAN). The semi-process-based BVOC emissions from LPJ-GUESS are not integrated into EMAC and are only evaluated against the new vegetation-sensitive empirically based emissions fluxes in EMAC.

2.3.2 Simulation set-up

In this work, the coupling strategy employed in Forrest et al. (2020) is employed, where modifications are done in LPJ-GUESS, such that it provides its functionality (i.e. vegetation information) via a new submodel in the MESSy framework, yet keeping the LPJ-GUESS source code intact with minimal modification. At a regular interval (currently 24 h at 12:00 UTC) EMAC provides LPJ-GUESS with the daily mean 2 m temperature, daily mean net downwards shortwave radiation and the total daily precipitation. Daily CO₂ concentrations and nitrogen deposition are also transferred from

EMAC to LPJ-GUESS. As a result, the LPJ-GUESS land surface conditions are entirely determined by the EMAC atmospheric state and chemical fluxes. In Forrest et al. (2020), only one-way coupling was performed (see Fig. 1), which means that LPJ-GUESS calculations are entirely based on EMAC information; however, the land surface vegetation condition calculated in LPJ-GUESS has no effect on the atmospheric state in EMAC. In this study, existing LPJ-GUESS output variables (i.e. LAI and PFT fractional coverage) are utilised to calculate isoprene and monoterpene emission rates in EMAC, allowing, for the first time, dynamic vegetation information from LPJ-GUESS to be passed back to EMAC and used for BVOC fluxes calculations in ONEMIS and MEGAN. Note that the hydrological cycle uses the ECHAM5 native soil moisture, and the hydrological information from LPJ-GUESS does not feed back to the meteorology. Similarly, plant albedo and roughness height are not allowed to influence the meteorology either, but climatological values are used to drive the weather and climate conditions of EMAC in the applied model configuration, even though these links are also implemented and pending throughout the evaluation.

Figure 2 illustrates the model configuration for computing isoprene and monoterpene emissions fluxes in EMAC using the submodels of ONEMIS and MEGAN. Both models require emission factors for the various PFTs, the solar zenith angle, surface radiation and surface temperature. Additionally, ONEMIS requires the following vegetation variables: leaf area index (LAI), foliar density and leaf area density (LAD) canopy profile. In contrast, MEGAN requires the LAI and fractional coverage of broadleaf, needleleaf, grass and shrub ecosystem types. In the original set-up, the vegetation input variables are prescribed from offline climatological datasets, whereas, in the new configuration, the climatological vegetation variables are replaced with ones calculated online in LPJ-GUESS. This implies that the new set-up feeds dynamic vegetation states to the BVOC modules that are directly computed in LPJ-GUESS on a daily timescale and driven by atmospheric states and chemical fluxes in EMAC, allowing for estimates of isoprene and monoterpene emissions with dynamic vegetation. The coupling is performed from the EMAC side by implementing new calculations using LPJ-GUESS information to derive all vegetation variables needed in ONEMIS and MEGAN for the computation of BVOC emission fluxes. Details on the vegetation variables required in ONEMIS and MEGAN, in addition to the new parameterisations used for their calculation, are described in the next section.

2.3.3 Vegetation variables for the BVOC modules

- *Leaf area index.* Measurements of the number of leaves in the canopy are required for ecosystem studies such as this one. This metric is often defined as the leaf area index (LAI), which is the one-sided leaf area in the

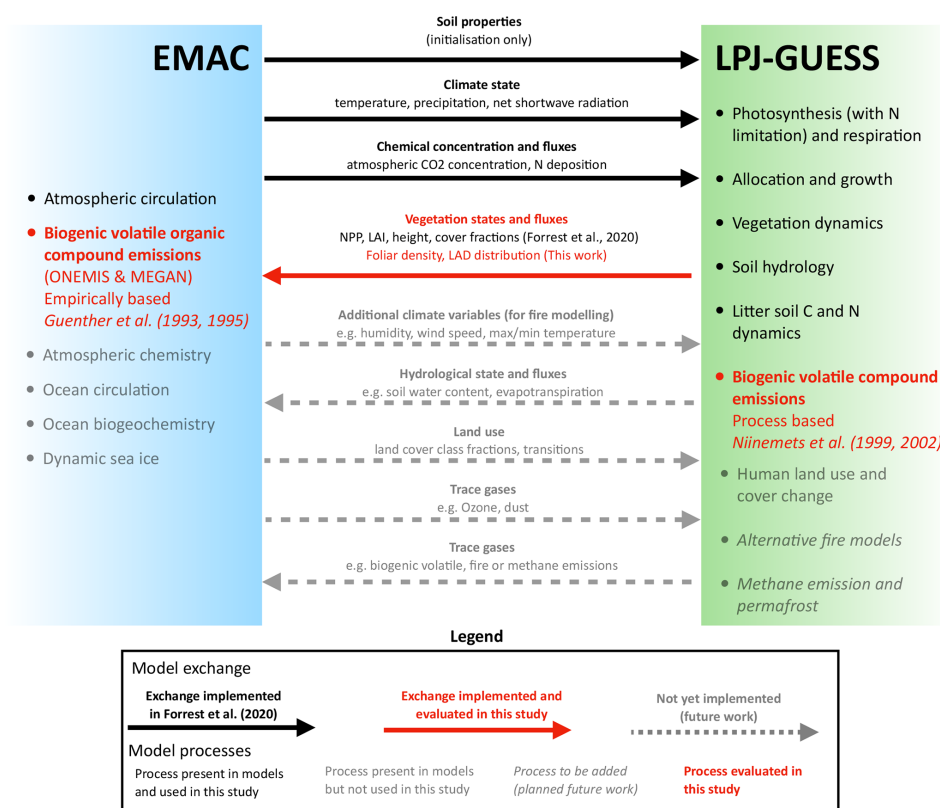


Figure 1. The main processes and exchanges in the coupled model framework adopted from Forrest et al. (2020). Model exchanges in normal black text were included in Forrest et al. (2020) and used in the simulations presented here. Exchanges in red are implemented in this work, while exchanges with grey text and grey dotted arrows are not currently included in the framework but are planned for use in future work. Model processes in normal black text were included in Forrest et al. (2020). Processes in normal grey text were included in Forrest et al. (2020) but not used here, while processes in italicised grey text are not currently included in the framework. Processes in red on the EMAC side (empirically based emissions with interactive vegetation information) are implemented and evaluated in this work, while BVOC emissions in LPJ-GUESS (semi-process based) were already implemented and are only used in this study for comparison with EMAC emissions.

canopy per unit surface area of the ground ($\text{m}^2 \text{m}^{-2}$ Jordan, 1969). In DGVMs, including LPJ-GUESS, this is a standard output variable.

- *Foliar density.* The foliar density D (grams of dry matter in metres squared), sometimes referred to as dry matter (DM), can be derived directly from the LAI as follows (Guenther et al., 1995):

$$D = \text{LAI} \cdot S_{\text{LW}}, \quad (1)$$

where S_{LW} is an average specific leaf weight (g m^{-2}) and is given for each ecosystem (or PFT) based on Box (1981).

- *Leaf area density distribution.* The leaf area density (LAD) is a metric describing the leaf area in a cubic volume within the canopy ($\text{m}^2 \text{m}^{-3}$). The original

ONEMIS configuration employs an expert-driven offline dataset from a dry deposition inferential model (DDIM; Weiss and Norman, 1985) to characterise the LAD distribution for the following three types of vegetation: (i) agricultural crops, (ii) deciduous forests and (iii) coniferous forests. The 12 PFTs (used in the applied LPJ-GUESS set-up) were classified into these three groups, with grass PFTs included in the “agricultural crop” category. The LAD distribution for each of the vegetation types is divided into four equal layers, i.e. LAD 1 (top layer), LAD 2, LAD 3 and LAD 4 (bottom layer).

These values are then used in ONEMIS for calculating the photosynthetic active radiation (PAR) within the canopy and subsequent BVOC emission estimates. For the LPJ-GUESS output, a parameterisation derived

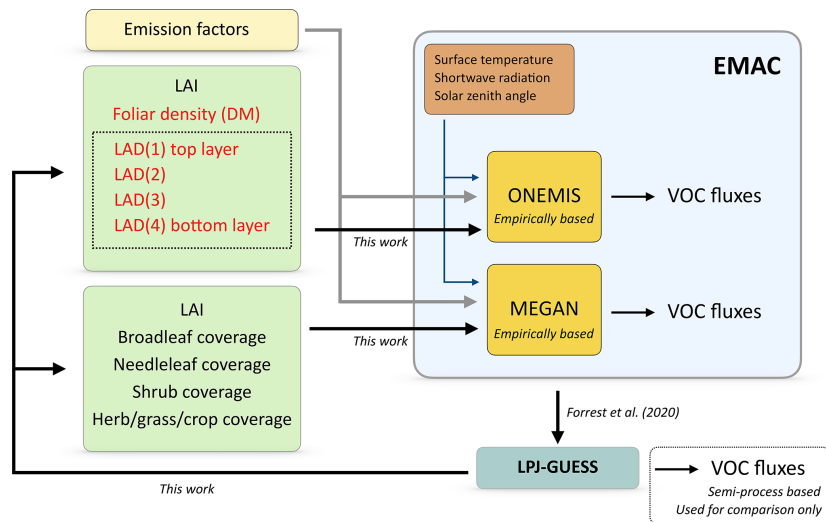


Figure 2. Model set-up for BVOC emission estimates in EMAC. The vegetation variables needed in ONEMIS and MEGAN are now provided by LPJ-GUESS, replacing offline climatological datasets. Vegetation variables that were already available at the interface between EMAC and LPJ-GUESS are in normal black text, while the new vegetation information derived in this work (i.e. foliar density and the LAD distribution) are in red text. Note that, in EMAC, only empirically based emissions are considered. VOC fluxes from the semi-process-based module in LPJ-GUESS are only used for comparison.

by Lalic et al. (2013) is employed to compute similar LADs at four canopy levels in the interface submodel between EMAC and LPJ-GUESS. The empirical relation describes the LAD at a height z (in m) as a function of the maximum LAD (LAD_{max}), the canopy height h and the height z_m corresponding to LAD_{max} (see Fig. 3).

$$LAD(z) = LAD_{max} \left(\frac{h - z_m}{h - z} \right)^n \times \exp \left[n \left(1 - \frac{h - z_m}{h - z} \right) \right] \quad \begin{matrix} n = 6 & \text{for } 0 \leq z < z_m \\ n = \frac{1}{2} & \text{for } z_m \leq z \leq h. \end{matrix} \quad (2)$$

First, the ratio between the canopy height (h) and the height corresponding to LAD_{max} (i.e. h/z_m) for each vegetation class using LAD canopy profiles from the DDIM is determined. The dataset has 21 layers, and the layer where LAD_{max} occurs (z_m) is utilised to compute h/z_m as follows:

- Agricultural crops, with $z_m/h = 12/21 \approx 0.57$.
- Deciduous forests, with $z_m/h = 15/21 \approx 0.71$.
- Coniferous forests, with $z_m/h = 17/21 \approx 0.81$.

LAD_{max} for each PFT is calculated from the corresponding LAI and PFT height h information from LPJ-GUESS, in addition to the ratio h/z_m , given the follow-

ing relation:

$$LAI = \int_0^h LAD dz = \int_0^h LAD_{max} \left(\frac{h - z_m}{h - z} \right)^n \times \exp \left[n \left(1 - \frac{h - z_m}{h - z} \right) \right] dz. \quad (3)$$

After a numerical value for LAD_{max} for each PFT is computed, the LAD at four canopy layers is calculated via Eq. (2) by integrating over four equal layers within the total canopy height h_{tot} . In all set-ups used in the study, the total canopy height h_{tot} is assumed to have a value of 25 m. This results in the following:

$$\begin{aligned} LAD(1) &= \int_{0.75h_{tot}}^{h_{tot}} LAD_{max} \left(\frac{h - z_m}{h - z} \right)^n \times \exp \left[n \left(1 - \frac{h - z_m}{h - z} \right) \right] dz, \\ LAD(2) &= \int_{0.5h_{tot}}^{0.75h_{tot}} LAD_{max} \left(\frac{h - z_m}{h - z} \right)^n \times \exp \left[n \left(1 - \frac{h - z_m}{h - z} \right) \right] dz, \\ LAD(3) &= \int_{0.25h_{tot}}^{0.5h_{tot}} LAD_{max} \left(\frac{h - z_m}{h - z} \right)^n \times \exp \left[n \left(1 - \frac{h - z_m}{h - z} \right) \right] dz, \end{aligned}$$

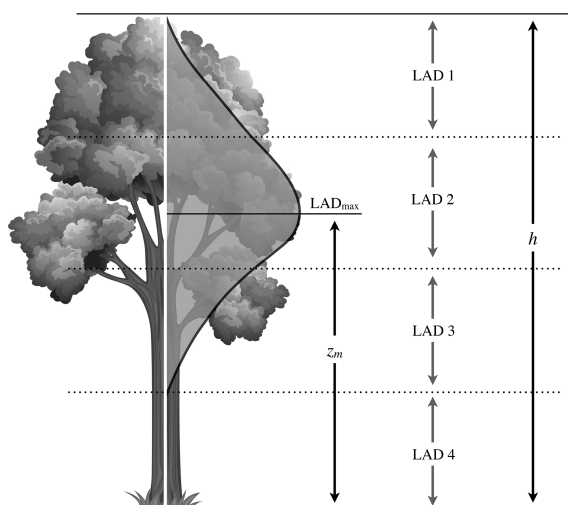


Figure 3. Graphical representation of the canopy LAD distribution. LAD_{max} is the maximum LAD within the canopy, z_m is the height from the ground at which LAD_{max} occurs, and h is the total canopy height. LAD 1, LAD 2, LAD 3 and LAD 4 are four equal canopy layers.

$$LAD(4) = \int_0^{0.25h_{tot}} LAD_{max} \left(\frac{h-z_m}{h-z} \right)^n \times \exp \left[n \left(1 - \frac{h-z_m}{h-z} \right) \right] dz, \quad (4)$$

where z_m is a fraction of h based on the PFT vegetation classes i, ii or iii, and h is the PFT height. Figure 4 compares the LAD distribution from DDIM point data, used in the previous set-up, with the new parameterisation described in Eq. (2).

- *Vegetation class coverage.* The vegetation coverage refers to the fraction of land area occupied by certain PFTs in one grid cell. This variable is used in MEGAN to adjust emission rates from different vegetation classes. This variable is already calculated in LPJ-GUESS for each of the 12 PFTs.

2.4 Set-up for double CO₂ scenarios

The submodel RAD in EMAC (Dietmüller et al., 2016) simulates the radiative transfer in the atmosphere accounting for the effects of the shortwave and longwave radiation fluxes from radiatively active trace gases. CO₂ has the largest radiative influence in the longwave range of the spectrum, resulting in radiative forcings leading to stratospheric cooling and tropospheric warming. The CO₂ value prescribed in

RAD mainly dictates the surface temperatures resulting from the greenhouse effect, while CO₂ in the vegetation scheme (i.e. in LPJ-GUESS) determines the carbon available for photosynthesis and, hence, accounts for CO₂ fertilisation effects.

Climatological monthly average sea surface temperature (SST) and sea ice content (SIC) from the Atmospheric Model Intercomparison Project (AMIP) database from 1987 to 2006 are used for Ref and Bio × 2, with a prescribed CO₂ of 348 ppmv in the radiation scheme. However, with 696 ppmv [CO₂] in the radiation scheme (i.e. in Atm × 2 and Both × 2), oceanic boundary conditions are prescribed using external data corresponding to SST and SIC at 696 ppmv to preserve radiative equilibrium. These data are acquired from a coupled atmosphere–ocean general circulation model (increased/decreased SSTs/SICs) performed under identical climate circumstances (696 ppmv [CO₂]; same approach as in Rybka and Tost, 2014).

3 Results and discussion

3.1 Vegetation characteristics as input to the emission routines

In this section, the LPJ-GUESS state variables needed as input for the BVOC routines are discussed. For that purpose, the LPJ-GUESS output is compared with the corresponding offline climatological datasets used for BVOC emission estimates in ONEMIS and MEGAN in the original model configuration i.e. with prescribed vegetation boundary conditions. For such comparisons, it has to be kept in mind that the neglect of land use in LPJ-GUESS means that currently only the natural biosphere is represented; however, climatological values for LAI also do not fully account for managed land. Nevertheless, the impact of crops is likely to modify the vegetation representation and emission results to a certain degree. The simulation results presented are from a 10-year temporal average with a 500-year offline spin-up phase at a horizontal resolution of T63 (approximately 1.9° × 1.9° at the Equator). The simulations are climatological, meaning that the same boundary conditions are used for each year of the simulation.

The spatial distribution of the LAI from LPJ-GUESS – shown in Fig. 5 – indicates an elevated LAI of more than 6 m² m^{−2} in the tropical rainforests of the Amazon, Congo and Southeast Asia. The LPJ-GUESS output is also compared with (1) the LAI prescribed for ONEMIS, (2) the LAI prescribed for MEGAN and (3) an external climatology dataset of the global monthly mean LAI averaged over the period from August 1981 to August 2015 (Mao and Yan, 2019). This product uses remotely sensed satellite data from the Moderate Resolution Imaging Spectroradiometer (MODIS) and the Advanced Very High Resolution Radiometer (AVHRR) instruments. Note that each emission scheme utilises its own climatological LAI, and an exchange

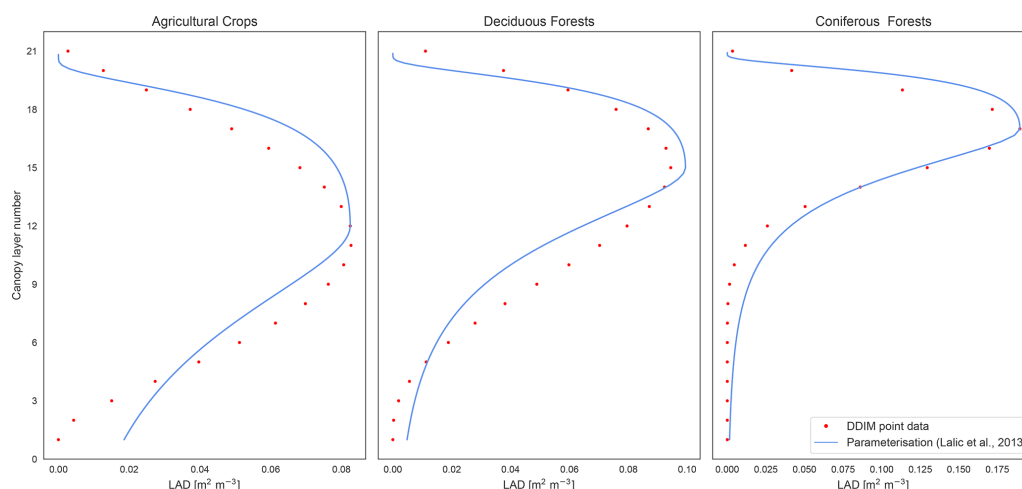


Figure 4. LAD distribution for a 21-layer canopy using DDIM point data versus the continuous distribution from the employed parameterisation for the three vegetation classes.

of this dataset results in substantially modified emission fluxes. The difference plots indicate that the climatological LAI used in ONEMIS is generally higher across the globe, while the LAI climatology used in MEGAN is generally lower, especially over the tropics. The external climatological dataset prescribes lower LAI compared to the LAI from LPJ-GUESS. Panel (e) in Fig. 5 shows the global monthly mean LAI from all datasets. LPJ-GUESS exhibits a difference of $0.73 \text{ m}^2 \text{ m}^{-2}$ between the lowest and highest month, while the variability is $1.40 \text{ m}^2 \text{ m}^{-2}$ for the ONEMIS dataset, $0.68 \text{ m}^2 \text{ m}^{-2}$ for the MEGAN dataset and $0.53 \text{ m}^2 \text{ m}^{-2}$ for the external LAI dataset. This means that the LAI variability from LPJ-GUESS is comparable to the LAI climatology used in MEGAN and the external LAI dataset; however, it differs significantly from the variability in the climatological LAI used in ONEMIS. The foliar density is not presented here, but, by definition, it is a function of the LAI (Eq. 1) and hence shows a similar spatial and temporal distribution. Even though LPJ-GUESS provides potential natural vegetation rather than all present vegetation, a good agreement with climatological LAI is found, particularly the LAI climatology used in MEGAN. Consequently, it is concluded that the current LPJ-GUESS vegetation representation is adequate for estimating BVOC fluxes in ONEMIS and MEGAN but that incorporating land use into the LPJ-GUESS-EMAC configuration will improve current (and future) representations of vegetation and the resulting BVOC estimates.

Figure 6 displays the LAD distribution derived from LPJ-GUESS and the climatology. The climatological LAD (derived from the DDIM model) has been used in the original set-up in ONEMIS to calculate BVOC emission estimates, whereas the LAD from LPJ-GUESS is derived from the parameterisation discussed in Sect. 2.3.3. In LPJ-GUESS, a to-

tal canopy height of 25 m was assumed, and thus, the four LAD layers are classified as follows: bottom canopy layer (LAD 4) represents the LAD between 0–6.25 m, LAD 3 is from 6.25 to 12.50 m, LAD 2 is from 12.50 to 18.75 m, and the top canopy layer (LAD 1) is from 18.75 to 25 m. The values of the four LAD layers (i.e. total canopy) add up to one. The four panels on the right of Fig. 6 show the geographic distribution of LAD from climatology (DDIM model). Even though the LAD canopy distribution from climatology data is oversimplified, it indicates higher densities in the leaf area in the uppermost layers of the canopy over the forested regions found in the tropics and boreal forests in the Northern Hemisphere. The panels on the left show the LAD geographic distribution from LPJ-GUESS at T63. With this approach, a better-resolved geographic distribution for the LAD at the different canopy layers is simulated. Increased LAD in the bottom layer (i.e. all vegetation below 6.25 m) highlights grasslands and desert areas. The layers above show how the LAD changes in the upper sections of the canopy, with plant biomass higher than 20 m mostly found in tropical forest areas. While the climatological LAD remains constant, the LAD from LPJ-GUESS changes from month to month. On average, climatological LAD 1 = 0.21, LAD 2 = 0.32, LAD 3 = 0.40, and LAD 4 = 0.08. LAD 1 from LPJ-GUESS stays constant at around 0.02, and LAD 2 varies between 0.12 (January) and 0.18 (July). LAD 3 and LAD 4 vary a bit but remain close to 0.44 and 0.50, respectively, with no apparent seasonal trends.

Figure 7 shows the fractional coverage required as input for MEGAN. The 12 PFTs from LPJ-GUESS are classified into the four vegetation classes, namely broadleaf trees, needleleaf trees, grass and shrub, and are compared to the corresponding climatology vegetation fraction. The climato-

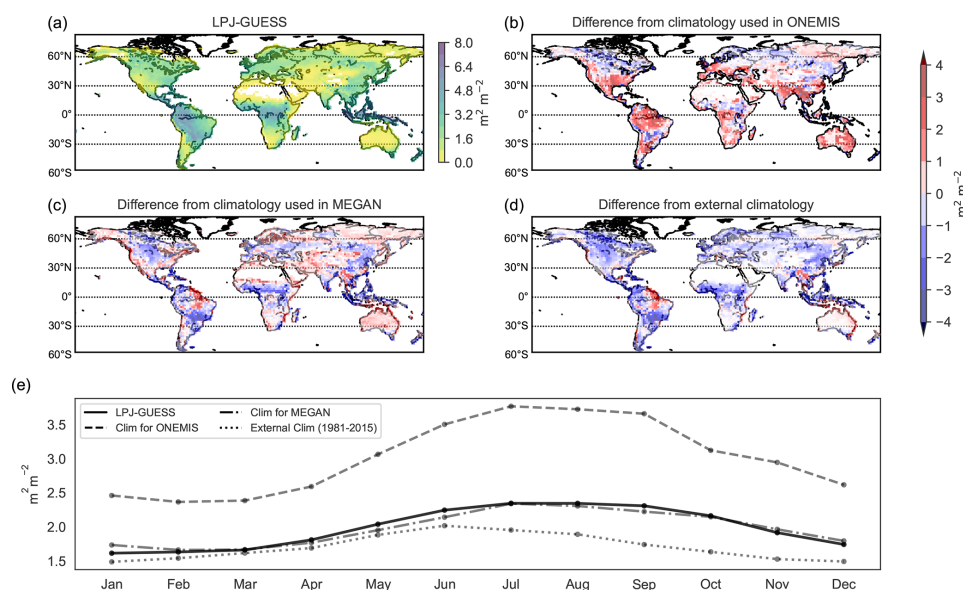


Figure 5. Geographic distribution of the LAI from LPJ-GUESS (a) and difference plots (LAI from LPJ-GUESS minus climatological LAI) from climatology used in the standard configuration in ONEMIS and MEGAN (panels b and c). An additional LAI climatology from 1981 to 2015 is also included (d). Panel (e) compares the global monthly averages of all datasets.

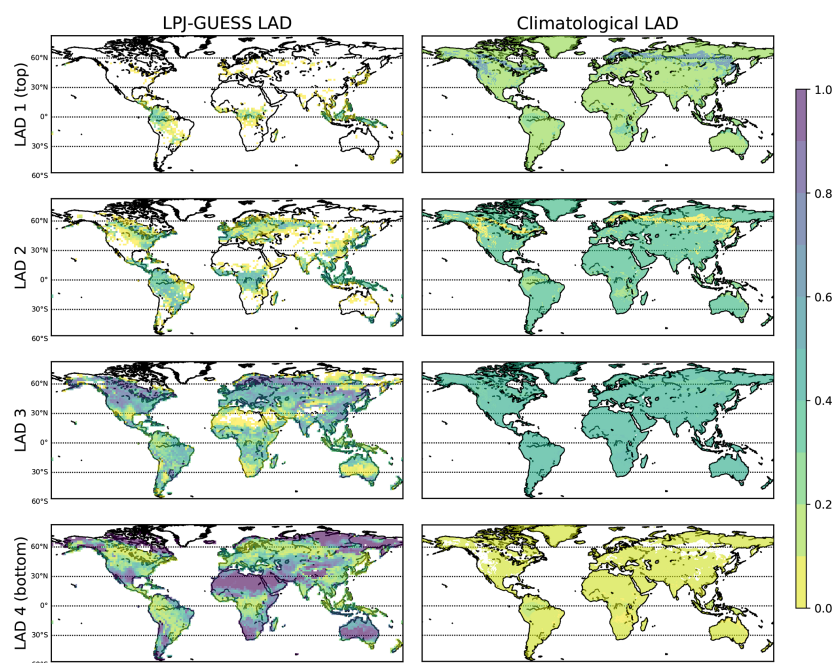


Figure 6. LAD fraction at four canopy layers from LPJ-GUESS (left panels) and the DDIM model (right panels). While the climatological LAD is oversimplified and homogeneous, LPJ-GUESS provides a higher-resolved LAD distribution at the four canopy layers.

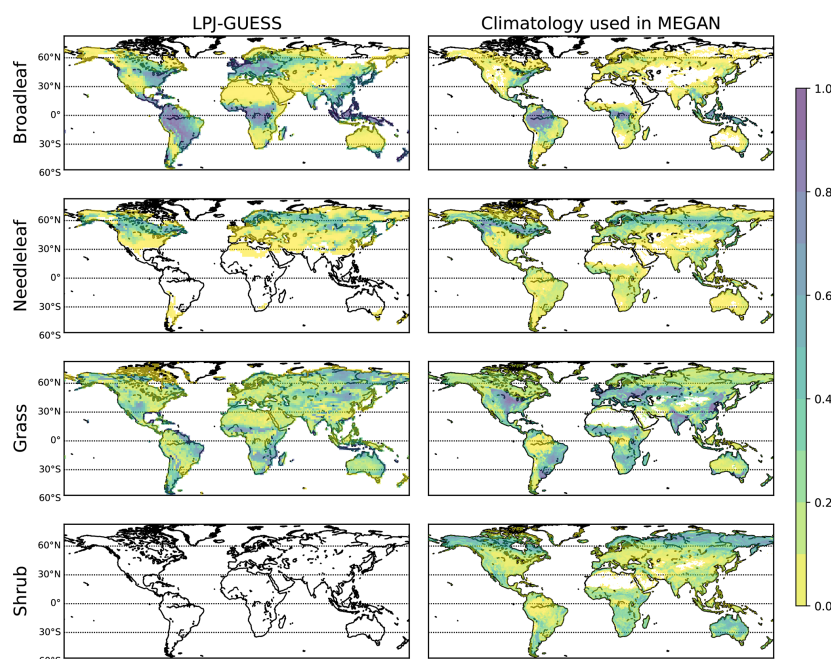


Figure 7. Fractional coverage for different vegetation classes used as inputs in MEGAN. The fractional coverage from LPJ-GUESS is displayed in the left panels, while the climatological fractional coverage is displayed in the right panels. Shrub PFTs are currently missing in the applied LPJ-GUESS global PFT set.

logical fractional coverage remains constant throughout the whole simulation, while the fractional coverage from LPJ-GUESS updates from year to year. The global averages of climatological fractional coverage for broadleaf trees, needleleaf trees, grass and shrubs are 0.13, 0.11, 0.23 and 0.14, respectively. LPJ-GUESS provides fractional coverage of 0.35, 0.11, 0.23 and 0 for broadleaf trees, needleleaf trees, grass and shrubs, respectively. Note that shrubs are not included in the currently applied LPJ-GUESS global PFT set; consequently, they are not considered in the applied simulation set-up. Studies by Forrest et al. (2015) did not use explicit shrub PFTs as well, and they have been explicitly included only in more recent studies (e.g. Allen et al., 2020). Even though this leads to less competition among some PFTs in certain regions, this is a limitation of the current study. However, including the new shrub PFTs is planned for future studies. Also, even though temperate needleleaf trees exist in LPJ-GUESS, they are very uncompetitive and thus not well represented in mixed forests.

3.2 Global isoprene and monoterpene emissions

3.2.1 Isoprene emissions

Figure 8 presents global isoprene emissions from ONEMIS and MEGAN with dynamic vegetation states from LPJ-GUESS, and offline climatological vegetation inputs, at spa-

tial resolution T63. The simulations have been conducted with fixed prescribed CO_2 of 348 ppmv (in both the radiation and vegetation schemes). Apart from the exchange of the vegetation, input parameters for the BVOC modules, the simulation set-up, model code and parameter settings are identical in all simulations. All calculations are climatological, i.e. with identical boundary conditions for all years, and the results presented here are from 10 ensemble years.

Panels a and b in Fig. 8 show the geographic distribution of isoprene emission rates (in $\text{mg m}^{-2} \text{d}^{-1}$) using LPJ-GUESS vegetation inputs. Strong isoprene emission fluxes can be seen over South America, central Africa, Southeast Asia and northern Australia, mostly corresponding to high vegetation densities in tropical rainforests. MEGAN emissions are significantly higher, with up to $90 \text{ mg m}^{-2} \text{d}^{-1}$ over the Amazon rainforest. Such differences in emissions between ONEMIS and MEGAN result from different canopy processes employed by the BVOC modules (see Sect. 2.1). Emission values shown in Fig. 8a and b are from the same simulation, meaning that input parameters (temperature, LAI, etc.) in ONEMIS and MEGAN are identical. Figure 8c and d compare our new emissions using LPJ-GUESS vegetation with emissions using climatological inputs. For both ONEMIS and MEGAN, emissions with dynamic vegetation are lower over tropical areas but higher over Australia compared to emissions using climatology. With climatological vegeta-

tion inputs, ONEMIS exhibits low but significant emissions over deserted regions, particularly the Sahara desert in North Africa, resulting from the poor representation of the LAD distribution in the original model set-up. This is not the case when using dynamic vegetation inputs.

Figure 8e and f depict the temporal profile of global monthly emission totals. In order to capture the true seasonal cycle, values in the Southern Hemisphere were shifted by 6 months before adding fluxes from both hemispheres. Figure 8g and h show the interannual variability in isoprene annual global totals. Over the 10 ensemble years, isoprene emissions appear to peak in the boreal summer months and decrease substantially in the boreal winter months. MEGAN includes a leaf age factor, which accounts for reduced emissions for young and old leaves based on observed LAI change. This explains the slight decrease in MEGAN emissions from April to May to June.

Over the 10-year simulation period considered, the global annual total isoprene fluxes from ONEMIS were found to be 546 Tg yr^{-1} (standard deviation (SD) = 8 Tg yr^{-1}) with dynamic vegetation and 558 Tg yr^{-1} (SD = 7 Tg yr^{-1}) with climatological inputs. MEGAN estimated 657 Tg yr^{-1} (SD = 11 Tg yr^{-1}) with dynamic vegetation and 631 Tg yr^{-1} (SD = 11 Tg yr^{-1}) with climatological vegetation inputs. The higher standard deviation when using LPJ-GUESS inputs indicates higher interannual variability in isoprene fluxes. While the year-to-year variability with climatological inputs is only determined by surface temperature and shortwave radiation (see Fig. 2), the interannual variability with LPJ-GUESS inputs is also influenced by interannual changes in vegetation states estimated in LPJ-GUESS. Jöckel et al. (2016) reported isoprene annual emissions of 488–624 Tg using ONEMIS, while other studies estimated fluxes of 642 Tg yr^{-1} (Shim et al., 2005) using 73 prescribed vegetation types, 571 Tg yr^{-1} (Guenther et al., 2006) using inventories and Olson ecoregion land covers, 467 Tg yr^{-1} (Arneth et al., 2007a) using 10 PFTs from LPJ-GUESS and, more recently, 594 Tg yr^{-1} using 16 PFTs (Sindelarova et al., 2014). It is important to note that no scaling factors were applied in our simulations, even though global annual totals from models are often scaled in atmospheric chemistry studies. For example, Pozzer et al. (2022) estimated 464 Tg of isoprene in the year 2010 using a MEGAN-EMAC set-up but employed a global scaling factor of 0.6. This means that the original emissions are similar to our MEGAN fluxes with climatological inputs. In general, our LPJ-GUESS-driven BVOC emissions in EMAC agree with past modelling estimates with similar spatial and temporal patterns.

3.2.2 Monoterpene emissions

Results for monoterpene emissions are shown in Fig. 9. Panels a and b present the spatial distribution of monoterpene emission rates (in $\text{mg m}^{-2} \text{ d}^{-1}$) from ONEMIS and MEGAN, while Fig. 9c and d show the difference in the

emission fluxes using climatological vegetation inputs. Elevated emission rates are also found over tropical rainforest regions, with ONEMIS prescribing much higher emission rates compared to MEGAN. The difference plots indicate that monoterpene emissions from ONEMIS are significantly higher with climatological inputs, except for some areas over southern Brazil and central Africa. Similarly, MEGAN generally prescribes higher emissions with climatological vegetation inputs compared to LPJ-GUESS inputs. ONEMIS emissions over deserted regions in north Africa and central Australia appear to be better resolved with dynamic vegetation, since ONEMIS with climatology inputs still attributes significant emission rates over such regions where vegetation is absent.

Monoterpene emission fluxes in ONEMIS only depend on the foliar density profile (i.e. $\text{DM} \times \text{LAD}$) and surface temperature. The high dependence of monoterpene emissions on foliar density explains both the lower seasonal variability and the lower yearly fluxes compared to emissions with climatological vegetation inputs. The lower magnitudes in monoterpene fluxes from MEGAN with dynamic vegetation result from the lack of representation of shrubs and needle-leaf tree PFTs in LPJ-GUESS, where such species are known to be strong emitters of monoterpenes. The seasonal variation, however, is satisfying, and the fact that the fractional coverages computed in LPJ-GUESS are dynamic (updating on a yearly time step) makes this set-up suitable for studying long-term variations in emissions. Annual totals from ONEMIS were found to be 102 Tg yr^{-1} (SD = 1 Tg yr^{-1}) with dynamic vegetation inputs and 175 Tg yr^{-1} (SD = 2 Tg yr^{-1}) with climatological inputs. MEGAN prescribes 54 Tg yr^{-1} (SD = 0.7 Tg yr^{-1}) and 76 Tg yr^{-1} (SD = 0.9 Tg yr^{-1}) with dynamic and climatological inputs, respectively. Guenther et al. (2012) gives a global annual monoterpene emission of 157 Tg, while Sindelarova et al. (2014) reported annual total emissions of monoterpenes ranging between 89 and 102 Tg yr^{-1} over a 30-year simulation period. Arneth et al. (2007a) reported 36 Tg yr^{-1} .

3.2.3 BVOC emissions from LPJ-GUESS

This section presents isoprene and monoterpene emission fluxes from the semi-processed-based module in LPJ-GUESS for comparison with the empirically based emissions from ONEMIS and MEGAN in the coupled model system. The LPJ-GUESS routine runs entirely within the LPJ-GUESS framework, meaning that emission values are provided on a daily basis. In this study, the LPJ-GUESS BVOC routine uses DTR computed in EMAC instead of climatological DTR (see Sect. 2.3). Panels a and b in Fig. 10 show the spatial distribution of isoprene and monoterpene emission rates from the LPJ-GUESS BVOC emissions routine. Figure 10c and d show the monthly total emissions from LPJ-GUESS and emissions from ONEMIS and MEGAN for comparison.

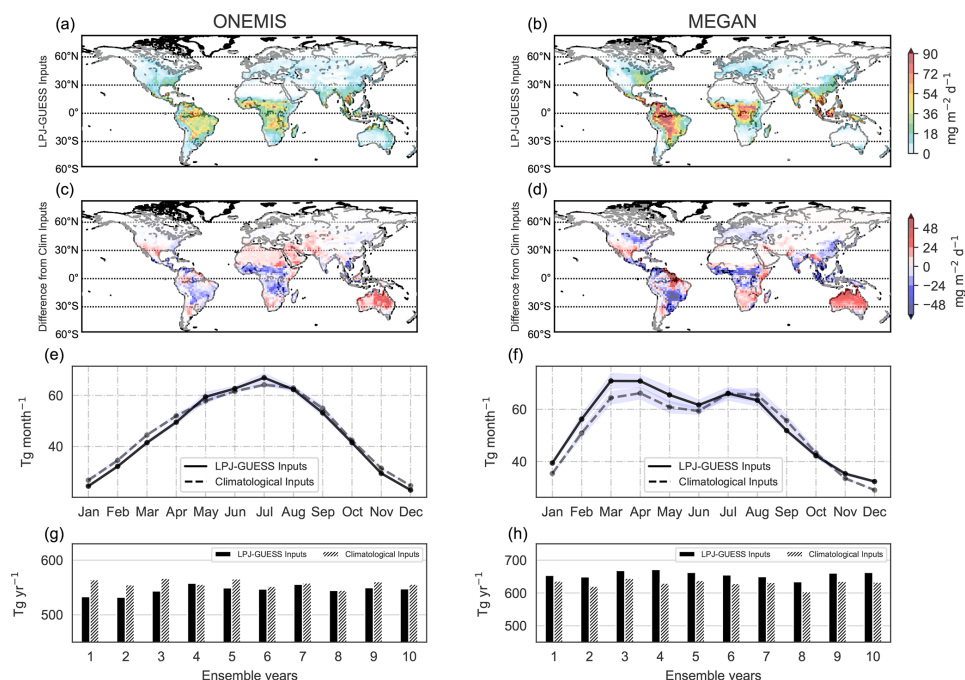


Figure 8. Spatial distribution of monthly isoprene emissions ($\text{mg m}^{-2} \text{d}^{-1}$) averaged over 10 ensemble years with LPJ-GUESS vegetation inputs from ONEMIS and MEGAN (panels **a** and **b**). Panels **(c)** and **(d)** show the difference in the emission flux compared to emissions from ONEMIS and GUESS with climatological vegetation inputs. The temporal profile of isoprene global totals in teragrams per month are depicted in panels **(e)** and **(f)**, and global annual totals (Tg yr^{-1}) for 10 ensemble years are shown in panels **(g)** and **(h)**.

Monthly isoprene emissions from LPJ-GUESS range from 48.3 Tg in December to 88.2 Tg in July, whereas monthly monoterpene emissions range from 1.8 Tg in January to 7.3 Tg in July. Figure 10e and f show yearly isoprene and monoterpene totals from LPJ-GUESS, ONEMIS and MEGAN for 10 ensemble years. The mean isoprene annual emission flux is 750 Tg yr^{-1} ($\text{SD} = 17 \text{ Tg yr}^{-1}$), while for monoterpenes it is 50 Tg yr^{-1} ($\text{SD} = 1 \text{ Tg yr}^{-1}$). The process-based isoprene emissions from LPJ-GUESS are marginally higher compared to the empirically based emissions from ONEMIS and MEGAN. For monoterpenes, emissions from LPJ-GUESS are similar to MEGAN emissions, while ONEMIS emissions are twice as much.

Currently, both empirical (e.g. Guenther et al., 2006) and process-based models (e.g. Niinemets et al., 1999; Bäck et al., 2005; Arneth et al., 2007b; Schurgers et al., 2009) are widely used to model and assess global BVOC emission estimates. Process-based models consider the emission inhibition from carbon and energy availability, allowing for some stress effects to be taken into consideration. In general, empirically based models suggest increased BVOC emissions in future climate scenarios resulting from temperature sensitivity and enhanced vegetation activity in a CO_2 -rich atmosphere (Sanderson et al., 2003; Naik et al., 2004; Lathiere et al., 2005). In contrast, process-based models indicate

that CO_2 inhibition of the leaf–isoprene metabolism can be large enough to offset increases in emissions (Arneth et al., 2007b; Heald et al., 2009). More recent laboratory studies provide evidence that certain plant species emit more isoprene in high CO_2 environments (e.g. Sun et al., 2013), indicating that there are still some gaps in the understanding of biochemical regulation of BVOC leaf emissions incorporated in such models. Grote and Niinemets (2007) compares the two model categories and argues that, in non-stressful conditions, empirical and process-based emission models predict BVOC emission dependencies on light and temperature in a similar way. A modelling study also showed that, regardless of whether CO_2 inhibitory effects are considered or not, temperature is the most important driver for increased isoprene emissions (Cao et al., 2021). In this study, it is shown that, even though process-based emissions from LPJ-GUESS might differ (in magnitude) from empirically based emissions in EMAC, the monthly distributions in emissions are similar (Fig. 10c and d). Even though emissions from ONEMIS and MEGAN in EMAC are empirically based, emissions in the new coupled configuration are now sensitive to vegetation changes, which is a substantial improvement over emissions with prescribed vegetation information in the previous model configuration.

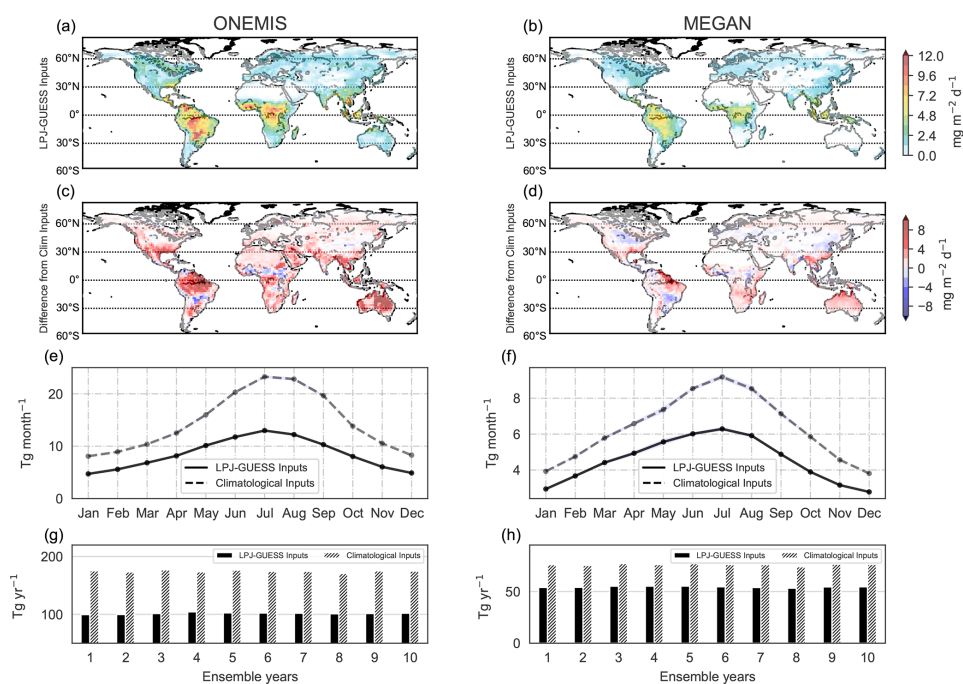


Figure 9. Same as Fig. 8 but for monoterpenes.

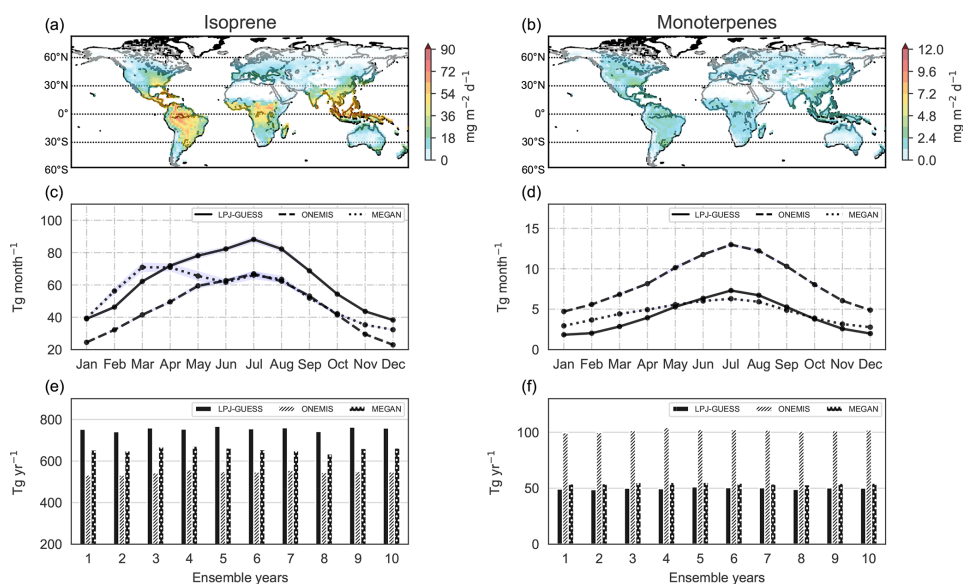


Figure 10. Spatial distribution of isoprene and monoterpene emissions from LPJ-GUESS at a spatial resolution of T63 (panels a and b). Isoprene and monoterpene mean monthly totals in teragrams per month from LPJ-GUESS, ONEMIS and MEGAN (panels c and d) and annual totals in teragrams per year for 10 ensemble years (panels e and f).

Table 1. Prescribed CO₂ in the radiation and vegetation schemes for different studies.

Study	CO ₂ in radiation scheme	CO ₂ in vegetation scheme
Ref	348 ppmv	348 ppmv
Bio × 2	348 ppmv	696 ppmv
Atm × 2	696 ppmv	348 ppmv
Both × 2	696 ppmv	696 ppmv

3.3 Emission sensitivity to double CO₂ scenarios

In this section, the variability in global isoprene and monoterpene emission estimates in doubling CO₂ scenarios is investigated. In particular, the CO₂ fertilisation and temperature effects are evaluated by prescribing different CO₂ values in the radiation and vegetation schemes in the coupled model set-up, as described in Sect. 2.4. The scope here is not to develop realistic future scenarios but rather to assess the model's sensitivity to atmospheric and vegetation CO₂ – hence the use of doubling scenarios. Four experiments were conducted to explore the impact of doubling CO₂ scenarios (both in the radiation and vegetation scheme) on isoprene and monoterpene emission rates. Ref is the reference simulation, with 348 ppmv [CO₂] in both schemes. Table 1 lists the CO₂ levels prescribed for each study. The analyses shown here are based on 10 years of data from 50-year simulations with constant boundary conditions and a 500-year offline spin-up phase.

3.3.1 Surface temperature in 2 × CO₂ in the radiation scheme

The temperature sensitivity of BVOC emissions is studied by doubling the prescribed CO₂ value in the radiation scheme. This results in a consistent global increase in the surface temperature of up to 4 °C in conjunction with the prescribed enhanced SST and sea ice coverage (see Appendix A).

3.3.2 Vegetation response to 2 × CO₂ scenarios

The LAI can be used as a marker for vegetation activity. Figure 11 displays LAI estimates from LPJ-GUESS for the reference study, Ref, and also shows how LAI values in the other studies compare to it. Bio × 2 indicates consistently increased vegetation activity when doubling the prescribed CO₂ in the vegetation scheme. This is the CO₂ fertilisation effect. The LAI in Atm × 2 decreases as a result of warmer temperatures and higher losses of soil moisture (reported elsewhere; e.g. Dermody et al., 2007; Sun et al., 2015). In Both × 2, with 696 ppmv [CO₂] in both the vegetation and radiation scheme, an overall rise in LAI compared to the Ref simulation is found, except for some regions in North America, western Brazil and southern Europe. In these areas, the water stress from higher surface temperatures results in an

overall decline in the vegetation, e.g. grass species take over forested areas, partly decreasing the LAI in the region. To this end, this model set-up can be used to analyse changes in BVOC emissions due to shifts in vegetation patterns in future climate scenarios.

3.3.3 Global BVOC emissions

Figure 12 shows the geographic distribution of isoprene emission rates (in mg m⁻² d⁻¹) averaged over 10 ensemble years for Ref and difference plots for Bio × 2, Atm × 2, and Both × 2 using climatological and dynamic vegetation inputs using ONEMIS and MEGAN. The bottom panels (Fig. 12q, r, s, and t) show the averaged emission rates for distinct latitude bands (0–30° S, 0–30° N, 30–60° N) for all studies during the same period, from left to right. It is found that in Bio × 2, isoprene emissions increased only when using dynamic vegetation inputs in both ONEMIS and MEGAN (Fig. 12b and j) are applied. In contrast to the prescribed climatological vegetation data, the LPJ-GUESS coupled set-up is sensitive to increased CO₂, which subsequently leads to higher emissions. Note that, in this scenario, ONEMIS attributes lower emission values over the tropics (Fig. 12b). Isoprene emissions are more dependent on light than on leaf area, so increased foliage may limit isoprene emissions in closed canopies such as in dense tropical rain forests (Guenther et al., 2006). This is not the case for open canopies, where increased foliage drastically enhances isoprene emissions. This effect is not well captured by MEGAN (Fig. 12j), given that here the PCEEA algorithm is employed whereas the canopy environment model only considers above-canopy radiation and is not sensitive to sun and shaded leaves at each canopy depth. In Atm × 2, temperature effects on isoprene emissions are found, while in Both × 2 we see how the combined effects of CO₂ fertilisation and temperature become obvious. The bar plots reveal that, in places where most of the global emissions occur, i.e. between 0 and 30° S, the emission was the highest in Both × 2, with an average emission rate of 27.3 mg m⁻² d⁻¹ (ONEMIS) and 44.3 mg m⁻² d⁻¹ (MEGAN). Given the empirical nature of ONEMIS and MEGAN, both set-ups give a consistent increase in isoprene fluxes in elevated temperatures (Atm × 2). However, with LPJ-GUESS inputs, differences in the emission fluxes are found that also result from vegetation dynamics, e.g. a decrease in fluxes from lower vegetation activity caused by water stresses. This highlights the advantage of having BVOC fluxes derived from dynamic vegetation states rather than prescribed boundary conditions.

In Fig. 13, similar behaviour in the monoterpene emission estimates is depicted for all studies. Monoterpene emission rates increase in Bio × 2 only for scenarios with dynamic vegetation due to CO₂ fertilisation. A worldwide increase in monoterpene fluxes in Atm × 2 is detected with higher surface temperatures using climatological inputs. However, with dynamic vegetation inputs, a less substantial rise, and a drop

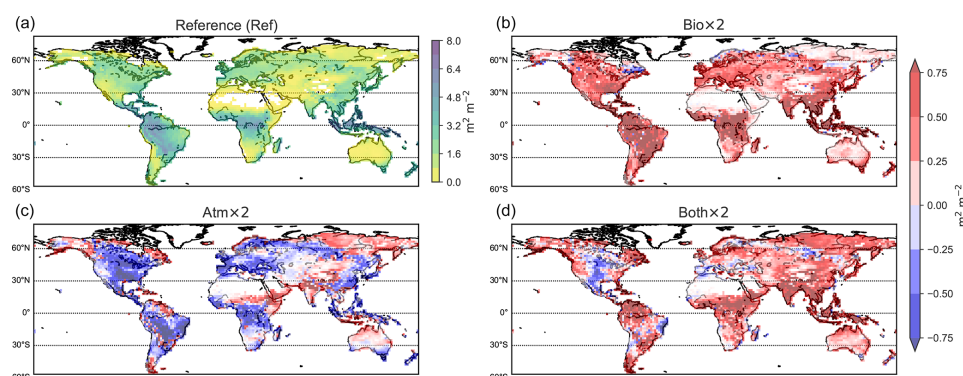


Figure 11. Geographic distribution of the LAI for the reference study (Ref) and difference plots. $\text{Bio} \times 2 - \text{Ref}$ (b), $\text{Atm} \times 2 - \text{Ref}$ (c) and $\text{Both} \times 2 - \text{Ref}$ (d) are shown.

in fluxes in certain regions, is determined. In $\text{Both} \times 2$, an increase in monoterpene emission rates is found as a result of the combined influence of temperature and CO_2 fertilisation.

4 Conclusions

The development of ESMs allows for a far more detailed analysis of a fully coupled and dynamic system addressing many complex biosphere–atmosphere interactions governed by BVOC emissions and thus shedding more light on the significance of such processes on global climate change and air quality. In this work, further development has been made towards producing a new atmospheric-chemistry-enabled ESM by coupling an atmospheric-chemistry-enabled atmosphere–ocean general circulation model (AOGCM) with a DGVM. This work also explores, for the first time, partial bi-directional interactions between the two modelling systems via BVOC emissions, building on recent technical developments (Forrest et al., 2020) that enabled one-way coupled simulations (in which climate information generated by EMAC is used to force LPJ-GUESS, but no land surface information is relayed back to EMAC). The updated model version described in this work allows computations of isoprene and monoterpene emissions based on dynamic vegetation states running on the EMAC time step. This is a substantial improvement compared to the earlier set-up in which BVOC emissions were only based on offline vegetation information and not coupled with dynamic vegetation states. The BVOC module in LPJ-GUESS (used in this study only to compare our new emissions in EMAC; Fig. 10) uses dynamic vegetation information but only provides daily average emissions, and these emissions are not yet integrated into EMAC.

Results show that the LAI and subsequent foliar density estimations from LPJ-GUESS are comparable to climatological datasets used as boundary layer conditions in the MEGAN-EMAC standalone configuration and an external

LAI climatology from 1981 to 2015. The LAI employed in the original ONEMIS-EMAC set-up, on the other hand, differs significantly in terms of magnitude and monthly variability. Such differences in the LAI inputs led to lower isoprene and monoterpene emissions using the coupled model set-up compared to the standalone configuration. The representation of the LAD distribution from the new parameterisation employed in LPJ-GUESS also gives a more realistic LAD profile compared to the oversimplified datasets used by the standard ONEMIS set-up. Given that our coupling only involves vegetative information going into ONEMIS and MEGAN in EMAC, our climate biases are considered to be consistent with those discussed in Forrest et al. (2020).

The new MEGAN-LPJ-GUESS configuration yielded satisfactory isoprene fluxes as well, with a monthly distribution comparable to MEGAN emissions with climatological inputs. Monoterpene emissions from the MEGAN-LPJ-GUESS set-up also give meaningful monthly variability with lower magnitudes compared to emissions with climatological inputs, given that the new set-up lacks emission contributions from shrubs and needleleaf tree PFTs in mixed forests. Future studies may include region-specific PFT groups (including shrub PFTs); however, for this study, the global PFT set is kept unchanged. Global isoprene emission estimates from the coupled model configuration, using both ONEMIS and MEGAN, give realistic global variability, corresponding to emissions with prescribed vegetation boundary conditions. The emission magnitudes are also comparable to modelled fluxes found in the literature and can be adjusted accordingly with global scaling factors for specific atmospheric chemistry studies. The new vegetation-sensitive emissions from ONEMIS and MEGAN agree with previous approaches with respect to the spatial and temporal patterns but provide more consistency and higher temporal resolution as required for interactive atmospheric chemistry simulations.

This study finds that (1) differences in BVOC emissions with implemented dynamic vegetation result almost entirely

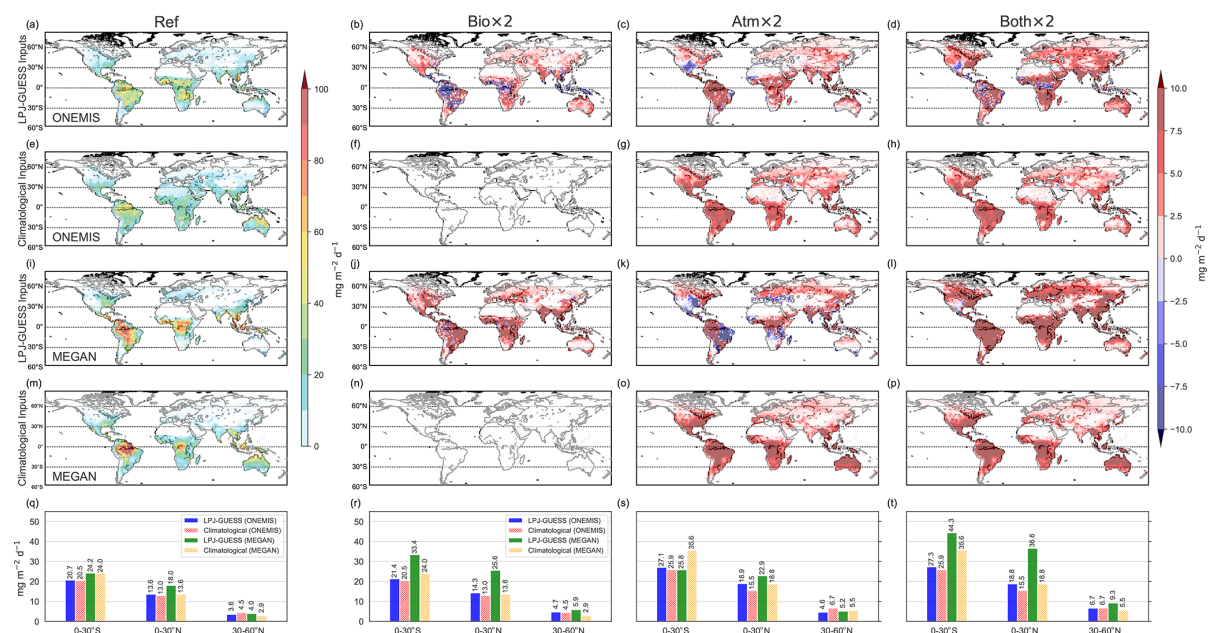


Figure 12. Global isoprene emission estimates from ONEMIS and MEGAN with LPJ-GUESS vegetation inputs and climatological inputs for the reference study (Ref) (a, e, i, m) and difference plots for Bio $\times$ 2 (b, f, j, n), Atm $\times$ 2 (c, g, k, o) and Both $\times$ 2 (d, h, l, p). The panels at the bottom (q, r, s, t) display the emission flux averages (in $\text{mg m}^{-2} \text{d}^{-1}$) over the latitude bands 0–30° S, 0–30° N, 30–60° N.

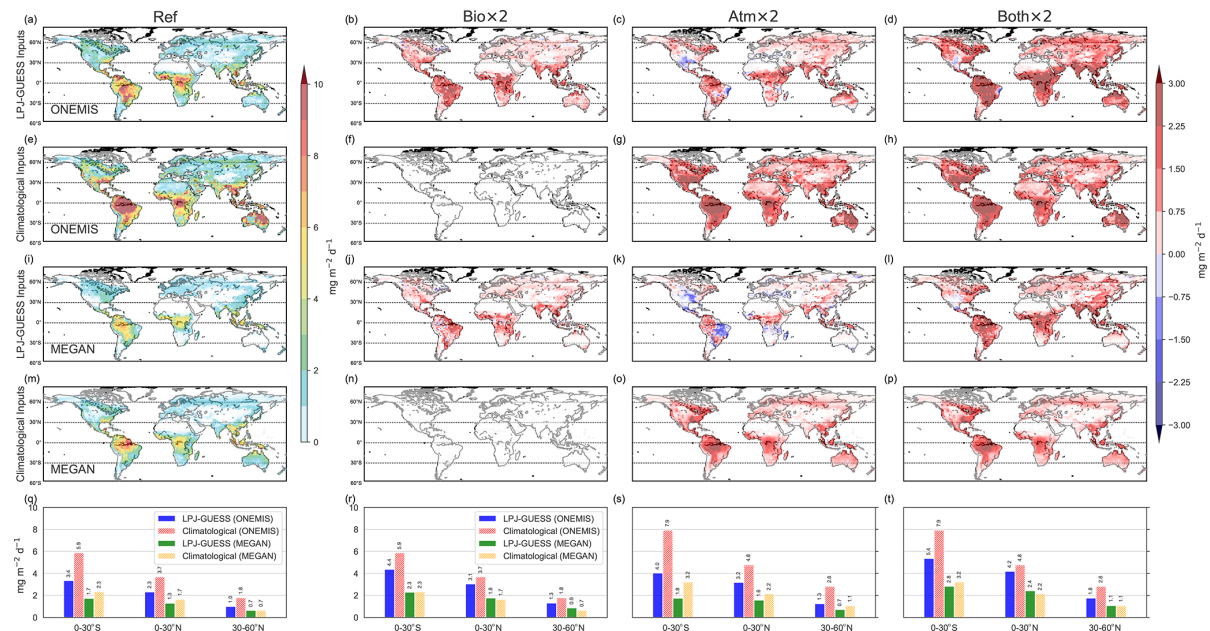


Figure 13. Same as Fig. 12 but for monoterpenes.

from changes in the input LAI, and the (2) dynamic vegetation increase the interannual variability in BVOC emissions by introducing new variability from dynamic vegetation states. These findings are comparable to those of Levis et al. (2003), where dynamic vegetation was implemented in the Community Climate System Model (CCSM).

CO₂ sensitivity studies also suggest that both isoprene and monoterpene emissions rise with warmer climatic scenarios ($2 \times \text{CO}_2$ in the radiation scheme); however, only the coupled configuration showed reduced emissions in some locations. The lower emissions result from the vegetation response to water stresses in a warmer climate. The new coupled model also allows for sensitivity studies for CO₂ fertilisation effects and indicates an increase in both isoprene and monoterpene emission rates in $2 \times \text{CO}_2$ scenarios in the vegetation scheme due to enhanced vegetation activity in a CO₂-rich atmosphere. This work provides evidence that the improved ESM, featuring dynamic vegetation, gives realistic BVOC emission estimates on a global scale, based on dynamic vegetation states, enabling further research into atmosphere–biosphere interactions and feedbacks with this model configuration.

Appendix A: Surface temperature at doubling CO₂ scenarios

Figure A1 illustrates temperatures (°C) at the lowermost vertical level in EMAC at spatial resolution T63 with prescribed CO₂ of 348 ppmv in the top panel and 696 ppmv in the middle panel. A plot comparing the reference simulations to the 2 × CO₂ simulations is given in the lower panel of Fig. A1.

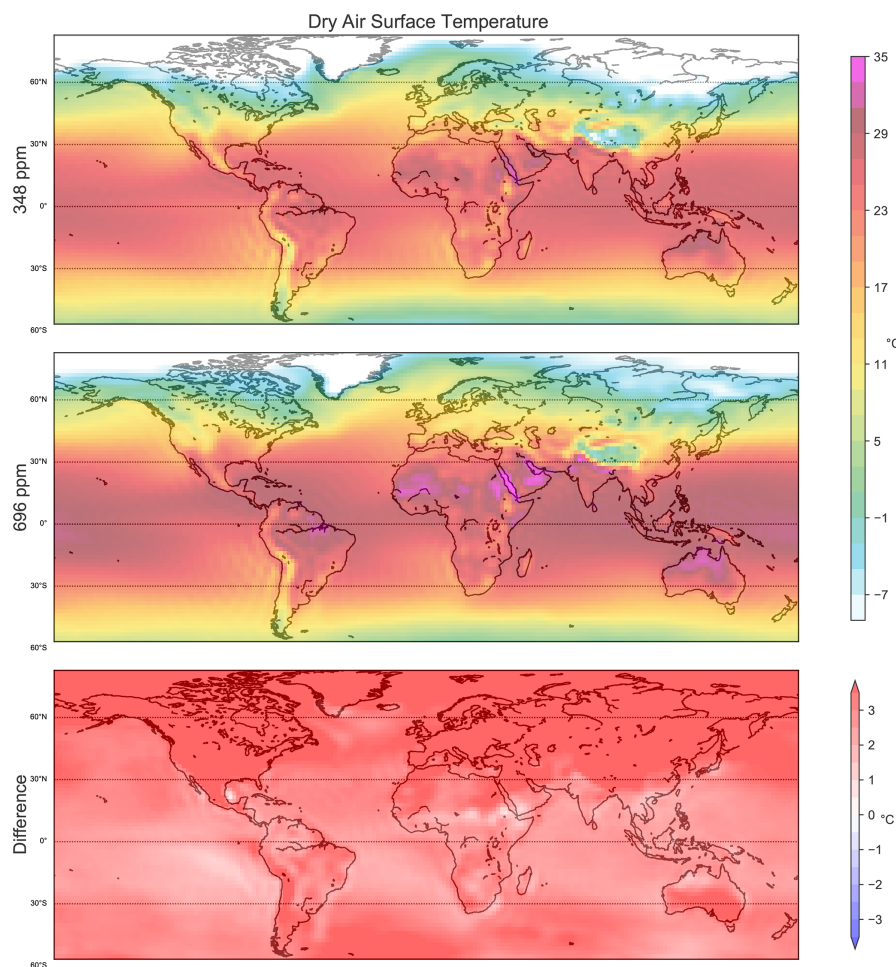


Figure A1. Surface temperature (°C) at spatial resolutions T63. In the radiation scheme, the first model configuration utilises a CO₂ volume mixing ratio of 348 ppm (parts per million), whereas the second set-up uses 696 ppm.

Code availability. The Modular Earth Submodel System (MESSy) is continuously developed and applied by a consortium of institutions. MESSy is licensed to all affiliates of institutions that are members of the MESSy Consortium, as is access to the source code. Institutions can become a member of the MESSy Consortium by signing the MESSy Memorandum of Understanding. More information can be found on the MESSy Consortium website (<http://www.messy-interface.org>, last access: 14 January 2023). LPJ-GUESS is used and developed globally; however, development is overseen at Lund University's Department of Physical Geography and Ecosystem Science in Sweden. Model codes can be made available to collaborators on entering into a collaboration agreement with the acceptance of certain conditions. Given that both MESSy and LPJ-GUESS are restricted, the code used here is archived with a restricted-access DOI (<https://doi.org/10.5281/zenodo.6772205>; Vella et al., 2022). The code will not be made publicly available, and sharing will be made possible only by the approval of the authors. The code described in this work has already been incorporated into the official development branch of the EMAC modelling system and will therefore be part of all future released versions.

Data availability. The datasets from this work will be made available on request to the corresponding author.

Author contributions. RV and HT performed the model coupling. RV performed the simulations and analysis. MF and JL contributed to the overall model development and interpretation of the results. RV wrote the article, with significant inputs from all co-authors.

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

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3.3 Impact of El Niño-Southern Oscillation on global BVOC emissions

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Changes in biogenic volatile organic compound emissions in response to the El Niño–Southern Oscillation

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Abstract. Emissions of biogenic volatile organic compounds (BVOCs) from the terrestrial biosphere play a significant role in major atmospheric processes. BVOCs are highly reactive compounds that influence the atmosphere's oxidation capacity and also serve as precursors for the formation of aerosols that influence global radiation budgets. Emissions depend on the response of vegetation to atmospheric conditions (primarily temperature and light), as well as other stresses, e.g. from droughts and herbivory. The El Niño–Southern Oscillation (ENSO) is a naturally occurring cycle arising from anomalies in the sea surface temperature (SST) in the tropical Pacific. ENSO perturbs the natural seasonality of weather systems on both global and regional scales and is considered the most significant driver of climate variability. Several studies have evaluated the sensitivity of BVOC fluxes during ENSO events using historical transient simulations. While this approach employs realistic scenarios, it is difficult to assess the impact of ENSO alone given the multiple types of climate forcing, e.g. from anthropogenic emissions of CO₂ and aerosol. In this study, a global atmospheric chemistry–climate model with enabled interactive vegetation was used to conduct two sets of simulations: (1) isolated ENSO event simulations, in which a single ENSO event is used to perturb otherwise baseline conditions, and (2) sustained ENSO simulations, in which the same ENSO conditions are reproduced for an extended period of time. From the isolated ENSO events, we present global and regional BVOC emission changes resulting from the immediate response of vegetation to atmospheric states. More focus is given to the sus-

tained ENSO simulations, which have the benefit of reducing the internal variability for more robust statistics when linking atmospheric and vegetation variables with BVOC flux anomalies. Additionally, these simulations explore long-term changes in the biosphere with potential shifts in vegetation in this possible climate mode, accounting for the prospect of increased intensity and frequency of ENSO with climate change. Our results show that strong El Niño events increase global isoprene emission fluxes by 2.9 % and that one single ENSO event perturbs the Earth system so markedly that BVOC emission fluxes do not return to baseline emissions within several years after the event. We show that persistent ENSO conditions shift the vegetation to a new quasi-equilibrium state, leading to an amplification of BVOC emission changes with up to a 19 % increase in isoprene fluxes over the Amazon. We provide evidence that BVOC-induced changes in plant phenology, such as the leaf area index (LAI), have a significant influence on BVOC emissions in the sustained ENSO climate mode.

1 Introduction

The terrestrial biosphere is a major source of natural volatile organic compounds (VOCs), such as isoprene and monoterpenes, which account for approximately 90 % of all VOC emissions to the atmosphere (Guenther et al., 1995). Isoprene and monoterpenes are thought to work against stress-induced reactive oxygen species or help plants in coping with

abiotic stress by changing membrane properties (Sharkey and Loreto, 1993; Vickers et al., 2009; Karl et al., 2010; Sharkey and Monson, 2017), while they can also be induced by other chemical, physical, or biological processes, such as herbivory (Laothawornkitkul et al., 2008) and signalling between organisms (Zuo et al., 2019). Biogenic volatile organic compounds (BVOCs) are highly reactive and short-lived (minutes to hours), as they quickly interact with tropospheric oxidant gases upon emission, exerting a significant influence on the atmosphere's oxidation capacity (Atkinson, 2000; Atkinson and Arey, 2003). The dominant reaction mechanisms of BVOCs are OH and O₃ oxidation, which have significant implications for secondary organic aerosol (SOA) formation and, in turn, for cloud formation and climate (Pöschl et al., 2010; Ehn et al., 2014; Palm et al., 2018). OH oxidation of BVOCs also has implications for greenhouse gas and pollutant concentrations (e.g. methane and CO; Arneth et al., 2010; Peñuelas and Staudt, 2010). BVOC emissions exhibit year-to-year variations attributed to the vegetation's sensitivity to climatic conditions. Our prior investigations have revealed that isoprene fluxes, spanning a decade, display a standard deviation of 8 Tg yr⁻¹ (Vella et al., 2023a).

The El Niño–Southern Oscillation (ENSO) is a periodic oscillation (occurring every 2 to 7 years) between anomalously warm (El Niño) and cold (La Niña) sea surface temperatures (SSTs) in the tropical Pacific (McPhaden et al., 2006). Because of the strong interactions between atmospheric and oceanic circulations, such anomalies in tropical Pacific SST have a significant impact on atmospheric processes. ENSO therefore exerts a marked influence on weather systems and global climate patterns (McPhaden et al., 2006). During ENSO events, the Walker circulation convective centres rearrange, inducing precipitation anomalies in the tropics as well as influencing monsoon systems via the Hadley circulation over the Pacific, Indian, and Atlantic oceans. Teleconnections with midlatitude westerlies can also result in consistent anomaly patterns in the extratropics (Dai and Wigley, 2000). To this end, tropical regions are often much warmer and drier than average during El Niño episodes (Gong and Wang, 1999; Dai and Wigley, 2000), but some regions tend to be cooler and wetter, e.g. western North America (Ropelewski and Halpert, 1986) and eastern Asia (Wu et al., 2003).

Several studies have explored the sensitivity of the terrestrial biosphere to different ENSO phases (e.g. Ahlström et al., 2015; Chang et al., 2017; Bastos et al., 2018; Wang et al., 2018; Teckentrup et al., 2021). The primary factors driving changes in vegetation are closely linked to the dominant meteorological drivers of net primary productivity (NPP) in different regions. Specifically, the primary drivers in the wet tropics and dry tropics and temperate regions are, respectively, radiation and moisture, while temperature is the primary driver in the western temperate and boreal regions (Nemani et al., 2003). The terrestrial biosphere often acts as

a carbon source during El Niño events, while carbon uptake increases during La Niña events, particularly in semi-arid ecosystems (Ahlström et al., 2015). Nevertheless, the complex relationships between ENSO-induced climatic variability and terrestrial ecosystem productivity, particularly the extent, amplitude, and underlying processes, remain poorly understood (Gonsamo et al., 2016; Wang et al., 2018; Zhu et al., 2017). Zhang et al. (2019) linked ENSO seasonality with global gross primary production (GPP) and found peak correlations occurring between global yearly GPP and ENSO conditions in August and October of the preceding year. Drought and warming in the Amazon Basin during the 2015/2016 El Niño event led to higher stress-related BVOC emissions, which, when combined with greater turbulent transport above the canopy, resulted in higher OH reactivity compared to non-El Niño years (Pfannerstill et al., 2018). Isoprene emissions based on satellite formaldehyde (HCHO) measurements also show interannual differences tied to temperature shifts and climate features such as El Niño (Wells et al., 2020). Modelling studies have suggested that BVOC emissions are generally higher during El Niño years and lower during La Niña years, with ENSO having a significant impact in both the tropics and the higher latitudes (Naik et al., 2004; Lathière et al., 2006; Müller et al., 2008). However, uncertainties persist due to the influence of climate change on ENSO variability.

Despite the numerous studies focusing on the effects of ENSO perturbation on the biosphere, only a limited number of studies have specifically investigated the sensitivity of BVOC emissions. Given that in situ observations of BVOC emission fluxes are scarce, many studies have relied on satellite-based remote sensing or modelling approaches. The modelling studies, in particular, are limited in number and only link BVOC emission trends during ENSO events without exploring in detail the underlying mechanisms driving such changes or separating the effects of ENSO from the transient emission changes in greenhouse gases and shorter-lived pollutants and their precursors. BVOC emissions are influenced by the overall response of the biosphere to atmospheric conditions; however, they are also strongly influenced by temperature and surface radiation, making them likely to be affected by the temperature and cloudiness variations associated with ENSO.

In this study, sea surface temperatures (SSTs) and sea ice cover (SIC) associated with ENSO years were used to construct different ENSO scenarios: very strong and moderate El Niño/La Niña scenarios as well as a baseline scenario. An Earth system model (ESM) with interactive vegetation representations was used to investigate ENSO-induced changes in meteorology, vegetation, and BVOC emissions on global and regional scales. We present two sets of simulations: (1) isolated ENSO event simulations, which employ a single ENSO event in otherwise baseline conditions, and (2) sustained ENSO simulations, which use the same ENSO conditions over a 30-year period. Both sets of simulations

are used to evaluate the various effects of ENSO. The isolated ENSO event simulations are used to study the temporal evolution of BVOC anomalies following a single ENSO event and to estimate global and regional changes resulting from the immediate response of the biosphere to atmospheric state anomalies. The most recent Intergovernmental Panel on Climate Change (IPCC) assessment based on CMIP6 simulations states that ENSO will continue to be the primary mode of interannual variability in a warmer climate, with a high degree of certainty (Lee et al., 2021). However, some studies have suggested the possibility of increased frequency of extreme ENSO events under greenhouse warming (e.g. Cai et al., 2015, 2021). Therefore, we use the sustained ENSO simulations to explore the upper range of impacts of ENSO, as well as long-term changes in the biosphere and the resulting BVOC emission fluxes in such a climate mode. We have examined various parameters, including surface temperature, surface radiation, aridity, net primary productivity (NPP), and leaf area index (LAI), to investigate the changes brought by ENSO. While changes in these parameters on a global scale can indicate overarching trends, anomalies connected to ENSO are often identified at a regional level. Given that the majority of BVOC emissions are emitted in tropical regions, our analysis focuses on seven specific regions within or in close proximity to the tropics: southwestern USA (SWUSA), the Amazon Basin (Amazon), central western Africa (CWAfr), southeastern Africa (SEAfr), India, southeastern Asia (SEAsia), and northern Australia (NAus), and these regions are commonly thought to be hotspots for ENSO-associated climate anomalies. To our knowledge, no study has comprehensively investigated global and regional BVOC emission changes in relation to vegetation and atmospheric state anomalies elicited by ENSO. Therefore, this research aims to shed light on the intricate interactions that drive shifts in BVOC emissions associated with the El Niño–Southern Oscillation.

2 Methods

2.1 Models

2.1.1 The EMAC modelling system

The EMAC (ECHAM/MESSy Atmospheric Chemistry) model is a numerical chemistry and climate modelling system that contains submodels that represent tropospheric and middle-atmospheric processes, as well as their interactions with oceans, land, and anthropogenic activities. It originally combined the ECHAM atmospheric general circulation model (GCM) (Roeckner et al., 2006) with the Modular Earth Submodel System (MESSy) (Jöckel et al., 2005) framework and philosophy, modularising physical processes as well as most of the infrastructure into submodels that can be further developed to improve existing process representa-

tions; new submodels can also be added to represent new or alternative process representations. In recent years, EMAC has been further developed to include a broader representation of atmospheric chemistry by coupling different processes such as representations for aerosols, namely aerosol–radiation and aerosol–cloud interactions (Tost, 2017). In this study, version 2.55 has been used, which is based on the well-documented version used in comprehensive model intercomparison studies (Jöckel et al., 2016).

2.1.2 LPJ-GUESS

The Lund–Potsdam–Jena General Ecosystem Simulator (LPJ-GUESS¹) (Smith et al., 2001, 2014) is a dynamic global vegetation model (DGVM) featuring an individual-based model of vegetation dynamics. These dynamics are simulated as the emergent outcome of growth and competition for light, space, and soil resources among woody plant individuals and a herbaceous understorey in each of a number (50 in this study) of replicate patches representing random samples of each simulated locality or grid cell. The simulated plants are classified into 12 plant functional types (PFTs) discriminated by growth form; phenology; photosynthetic pathway (C₃ or C₄); bioclimatic limits for establishment and survival; and, for woody PFTs, allometry and life history strategy. LPJ-GUESS has previously been implemented in global ESMs (e.g. Weiss et al., 2014; Alessandri et al., 2017) and, more recently, coupled with EMAC (Forrest et al., 2020; Vella et al., 2023a). The LPJ-GUESS version used in this study currently provides information on potential natural vegetation, and it does not incorporate changes in land use. This is a limitation within our current model configuration, as the implemented version of LPJ-GUESS lacks the capability to account for land use changes. This functionality will be included in the next version of LPJ-GUESS.

2.2 EMAC-LPJ-GUESS configuration

Once fully coupled, the EMAC-LPJ-GUESS configuration will be a sophisticated Earth system model (ESM) capable of studying interactions between the land and atmosphere. This includes examining the methane cycle and its lifespan, the atmospheric chemistry of various carbon compounds, the impact of fires and associated feedbacks, future nitrogen deposition rates and scenarios for fertilisation, the effects of ozone on plants, and the role of biogenic volatile organic compounds in aerosol production and their contribution to cloud formation and precipitation patterns. While efforts towards a fully coupled configuration are ongoing, in this work, we use the standard EMAC-LPJ-GUESS coupled configuration, where the vegetation in LPJ-GUESS is entirely determined by the EMAC atmospheric state, soil, N deposition, and

¹The following section is based on the template for a standard copyright-free LPJ-GUESS model description (<https://web.nateko.lu.se/lpj-guess/resources.html>, last access: 3 July 2023).

fluxes (Forrest et al., 2020), but there is no feedback from the vegetation to climate variables (e.g. changes in albedo and roughness length). After each simulation day, EMAC computes the average daily values of 2 m temperature, net downward shortwave radiation, and total precipitation and passes these state variables to LPJ-GUESS. Vegetation information (LAI, foliar density, leaf area density distribution, and PFT fractional coverage) from LPJ-GUESS is then fed back to EMAC for the calculation of BVOC emission fluxes using EMAC's BVOC submodules (Vella et al., 2023a), namely ONEMIS (Kerkweg et al., 2006) and MEGAN (Guenther et al., 2006). Both ONEMIS and MEGAN are based on the Guenther algorithms (Guenther et al., 1993, 1995), where the BVOC emission flux (F) is calculated as a function of the foliar density and its vertical distribution (D , kg dry matter m^{-2}), ecosystem-specific emission factors (ϵ), and a non-dimensional activity factor (γ) that accounts for the photo-synthetically active radiation (PAR) and temperature:

$$F = [D][\epsilon][\gamma]. \quad (1)$$

In this work, we evaluate fluxes from ONEMIS, which is the standard and most established emission module in EMAC. Emissions are calculated at four distinct canopy layers, which are defined by the leaf area density (LAD) and the leaf area index (LAI). The attenuation of the PAR is determined for each level by considering the direct visible radiation and the zenith angle. Using the proportions of sunlit leaves and the overall biomass, emissions from both sunlit and shaded leaves within the canopy are estimated. Further technical details for canopy processes employed in ONEMIS can be found in Ganzeveld et al. (2002). While validating pure BVOC fluxes from models using observations remains challenging, this setup was evaluated and demonstrated to be adept in capturing global BVOC estimates and responses when compared to other modelling studies (Vella et al., 2023a). As described in Eq. (1), BVOC emission calculations in this setup are governed by vegetation states (D) from LPJ-GUESS that are largely based on temperature, radiation, and soil moisture. Furthermore, the instantaneous surface radiation and temperature levels (γ) have a large impact on the emission rates. On the basis of such model parameterisations, we explore the impact on BVOC emission anomalies by evaluating changes in the surface temperature and radiation, aridity index (AI), NPP, and LAI.

2.3 Experimental design

A 30-year (1980–2009) SST and SIC dataset from the Atmospheric Model Intercomparison Project Phase II (AMIP II) was used to evaluate the Oceanic Niño Index (ONI) and classify the strength of different ENSO events (<https://pcmdi.llnl.gov/mips/amip/amip2/>, last access: 3 July 2023). The 3-month mean SST anomalies in the Niño 3.4 region (5°N – 5°S , 120°W – 170°W), based on this 30-year base period, are shown in the top panel of Fig. 1. Events are defined when the

anomaly is greater than or equal to 0.5°C for five consecutive overlapping 3-month periods ($+0.5^{\circ}$ for El Niño events and -0.5° for La Niña events). Furthermore, events are categorised as weak (0.5 to 0.9 SST anomaly), moderate (1.0 to 1.4), strong (1.5 to 1.9), or very strong (≥ 2.0) when the threshold is reached or exceeded for at least three consecutive overlapping 3-month periods. Even though not officially published, this ONI threshold classification has been used by the National Oceanic and Atmospheric Administration (NOAA)² and also in several research articles (e.g. Jimenez et al., 2021; Abish and Mohanakumar, 2013).

Global SST and SIC are used as forcing data to construct five scenarios: (1) base conditions (based on the 1980–2009 average), (2) very strong El Niño (based on 1997/1998), (3) moderate El Niño (based on 2002/2003), (4) very strong La Niña (based on 1988/1989), and (5) moderate La Niña (based on 1995/1996). The event typically spans March to June of the following year. The lower panel of Fig. 1 shows SST starts to deviate from baseline conditions in March/April of the first event year and continues until April/May/June of the following year (event year 2). For all simulations, the CO_2 concentration was kept fixed at 348 ppmv, representing the year 2000. This study looks at atmospheric, vegetational, and BVOC emission changes in sustained ENSO scenarios with continuous ENSO forcing, as well as the temporal evolution of BVOC emissions following an isolated event under otherwise baseline conditions. To do so, two sets of global simulations, hereinafter referred to as *sustained* and *isolated* simulations, are performed. Both sets of simulations run at a horizontal resolution of T63 (approximately $1.9^{\circ} \times 1.9^{\circ}$) and have a 500-year offline spin-up phase. In this work, we focus especially on seven regions, defined in the Supplement and also presented graphically in Figs. 4, 5, and 7. The regions considered are hotspots for ENSO (apart from NAus) and places with generally high BVOC emissions in the tropics (except SWUSA) (Bastos et al., 2013; Vella et al., 2023a; Sindelarova et al., 2014). Additionally, we used the BVOC anomaly distribution maps (Fig. 7) to establish the exact dimensions of the bounding box for regions with relatively consistent BVOC anomalies. Throughout our analysis, we applied an ocean mask to focus solely on anomalies occurring over land.

2.3.1 Isolated ENSO event simulations

This simulation setup is designed to trigger a single ENSO event (spread over 2 years) in otherwise baseline conditions. The evolution of the SST temporal simulations is depicted in the lower panel of Fig. 1. The simulations used base conditions for the whole simulation time except for the 2 event years, where a perturbation in the SST and SIC is introduced

²<https://www.climate.gov/news-features/blogs/enso/united-states-el-nino-impacts-0>, last access: 3 July 2023.

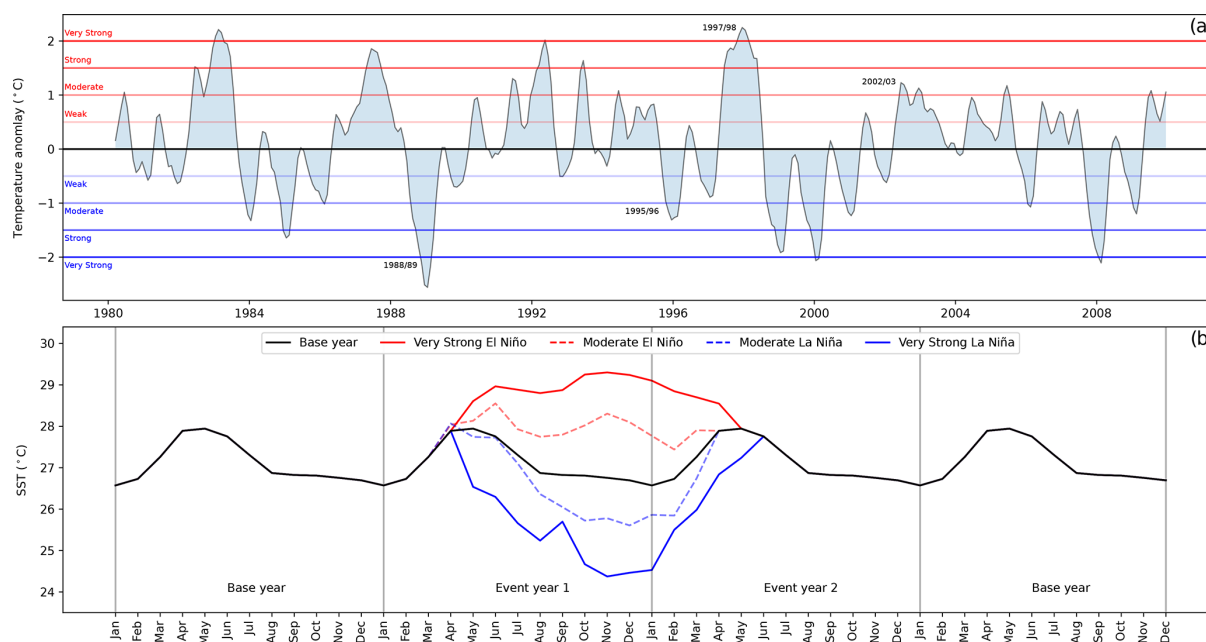


Figure 1. (a) SST anomalies in the Niño 3.4 region from 1980 to 2009. The horizontal lines indicate the anomaly strength (El Niño in red and La Niña in blue) with incremental thresholds every 0.5° . The dates of significant ENSO episodes considered in this study are highlighted. (b) A total of 4 consecutive years of SST in the Niño 3.4 region. The 2 years in the middle indicate four ENSO scenarios as well as the base scenario. The base year (i.e. the 30-year average SST over El Niño 3.4) is shown in black, while the ENSO events are shown in red (El Niño) and blue (La Niña).

according to the specific ENSO event considered. The ENSO anomaly perturbation is employed in the 31st and 32nd years of the 50-year simulation, which means that the simulation runs with base SST/SIC from the 1st to the 30th year, and then from the 33rd to the 49th year.

The events considered with this setup are a very strong La Niña event (based on SST and SIC from May to December 1988 and January to April 1989) and a very strong El Niño event (based on SST and SIC from May to December 1997 and January to March 1998). With this setup, temporal variations in BVOC emissions during very strong ENSO events and in subsequent years can be assessed. We also investigate correlations between the BVOC flux anomalies and the temperature, radiation, aridity index (AI), net primary production (NPP), and leaf area index (LAI) anomalies during the event years and the subsequent 2 years. Here, AI is defined as the total precipitation divided by the potential evaporation/sublimation (including evapotranspiration, PET). The standardised anomaly is calculated by dividing the anomalies by the standard deviation of the base scenario. The simulations conducted in this study differ from previous studies, where BVOC emission changes due to ENSO are typically evaluated using satellite data (e.g. Zhang et al., 2019; Wells et al., 2020) or with transient (i.e. historical) simulations (e.g. Naik et al., 2004; Bastos et al., 2018; Teckentrup

et al., 2021). The advantage of conducting isolated ENSO simulations with constant climate boundary conditions is that it allows for the study of the specific impacts of ENSO on the system being simulated. By isolating the ENSO signal and keeping other climatic forcing factors, such as trends from CO_2 , aerosol loading, etc., constant, we can attribute any observed changes in BVOC fluxes solely to the ENSO phenomenon.

2.3.2 Sustained ENSO simulations

Even though the isolated ENSO simulations give insights into changes in BVOC fluxes with respect to the magnitude and evolution over time, it is hard to constrain ENSO-induced changes statistically from a single simulation run given the high internal variability within the system. We therefore further analysed model results from sustained ENSO simulations. These simulations describe ENSO conditions continuously over many years by using the same yearly cycle of SST and SIC data over 30 simulated years. The five scenarios employ the following global SST and SIC data: (1) base simulation – monthly average SST and SIC from 1980 to 2009 (30 years), (2) moderate El Niño scenario – April to December 2002 and January to March 2003, (3) very strong El Niño scenario – May to December 1997 and January to March 1998, (4) moderate La Niña scenario

– April to December 1995 and January to March 1996, and (5) very strong La Niña scenario – May to December 1988 and January to April 1989. The corresponding SSTs used in these simulations can be seen in the lower panel of Fig. 1. The baseline simulation employs SST/SIC spanning January to December, whereas the ENSO simulations adopt distinct 12-month sequences spanning the event years 1 and 2 (see Fig. 1). In these subsequent simulations, there could be a disruption in the sequential order of months given that the ENSO event occurs over 2 years and could start in March, April, or May of the first event year. Nevertheless, the annual ENSO cycle remains consistent given that these 12 monthly SST/SIC data are used perpetually.

Compared to the isolated simulations, the sustained simulations constrain the correlations among BVOC flux emissions, meteorology, and vegetation changes better, and they provide statistical confidence that the characterised perturbations are caused by ENSO rather than other variability attributed to the climate system. However, we emphasise that these simulations express ENSO scenarios where the vegetation comes into quasi-equilibrium with the new climate system. The reported changes in BVOC emissions are therefore exaggerated as they include drifts in the vegetation states resulting from years to decades of plant establishment and mortality. The focus here is to link ENSO with the driving variables for BVOC emissions; however, given the possible increased frequency of more intense ENSO with climate change (Cai et al., 2021, 2015), these simulations provide insights into possible vegetation changes and the subsequent effects on BVOC emission fluxes in these scenarios.

2.4 Principle component analysis

Principal component analysis (PCA) was used on climate variables to assess their correlation with isoprene emissions during El Niño events (Sect. 3.2.3). PCA is a statistical method used to simplify and understand complex datasets by transforming the original variables into a new set of orthogonal (uncorrelated) variables called principal components. These components are linear combinations of the original variables that capture the maximum variance in the data. For each pixel of the model output, we perform PCA to extract the first principal component for each driving variable (temperature, radiation, aridity index, NPP, and LAI). The correlation between the first principal component of each driving variable and isoprene/monoterpene emission fluxes is computed for each pixel. These correlation values are then used to rank the variables' importance in driving isoprene/monoterpene emissions during El Niño events for each pixel.

3 Results

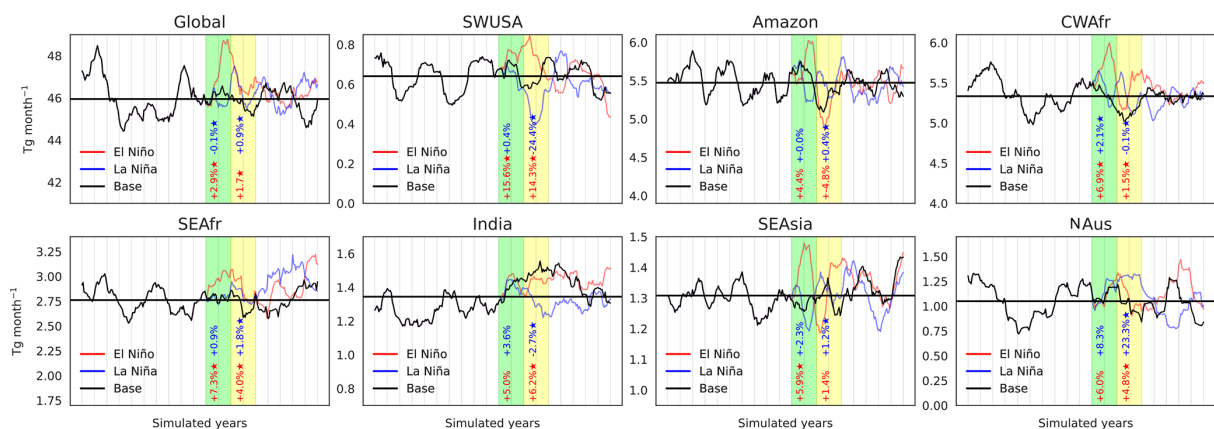
3.1 Isolated ENSO event simulations

This section presents results from the isolated simulations described in Sect. 2.3. Figure 2 shows global 12-month moving averages of monthly isoprene emissions globally and over seven regions (see Table 1 in the Supplement). The event, spanning over 2 years, is marked in green, while the following 2 years are marked in yellow. Isoprene fluxes from El Niño, La Niña, and base conditions are shown in red, blue, and black, respectively. Notice that prior to the event, the isoprene fluxes in all cases are identical; however, following the ENSO perturbation, fluxes diverge. The anomalies in BVOC emission fluxes result from the immediate response of vegetation due to changes in meteorological states elicited by ENSO. Given that the simulations are identical, the changes noted here can be solely attributed to the corresponding SST/SIC anomalies, and the influence of other forcing factors within the Earth system can be excluded.

Figure 2 shows the percentage changes in El Niño (in red) and La Niña (in blue) fluxes compared to the base simulation during the event (in green) and the subsequent 2 years (in yellow). The percentage changes include an asterisk when the difference from the base emissions is statistically significant (99 % confidence with a two-tailed Student's *t* test, i.e. $p < 0.01$) over that time frame. Global isoprene emissions increase by 2.9 % during an El Niño and remain elevated by 1.7 % in the 2 years following the event. During El Niño and the subsequent 2 years, SWUSA experienced a rise of 15.6 % and 14.3 %, respectively, while a decline of 24.4 % is found in SWUSA during the 2 years following La Niña. Other notable emission changes were observed in CWAfr, SEAfr, and SEAsia for the El Niño event. A significant increase of 23.3 % in the 2 years following La Niña was also observed in NAus. Our results suggest that changes in isoprene fluxes due to an ENSO event are highly regional and, on a global scale, higher emissions during and following a strong El Niño event are present. These simulations also show that the perturbation introduced by the ENSO event stretches for a long time following the event, even though the SST and SIC are restored to base conditions. Over the course of 7 years following the event, global isoprene emissions deviated from baseline levels by 0.74 Tg per month during El Niño and 0.37 Tg per month during La Niña on average. However, our findings indicate that the maximum monthly deviation reached 1.01 Tg during El Niño and 1.49 Tg during La Niña within this 7-year period. Monoterpene emissions follow similar tendencies to isoprene fluxes (see Fig. S3 in the Supplement). El Niño results in a global increase of 3.2 % in monoterpene fluxes, while regionally, emissions increase by 18.4 % over SWUSA, 6.9 % over the Amazon, 6 % over CWAfr, 5.1 % over SEAfr, and 2.6 % over SEAsia ($p < 0.01$). La Niña events do not result in statistically significant changes in monoterpene emission changes glob-

Table 1. Simulation details for ENSO experiments in this study.

Simulation name	Simulation type	Details	
Base Very strong El Niño Very strong La Niña	Isolated ENSO simulations	T63 horizontal resolution 500-year offline spin-up phase 40 years long, using the last 11 years for analysis Base global SST and SIC conditions except for the event years On the 31st/32nd year of the simulation	
Base Very strong El Niño Moderate El Niño Very strong La Niña Moderate La Niña		Sustained ENSO simulations	T63 horizontal resolution 500-year offline spin-up phase 50 years long, using the last 30 years for analysis The same conditions applied in every year of the simulation

**Figure 2.** Evolution over time of global and regional isoprene fluxes in response to El Niño (red) and La Niña (blue) events over 2 years (green columns). The black line shows fluxes without the ENSO perturbation, while the horizontal black line illustrates the mean over base conditions throughout all simulations. Percentage changes in El Niño (in red) and La Niña (in blue) fluxes compared to the base simulation during the event (in green) and the subsequent 2 years (in yellow) are included with statistically significant changes ($p < 0.01$) marked with an asterisk. The vertical grey lines correspond to the simulated years. Only 10 of the 30 simulated years before the ENSO event are shown.

ally, but notable changes are seen in the 2 years following the event over SWUSA (-24.2%) and NAus ($+24.0\%$).

Figure 3 shows correlations between the isoprene standardised flux anomalies and the standardised temperature, radiation, aridity index (AI), net primary production (NPP), and leaf area index (LAI) anomalies during the event years and the following 2 years (total of 4 years) for the regions considered during El Niño and La Niña. Table 2 lists the Pearson correlation coefficients (r) from Fig. 3 but also includes r values for the 2-year event period and the following 2 years separately. In our assessment, we classify the Pearson correlation coefficients as follows: 0.00–0.29 as *negligible*, 0.30–0.49 as *weak*, 0.50–0.69 as *moderate*, 0.70–0.89 as *strong*, and ≥ 0.90 as *very strong* for positive correlations and similarly for negative correlations between 0 and -1 . The investigated correlation coefficients between modelled isoprene emission fluxes and various driving variables pro-

vide insights into the relationship between these factors in different regions during El Niño and La Niña events.

We note that the dependencies in isoprene emissions are very region-specific, so much so that correlations on a global scale are generally poor for both El Niño and La Niña events. In the Amazon region, during both El Niño and La Niña events, there are moderate-to-strong positive correlations observed between isoprene flux and temperature (El Niño, 0.89; La Niña, 0.85), as well as surface radiation (El Niño, 0.73; La Niña, 0.87). Additionally, there are strong negative correlations between isoprene emissions and AI (El Niño, -0.77 ; La Niña, -0.75). These findings suggest that higher temperatures and surface radiation tend to increase isoprene emissions in this region, while increased drought conditions limit the emissions to some degree. In CWAfr we see moderate-to-weak correlations between the isoprene flux and temperature (0.51), radiation (0.58), and AI (-0.46) during an El Niño event and only a weak correlation with temperature (0.45)

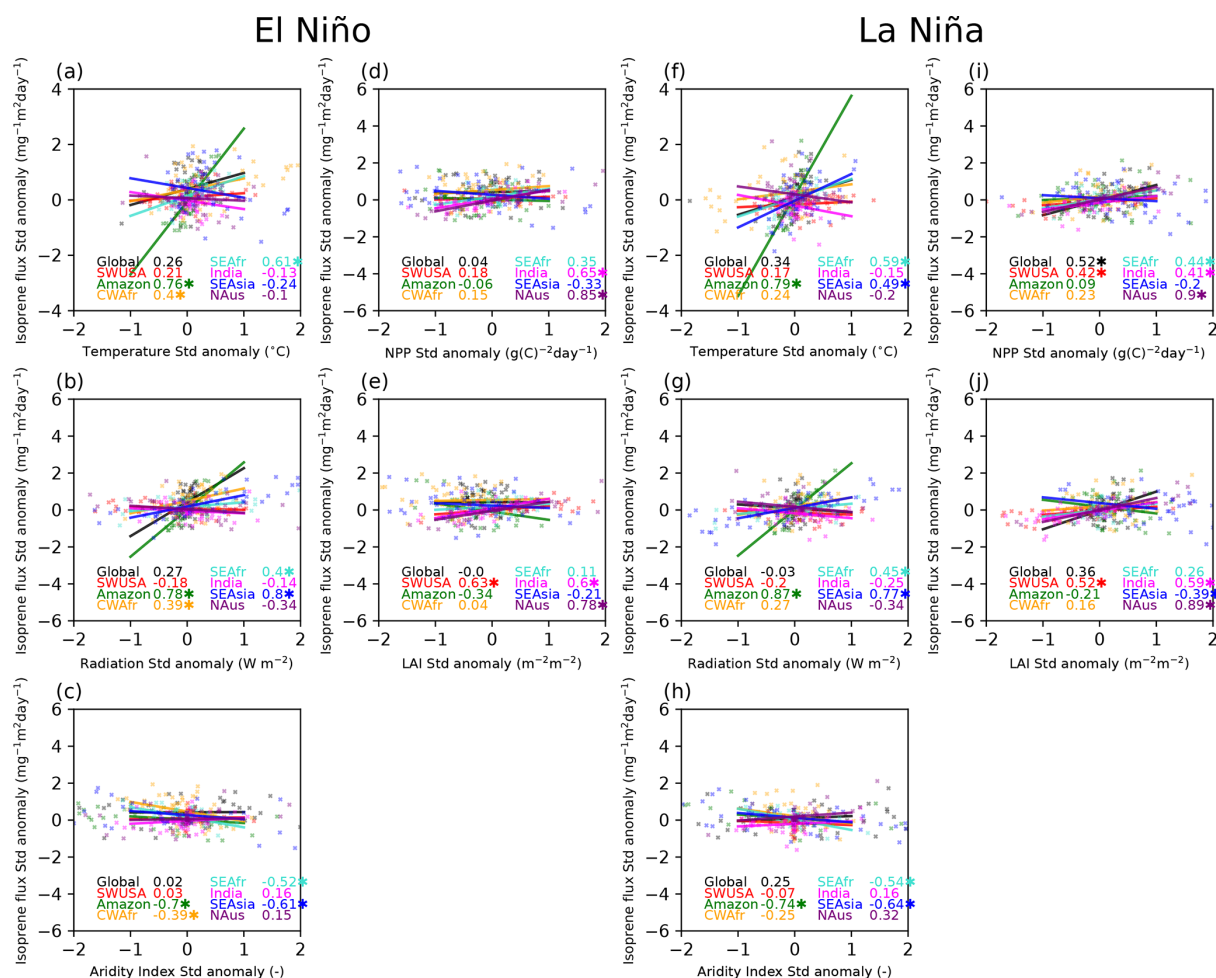


Figure 3. Scatterplots with a linear regression fit including the Pearson correlations (r) between the standardised isoprene flux anomalies and standardised temperature, radiation, aridity index (AI), net primary production (NPP), and leaf area index (LAI) anomalies in different regions for the event years and the following 2 years (4 years in total). Very strong El Niño events are shown in the panels on the left-hand side, while very strong La Niña events are shown in the panels on the right-hand side. Correlations with $p < 0.01$ are marked with an asterisk.

during a La Niña event. In SEAfr we also observe a moderate correlation with temperature (El Niño, 0.61; La Niña, 0.63) and a weak negative correlation with AI (El Niño, -0.45 ; La Niña, -0.40). In SEAsia, we observe a strong positive correlation (0.71) between temperature and isoprene flux during La Niña events. Additionally, there are strong correlations with radiation during both El Niño (0.87) and La Niña (0.84) events. In this region, we also find weak-to-moderate negative correlations between isoprene emissions and the NPP and LAI for both El Niño and La Niña events. In India, the correlation between isoprene flux and temperature is negligible to weak across all time frames, suggesting that temperature alone may not be a strong driver of isoprene emissions in India; however, moderate-to-strong correlations are seen

with AI, NPP, and LAI during La Niña. In SWUSA there are moderately positive correlations between isoprene flux and NPP/LAI during La Niña. Overall, the correlations in the NAus region suggest that surface NPP and LAI play significant roles in driving isoprene emission fluxes.

3.2 Sustained ENSO simulations

The isolated ENSO simulations that have been discussed so far give insights into the temporal evolution of changes in BVOC emission changes from single ENSO events. Here we present results from sustained ENSO conditions based on the last 30 ensemble years from 50-year simulations at a horizontal resolution of T63 with a 500-year offline spin-up phase. These simulations have the advantage of minimising

Table 2. The Pearson correlations (r) between the standardised isoprene flux anomalies and standardised temperature, radiation, aridity index (AI), net primary production (NPP), and leaf area index (LAI) anomalies at different regions for both events as well as the 2 subsequent years (as depicted in Fig. 3), for the ENSO event only (2 years) and for the 2 years following the event. Correlations with $p < 0.01$ are in bold.

	Temperature			Radiation			AI			NPP			LAI		
	Both	Event	+2 years	Both	Event	+2 years	Both	Event	+2 years	Both	Event	+2 years	Both	Event	+2 years
El Niño															
Global	0.26	0.22	0.25	0.27	0.27	0.25	0.02	0.12	−0.03	0.04	−0.39	0.57	0	−0.17	0.18
SWUSA	0.21	0.20	0.35	−0.18	−0.17	−0.34	0.03	0.19	0.04	0.18	0.25	−0.07	0.63	0.5	0.68
Amazon	0.76	0.89	0.53	0.78	0.73	0.85	−0.70	−0.77	−0.56	−0.06	−0.23	0.27	−0.34	−0.68	0.28
CWAfr	0.40	0.51	−0.07	0.39	0.58	0.1	−0.39	−0.46	−0.32	0.15	0	0.43	0.04	−0.02	0.11
SEAfr	0.61	0.61	0.61	0.4	0.3	0.53	−0.52	−0.45	−0.56	0.35	0.28	0.54	0.11	0.23	0.04
India	−0.13	−0.26	−0.04	−0.14	−0.4	0.14	0.16	0.35	0.02	0.65	0.72	0.2	0.60	0.59	0.19
SEAsia	−0.24	−0.2	0.72	0.80	0.87	0.81	−0.61	−0.76	−0.38	−0.33	−0.62	−0.01	−0.21	−0.59	−0.15
NAus	−0.10	0.18	0.23	−0.34	−0.28	−0.36	0.15	0.32	0.09	0.85	0.85	0.87	0.78	0.79	0.81
La Niña															
Global	0.34	0.44	0.27	−0.03	−0.25	0.13	0.25	0.27	0.22	0.52	0.42	0.59	0.36	0.08	0.61
SWUSA	0.17	0.24	0.01	−0.20	−0.09	−0.47	−0.07	−0.09	0.17	0.42	0.26	0.51	0.52	0.45	0.48
Amazon	0.79	0.85	0.72	0.87	0.87	0.87	−0.74	−0.75	−0.72	0.09	−0.15	0.33	−0.21	−0.22	−0.17
CWAfr	0.24	0.45	0.14	0.27	0.3	0.35	−0.25	−0.32	−0.19	0.23	0.15	0.38	0.16	0.2	−0.04
SEAfr	0.59	0.63	0.61	0.45	0.15	0.71	−0.54	−0.40	−0.68	0.44	0.41	0.46	0.26	0.42	0.11
India	−0.15	−0.07	−0.08	−0.25	−0.29	0.01	0.16	0.35	0.02	0.41	0.72	0.2	0.59	0.59	0.19
SEAsia	0.49	0.71	0.62	0.77	0.84	0.72	−0.64	−0.62	−0.67	−0.20	−0.49	−0.01	−0.39	−0.59	−0.15
NAus	−0.20	−0.20	−0.1	−0.34	−0.44	−0.25	0.32	0.58	0.11	0.90	0.93	0.88	0.89	0.90	0.87

the influence of internal variability and thus enhancing potential links among ENSO, meteorology, vegetation, and BVOC emission fluxes.

While the current climatic conditions do not align with this scenario, it has been suggested that under the influence of climate change, both El Niño and La Niña could become more intense and prolonged (Cai et al., 2015, 2021). The main objective of the presented simulations is to statistically analyse the response of the driving variables to ENSO, enabling us to examine potential lasting impacts on the biosphere and BVOC emissions.

3.2.1 Changes in atmospheric drivers during ENSO

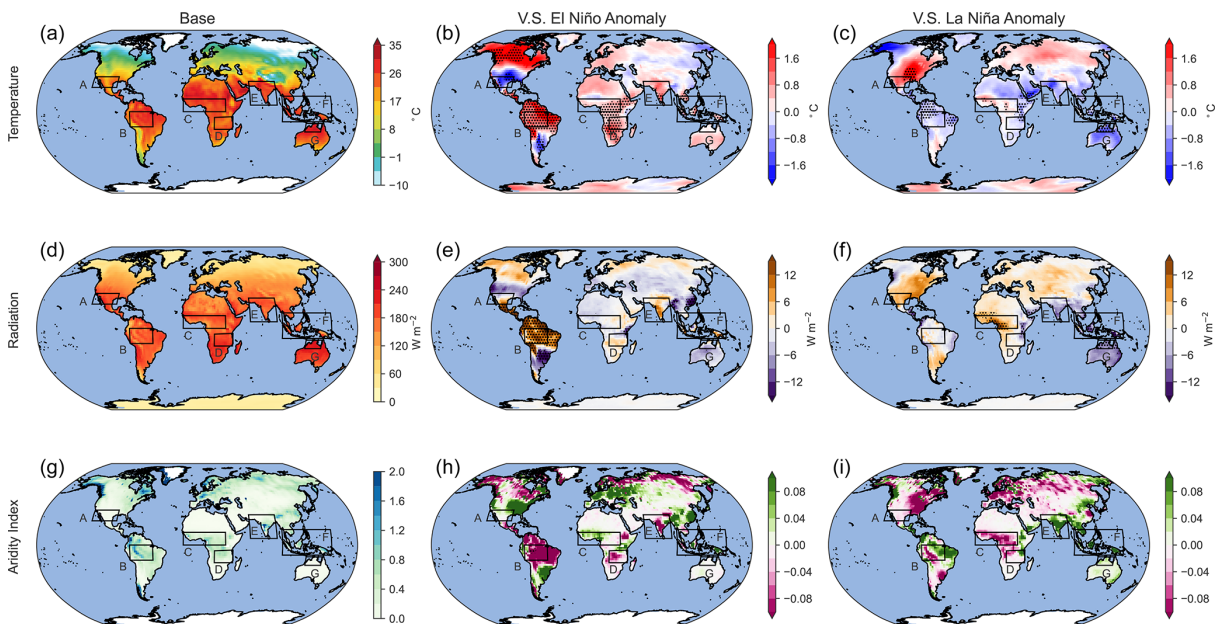
Figure 4 shows global distributions of surface temperature, net solar radiative flux at the surface, and AI averaged over 30 years for the base scenario as well as anomalies from very strong El Niño (very strong El Niño – base) and La Niña (very strong La Niña – base) scenarios. Table 3 provides an overview of the absolute and percentage changes in surface temperature, radiation, and AI over land during the ENSO conditions compared to the base scenario, globally, and the indicated areas of interest with statistically significant anomalies ($p < 0.01$) in bold.

On a global scale, there is a notable increase in surface temperature over land (2.11 %) during El Niño and a decrease (−0.26 %) during La Niña. Global surface radiation anomalies and AI anomalies are below 1 %. SWUSA experiences a decrease in temperature (−3.93 %) during El Niño, coupled with an increase in AI (57.57 %). Conversely, during La Niña, SWUSA encounters a temperature increase (3.74 %) along with an increase in radiation (3.05 %) but a substantial decrease in AI (−47.23 %). The Amazon region

exhibits a statistically significant increase in temperature (5.84 %) and radiation (6.02 %) during El Niño, while the aridity index undergoes a considerable decrease (−22.59 %). For La Niña, the Amazon region experiences a statistically significant decrease in temperature (−1.49 %) and an increase in AI of 5.45 %. In CWAfr, El Niño brings about an increase in temperature (2.35 %), while AI decreases (−13.64 %). During La Niña, CWAfr sees a decrease in temperature (−0.33 %), a decrease in radiation (−2.71 %), and an increase in AI (10.80 %). SEAfr experiences a statistically significant increase in temperature (2.80 %) during El Niño, while La Niña brings a decrease (−1.21 %). Changes in the surface radiation are relatively small for SEAfr, while AI decreases (−4.54 %) and increases (2.47 %) during El Niño and La Niña, respectively. India experiences a temperature increase (2.44 %) during El Niño and a decrease during La Niña (−1.75 %). AI shows contrasting changes, decreasing during El Niño (−11.67 %) and increasing during La Niña (17.22 %). SEAsia exhibits a slight temperature increase (0.62 %), a significant increase in radiation (6.29 %), and a decrease in AI (−20.04 %) during El Niño. Conversely, La Niña brings a slight temperature decrease (−0.42 %), a decrease in radiation (−3.36 %), and an increase in AI (29.35 %). NAus experiences a slight increase (0.36 %) in temperature during El Niño, while temperature (−2.55 %) and radiation (−3.51 %) decrease during La Niña. AI increases by 82.74 % during La Niña. Overall, both El Niño and La Niña substantially modulate key meteorological variables at a regional level without disturbing the global energy and water budgets.

Table 3. Absolute and percentage changes in temperature, radiation, and the aridity index (AI) for El Niño and La Niña scenarios corresponding to Fig. 4. Statistically significant anomalies ($p < 0.01$) are in bold.

Area	El Niño anomaly			La Niña anomaly		
	Temp [% (°C)]	Rad [% (W m ⁻²)]	AI [% (-)]	Temp [% (°C)]	Rad [% (W m ⁻²)]	AI [% (-)]
Global	2.11 (0.25°)	0.49 (0.72)	-0.34 (-0.0008)	-0.26 (-0.03°)	0.04 (0.06)	-0.79 (-0.0019)
SWUSA	-3.93 (-0.81°)	0.02 (0.05)	57.57 (0.027)	3.74 (0.77°)	3.05 (5.96)	-47.23 (-0.0222)
Amazon	5.84 (1.51°)	6.02 (10.61)	-22.59 (-0.123)	-1.49 (-0.39°)	-0.12 (-0.22)	5.45 (0.0297)
CWAfr	2.35 (0.64°)	3.44 (6.85)	-13.64 (-0.0442)	-0.33 (-0.40°)	-2.71 (2.56)	10.80 (0.035)
SE Afr	2.80 (0.65°)	0.03 (0.07)	-4.54 (-0.0126)	-1.21 (-0.28°)	-0.82 (-1.60)	2.47 (0.0069)
India	2.44 (0.62°)	1.45 (2.72)	-11.67 (-0.0187)	-1.75 (-0.45°)	-2.89 (-5.42)	17.22 (0.0276)
SE Asia	0.62 (0.17°)	6.29 (12.91)	-20.04 (-0.1043)	-0.42 (-0.12°)	-3.36 (-6.89)	29.35 (0.1527)
NAus	0.36 (0.11°)	-0.53 (-1.26)	2.55 (0.0007)	-2.55 (-0.76°)	-3.51 (-8.28)	82.74 (0.0236)

**Figure 4.** Global distribution of surface temperature, solar radiation, and AI during the base scenario (a, d, g) averaged over the last 30 years of 50-year simulations. Panels (b), (e), and (h) show El Niño anomalies, and (c), (f), and (i) show La Niña anomalies. Areas with statistically significant anomalies ($p < 0.01$) are hatched.

3.2.2 Vegetation response

The model configuration used in this study links atmospheric states with vegetation dynamics. Given that the vegetation is responsive to temperature, solar radiation, and soil moisture, changes in atmospheric states during an ENSO event also modify the state of the vegetation. Figure 5 shows the spatial distribution of the net primary production (NPP) and leaf area index (LAI) during the base case as well as anomalies in the same parameters during El Niño and La Niña scenarios. Percentage and absolute changes in NPP and LAI are presented in Table 4, showing the contrasting effects of El Niño and La Niña anomalies on NPP and LAI across different re-

gions. On a global scale during El Niño occurrences, a slight rise (0.20 %) in NPP is noted, although this change lacks statistical significance. However, LAI experiences a significant decrease (-3.05 %), suggesting a reduction in vegetation cover. Regionally, SWUSA exhibits a notable increase in both NPP (12.11 %) and LAI (22.79 %) during El Niño, while the Amazon region experiences decreases in both NPP (-3.10 %) and LAI (-6.86 %). In contrast, during La Niña, positive changes are observed, with notable increases in NPP and LAI in NAus.

The biosphere responds on different timescales to atmospheric changes: while NPP responds nearly instantaneously, LAI responds rather slowly (within years to decades), but

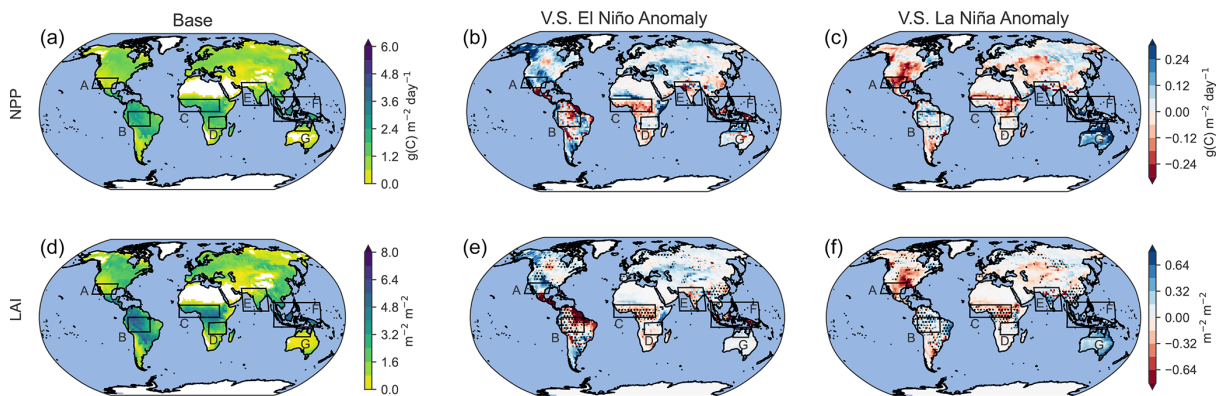


Figure 5. Distribution of NPP and LAI for base conditions (a, d) and anomalies in very strong El Niño (b, e) and very strong La Niña scenarios (c, f) averaged over the last 30 years of 50-year simulations.

Table 4. Absolute and percentage changes in the net primary productivity (NPP) and leaf area index (LAI) for El Niño and La Niña scenarios corresponding to Fig. 5. Statistically significant anomalies ($0 < .01$) are in bold.

Area	El Niño anomaly		La Niña anomaly	
	NPP [% ($\text{g C m}^{-2} \text{yr}^{-1}$)]	LAI [% ($\text{m}^2 \text{m}^{-2}$)]	NPP [% ($\text{g C m}^{-2} \text{yr}^{-1}$)]	LAI [% ($\text{m}^2 \text{m}^{-2}$)]
Global	0.20 (0.28)	−3.05 (−0.06)	1.45 (1.96)	2.54 (0.05)
SWUSA	12.11 (9.63)	22.79 (0.21)	−16.44 (−13.08)	−30.08 (−0.28)
Amazon	−3.10 (−6.35)	−6.86 (−0.29)	2.58 (5.30)	2.46 (0.10)
CWAfr	−1.11 (−1.99)	−13.17 (−0.46)	1.26 (2.28)	4.43 (0.16)
SE Afr	2.07 (3.16)	1.54 (0.04)	1.13 (1.73)	3.32 (0.08)
India	−3.60 (−3.88)	−2.28 (−0.03)	−0.66 (−0.72)	10.08 (0.15)
SE Asia	−3.66 (−7.85)	−23.24 (−1.01)	4.47 (10.28)	16.67 (0.72)
NAus	12.32 (9.66)	13.81 (0.09)	40.60 (31.85)	72.40 (0.49)

changes in PFT composition are often very slow, as they are the result of decades to hundreds of years of plant dispersal, establishment, and mortality (Hickler et al., 2012). Note that in the vegetation model here, dispersal limitations are not included; i.e. if the climate is suitable for a PFT, it can establish immediately. Thus, the model is likely to overestimate range shifts. However, recent observations of increasing drought- and heat-related tree mortality in many areas across the globe (Parmesan et al., 2022) have shown that PFT compositions can also change rapidly if the dominant trees die. With the intensification of ENSO, we could be moving towards a new quasi-equilibrium in the NPP and LAI states where the vegetation state adapts to the new climate. Figure 6 shows changes in the vegetational fractional coverage of the dominant PFTs over SWUSA, the Amazon, SEAsia, and India in the different scenarios of this climate mode. PFT fractional coverage in CEAfr, SE Afr, and NAus exhibited minimal variations and is therefore excluded from Fig. 6.

SWUSA is a semi-arid region dominated by grass but also includes temperate broad-leaved evergreen trees (TeBEs). In comparison to the base case, SWUSA experiences an increase in vegetation during El Niño. TeBE's area coverage

expands by 2 %, and warm C₄ grass (C₄G) increases by 4.4 %. On the other hand, during La Niña, both TeBE and C₄G diminish by 4.9 % and 3.4 %, respectively. Over the Amazon, there are also notable changes in PFT coverage, especially in the El Niño scenario, where tropical broad-leaved trees (TrBEs) decrease by 10.2 %. On the other hand, TrBE increases by 2 % during La Niña. Over SEAsia, TrBE decreases by 8.6 % during El Niño and increases by 4.9 % during La Niña. The area over India consists of more PFTs and changes compared to the base case, which mostly occur in the La Niña scenario where the fractional coverage of TrBE increases by 2.5 %. Over CWAfr, SE Afr, NAus, and globally, changes in the fractional coverage of different PFTs during the ENSO events were found to be negligible. We observed plant growth expanding into unvegetated territory in scenarios with increased total vegetation. Even in densely forested areas, such as the Amazon region considered in this study, there are patches of land where conditions do not support vegetation. For example, the total vegetated area (including all 12 PFTs) across the Amazon adds up to 92.8 % in the base case and 94.8 % in the La Niña scenario, meaning that new vegetation was established on 2 % of the land area.

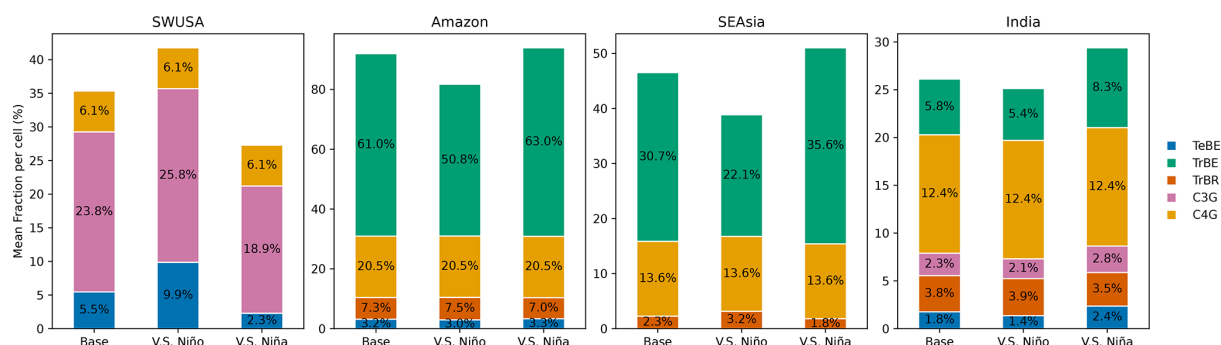


Figure 6. Stacked bar plots for the fractional coverage (% of the total area) per PFT during the base case, very strong El Niño, and very strong La Niña scenarios over the USA, the Amazon, Asia, and India. Only PFTs with a fractional coverage higher than 1 % are shown. The PFTs shown are temperate broad-leaved evergreen trees (TeBEs), tropical broad-leaved evergreen trees (TrBEs), tropical broad-leaved rain-green trees (TrBRs), cool C₃ grass (C₃G), and warm C₄ grass (C₄G). Based on the average of the last 30 years of 50-year simulations.

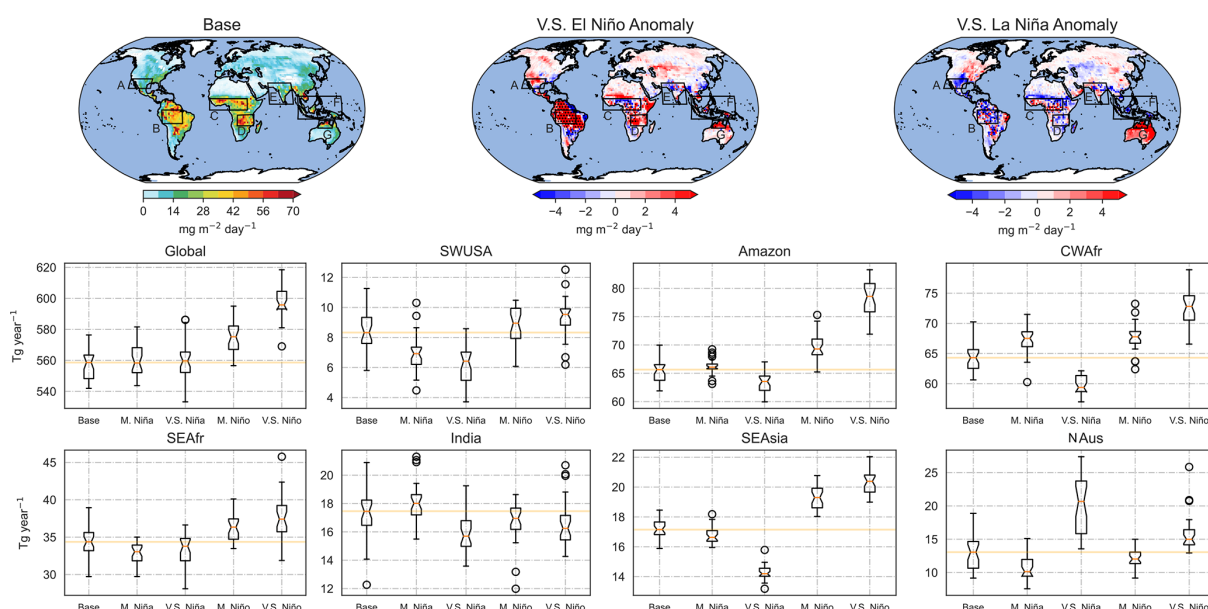


Figure 7. Global isoprene fluxes for the base scenarios as well as changes in emissions during very strong El Niño and La Niña events. The bar plots show emission changes for the different scenarios. The median of the base scenario is indicated in orange.

3.2.3 BVOC emission changes

Figure 7 shows isoprene fluxes for base conditions as well as emission changes during very strong El Niño and La Niña scenarios averaged over 30 years. The box-and-whisker plots compare emission values for all scenarios over different regions. Global annual isoprene emissions in the base case are 557 Tg. Only El Niño scenarios yield a statistically significant increase in yearly emissions with 573 Tg (+2.87 %) and 597 Tg (+3.95 %) for moderate and very strong scenarios, respectively. Over SWUSA, isoprene emissions during the base scenarios are 8.3 Tg yr⁻¹, while in the

moderate and very strong La Niña scenarios emissions are 7 Tg yr⁻¹ (−15.66 %) and 6.1 Tg yr⁻¹ (−26.51 %), respectively. Changes during El Niño over SWUSA are statistically insignificant. In the base scenario, emissions over the Amazon are 64.5 Tg yr⁻¹, and while anomalies during La Niña are statistically insignificant, isoprene emissions increase to 68.15 Tg yr⁻¹ (+5.66 %) and 77.35 Tg yr⁻¹ (+19.84 %) in moderate and very strong El Niño scenarios, respectively. In CWAfr, anomalies are statistically significant in all scenarios. There is an increase of 2.87 % and a decrease of 6.96 % in moderate and very strong La Niña scenarios, respectively,

and an increase of 4.26 % and 11.48 % in moderate and very strong El Niño scenarios, respectively.

In SE Afr, statistically significant anomalies only occur in the very strong El Niño scenario with an increase from 34.13 Tg yr⁻¹ during the base scenarios to 38.13 Tg yr⁻¹ (+11.72 %) during the very strong El Niño. Over India, changes in emissions in all scenarios are statistically insignificant. Emissions from SE Asia are 16.56 Tg yr⁻¹, and while anomalies for moderate La Niña are not statistically significant, during very strong La Niña emissions drop to 14.99 Tg yr⁻¹ and increase to 19.22 and 19.92 Tg yr⁻¹ during moderate and very strong El Niño scenarios, respectively. Lastly, over NAus, base emissions are 12.95 Tg yr⁻¹ and increase to 20.14 Tg yr⁻¹ during very strong La Niña, the only statistically significant anomaly. We again emphasise that these emission changes may be exaggerated as they include long-term changes in the biosphere in this climatic mode. These changes could be seen as an upper range for ENSO-induced emission flux changes in this climatic mode.

Correlations between fluxes and driving variables

Figure 8 displays relationships between the standardised isoprene flux anomalies and surface temperature, radiation, and AI standardised anomalies over land for the different regions during sustained El Niño and La Niña simulations over a 30-year period. Surface temperatures and isoprene fluxes are generally positively correlated during El Niño. The standardised isoprene emissions anomalies correlate well with standardised anomalies for the net solar surface radiation standardised anomaly over the Amazon (0.68), SE Afr (0.47), and SE Asia (0.53) during El Niño and also during La Niña with $r = 0.81$, 0.49, and 0.68 for the Amazon, SE Afr, and SE Asia, respectively. This highlights the influence of light availability on photosynthesis and subsequent isoprene production. The isoprene flux anomaly correlates negatively with AI with moderate correlations over the Amazon and SE Afr during El Niño and over the Amazon, SE Afr, and SE Asia during La Niña, highlighting the link between water availability and isoprene emissions in these regions. Generally, the vegetation activity (both NPP and LAI) positively correlates with isoprene emissions. In both El Niño and La Niña events, NPP moderately correlates with isoprene fluxes over SE Afr (El Niño $r = 0.51$, La Niña $r = 0.55$), India (El Niño $r = 0.65$, La Niña $r = 0.57$), and NAus (El Niño $r = 0.80$, La Niña $r = 0.80$), while moderate positive correlations with the LAI are seen over northwestern USA (NWUSA; El Niño $r = 0.58$, La Niña $r = 0.43$), India (El Niño $r = 0.65$, La Niña $r = 0.68$), and NAus (El Niño $r = 0.87$, La Niña $r = 0.86$).

For each grid cell of the model output, principal component analysis (PCA) was used to determine the variable whose ENSO anomaly correlates most strongly with the BVOC anomaly in order to find the primary cause of the variations in BVOC emissions caused by ENSO. Figure 9 shows

the spatial distribution of the dominant variable that correlates most strongly with changes in isoprene fluxes. BVOC emission changes in the tropics exhibit a predominant influence on temperature, particularly in Africa. However, there are also regions where the flux of BVOCs is primarily driven by surface radiation and anomalies in LAI, most notably in the western part of South America. ENSO-induced BVOC emission anomalies in subtropical regions of Africa, as well as in the southern United States, southeastern Asia, and Australia, seem to be mostly driven by changes in LAI. In the northern latitudes, particularly across the boreal forest, surface radiation emerges as the primary driver of changes in BVOC emissions.

4 Discussion

We suggest that the simulations presented in this study are particularly useful as they isolate the response of BVOC emissions resulting from SST/SIC anomalies during ENSO events, thus avoiding additional forcing from anthropogenic emissions of aerosols and CO₂, which are known to influence BVOC fluxes (Heald et al., 2009). However, we acknowledge that these simulations may not fully capture the Earth system's response to ENSO. For instance, our approach simplifies the complexity of the climate system by disregarding the influence of other climate drivers that may interact with ENSO, for example, feedbacks via chemistry, SOA, radiation, and aerosol–cloud interactions. The sustained ENSO simulations also result in inflated BVOC emissions as they also capture long-term changes in the biosphere. However, these simulations are mostly intended for the statistical evaluation of the driving variables' response to ENSO and the subsequent changes in BVOC emissions. Furthermore, the simulated vegetation does not include anthropogenic deforestation (and land use changes), so the simulated vegetation patterns represent potential natural vegetation. EMAC has been shown to have a good representation of the hydrological cycle with a moist bias at the southern edge of the Himalayas and a dry bias in Amazonia (Xin et al., 2019; Jöckel et al., 2016). This known bias could also lead to amplified water stress affecting BVOC emissions in this region.

While we are confident that the results presented here are relatively robust, we must acknowledge some caveats in our modelling system configuration. Parameterisations employed by the model, especially the empirical nature of the BVOC submodels, might not be able to fully capture all the complexities influencing BVOC emissions from ENSO perturbations. In this study, we also evaluate pure flux emissions from the land biosphere without taking into account changes in atmospheric chemistry and aerosols that may have feedback effects on vegetation and therefore BVOC emissions. For example, recent studies have suggested that SOA from BVOC emissions influences surface radiation both directly through scattering and indirectly via cloud interactions

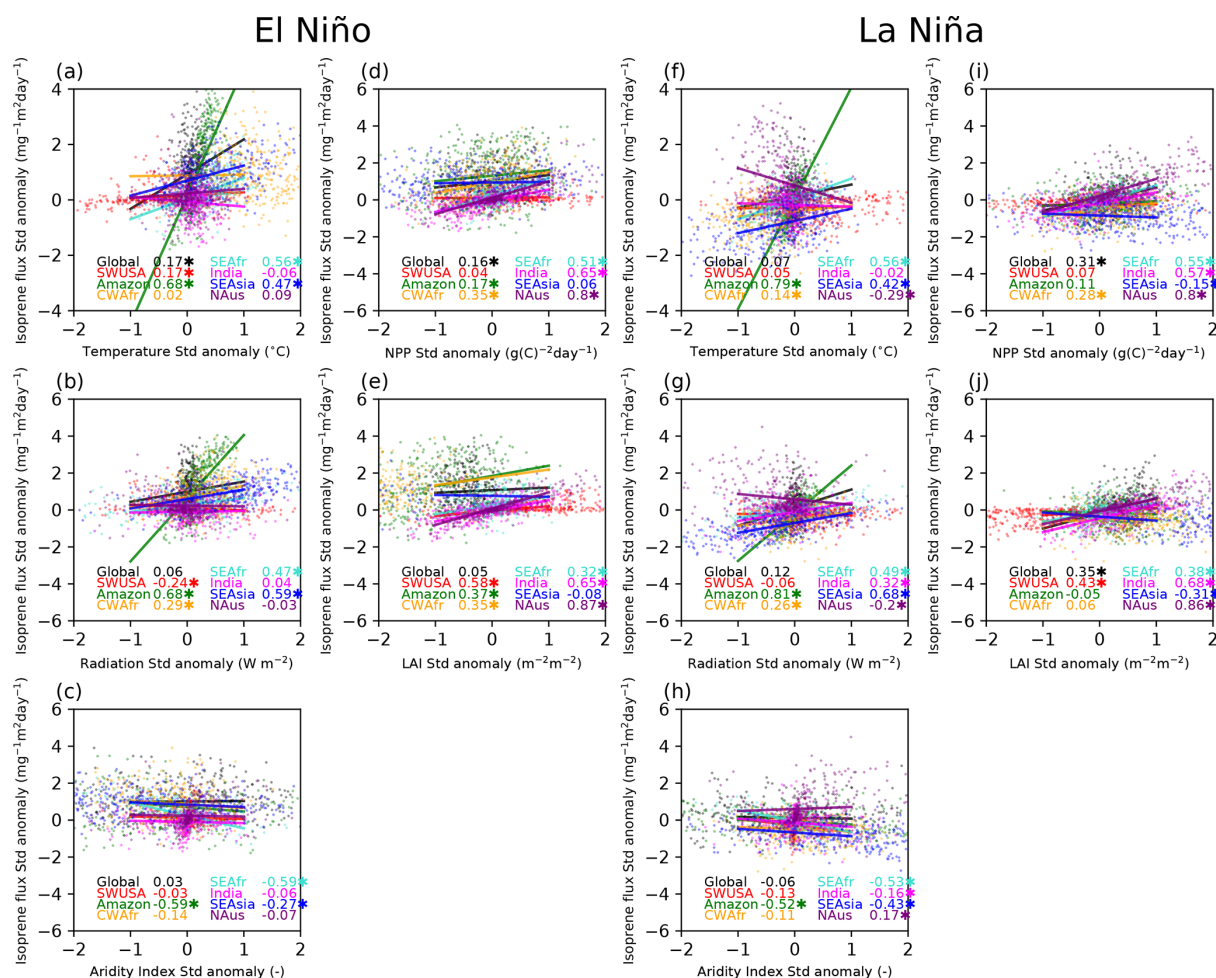


Figure 8. Scatterplots with a linear regression fit including the Pearson correlation (r) between the standardised isoprene flux anomalies and standardised temperature, radiation, AI, NPP, and LAI anomalies in different regions for very strong El Niño (top panels) and La Niña (bottom panels) events. Correlations with $p < 0.01$ are marked with an asterisk.

and that higher aerosol scattering is associated with higher BVOC emissions, while chemical mechanisms also play a role in such feedbacks (Scott et al., 2014; Sporre et al., 2019; Weber et al., 2022).

4.1 Isolated ENSO simulations

The BVOC emission anomalies from our isolated simulations agree with previous work linking high BVOC emissions with El Niño years (e.g. 1983, 1987, 1990–1991, and 1994–1995) and lower emissions with La Niña years (e.g. 1984–1985 and 1988–1989) (Lathière et al., 2006; Naik et al., 2004). Lathière et al. (2006) showed that, with CO_2 concentrations fixed to 1983, isoprene emissions are higher (1.92 %) during El Niño years and lower (−0.63 %) during La Niña years compared to the 1983–1995 average. Our simulations,

based on 1997/1998 (very strong El Niño) and 1988/1989 (very strong La Niña), suggest an increase (2.9 %) and a decrease (−0.1 %) for El Niño and La Niña, respectively. The disagreement in the magnitudes could arise from differences in the model configurations and the fact that the time frames for base conditions differ. Our results suggest that this asymmetry in BVOC emissions during El Niño and La Niña arises from regional anomalies within the corresponding ENSO occurrences. However, the observed dissimilarity in asymmetry among various regions implies that this phenomenon is influenced by the distinctive responses of varied vegetation types. While LPJ-GUESS simulates consistent responses in terms of water uptake and water limitation on photosynthesis across all PFTs, the phenological response varies among them. Notably, rain-green trees and grasses, common in sa-

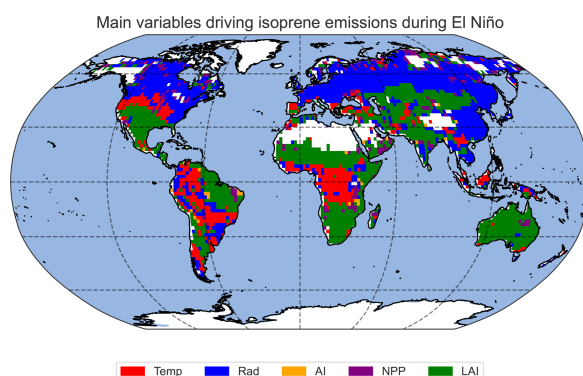


Figure 9. Map showing the variables whose anomalies correlate most strongly with the isoprene flux change for every pixel of the model output. The correlations between the isoprene flux and the temperature, radiation, AI, NPP, and LAI anomalies are classified using PCA.

vanna ecosystems, tend to “switch off” their leaves at low soil moisture levels to better handle reduced precipitation. This can lead to reduced GPP/NPP, subsequently affecting emissions of BVOCs.

We demonstrate that a single ENSO event has a lasting effect on BVOC emission fluxes, persisting for several years after the event. Even when the ENSO event subsides and normal climatic conditions resume, the biosphere may take time to recover. The prolonged perturbations in BVOC emissions can be attributed to the intricate interplay of various factors, including small changes in microclimate and longer-lasting changes in vegetation. Previous studies have also documented the prolonged impact of ENSO on climatic variables, particularly precipitation and vegetation dynamics (e.g. Lv et al., 2022; Detsch et al., 2016).

Generally, the correlations during the event years compared to the subsequent 2 years are stronger for temperature and radiation, but for AI, NPP, and LAI the correlations in the 2 years following the event are very comparable and in some cases stronger. This indicates that BVOC fluxes respond immediately to anomalies in temperature and radiation, but other driving variables such as AI, NPP, and LAI influence the emissions over longer timescales. It is evident that the correlation patterns vary in different regions during and following El Niño and La Niña events. While temperature and surface radiation seem to play an important role in driving BVOC emissions in most regions, the correlations with the other driving variables show that the biosphere is indeed responding to other signals, emphasising the need for a comprehensive understanding of the underlying mechanisms influencing BVOC emission patterns. This also implies that statistical regression models can hardly capture the processes and dynamics that more complex ecophysiological models like LPJ-GUESS simulate. It is important to note that this

approach has limitations. It simplifies the complexity of the climate system by disregarding the influence of other climate drivers that may interact with ENSO. These drivers can affect the overall climate and may modulate the impact of ENSO on BVOC fluxes in the real world. Therefore, while isolated ENSO simulations provide valuable insights into the ENSO–BVOC relationship, they should be interpreted in the context of the broader climate system and the potential interactions with other drivers.

4.2 Sustained ENSO simulations

It has been suggested that changes in weather patterns during ENSO events are linked to the rearrangement of the Walker circulation convective centres and teleconnections with mid-latitude westerlies (McPhaden et al., 2006; Dai and Wigley, 2000). Our simulations agree with previous studies suggesting that during El Niño the tropics become warmer and drier (Gong and Wang, 1999; Dai and Wigley, 2000), while some areas such as western North America and eastern Asia tend to be cooler and wetter (Ropelewski and Halpert, 1986; Wu et al., 2003). Bastos et al. (2013) investigated the variations in temperature, radiation, and precipitation during El Niño and La Niña events from 2000 to 2011 and revealed significant changes in these variables. Positive temperature anomalies exceeding 1 °C were observed in the Amazon, central and southern Africa, and northern Australia during El Niño, while cooler temperatures were detected in the USA and Europe. In our study, we also observe similar trends, with the strong signal over Australia coming from the significantly cooler temperatures during La Niña. However, we did not observe such a pronounced influence on European temperatures. Furthermore, our results align with the findings from Bastos et al. (2013) in terms of surface radiation changes. Regarding precipitation, the signal in Bastos et al. (2013) was less distinct, but a decrease in precipitation in the Amazon region during El Niño was suggested. Consistent with these findings, our study revealed higher aridity in the Amazon, supporting the notion of decreased precipitation in this region during El Niño events.

The changes in temperature, surface radiation, and AI during El Niño and La Niña events can have significant impacts on vegetation, as reflected in the changes in NPP and LAI shown in Fig. 5 and Table 4. Our findings are consistent with previous studies linking low global NPP to El Niño years and high global NPP to La Niña years (Zhang et al., 2019; Bastos et al., 2013; Nemani et al., 2003; Behrenfeld et al., 2001). In areas where vegetation growth is at least partly limited by water availability (see Nemani et al., 2003, for global estimates of limiting factors), higher temperatures lead to higher evapotranspiration rates and increased water stress on vegetation, which may result in reduced NPP and decreased LAI. Cooler temperatures can have varying effects on vegetation, depending on the specific ecological conditions of the region. However, increased surface radiation can enhance

photosynthesis and potentially lead to higher NPP. The positive anomalies in NPP observed during La Niña in several regions, such as SWUSA, SEAsia, and NAus, may be attributed to the combined effects of cooler temperatures and increased radiation.

4.2.1 Global and regional BVOC emission response

BVOC emission changes observed in the sustained ENSO simulations exhibit similar tendencies to those observed in the isolated ENSO simulations. However, the sustained simulations show a stronger signal and larger amplitudes in the anomalies of BVOC emissions. This enhanced signal can be attributed to the inclusion of long-term changes in the biosphere, as illustrated in Fig. 6. Lathière et al. (2006) showed stronger correlations between biogenic emissions and the Southern Oscillation Index (defined as the pressure difference between Tahiti and Darwin, which provides a measure of the intensity of El Niño and La Niña episodes) over tropical regions compared to other parts of the world, in particular in the Amazon forest, southeastern Africa, and southeastern Asia. Our findings also show statistically significant changes in isoprene emission fluxes over these regions as shown by the hatched areas in Fig. 7 (and similarly for monoterpenes; see Supplement).

This simulated positive correlation between BVOC emissions and temperature can be attributed to the enhanced enzymatic activity of plants in higher temperatures (Monson et al., 1992; Sharkey et al., 1996), while the positive correlation with surface radiation could be explained by increased rates of photosynthesis, resulting in enhanced BVOC emissions from vegetation (Sharkey et al., 1996; Harley et al., 1999). It has recently been suggested that limited access to water through soil seems to influence isoprene emissions predominantly through growth stress and, to a lesser extent, closure of stomata, while monoterpene emissions are mostly limited by stomatal closure (Bonn et al., 2019). Similarly, temperature stress also substantially influences BVOC emission fluxes, as can be seen for example by the power-law functions of temperature in the description of isoprene emissions (Guenther et al., 2012), e.g. linking a 2 °K temperature anomaly to 23 % increased isoprene emissions in China. Areas with higher vegetation productivity tend to exhibit increased isoprene emissions as more carbon resources may be available for isoprene synthesis within actively growing vegetation. Higher LAI values indicate greater foliage density and, consequently, increased potential for isoprene production. The correlations between the driving variables and BVOC anomalies in the regions considered are characterised separately in the following sections.

Southwestern USA

In SWUSA, BVOC emissions are higher during El Niño and lower during La Niña. However, the correlations between

isoprene emission fluxes and factors such as temperature, radiation, AI, and NPP are negligible (with a correlation coefficient below 0.29) in both scenarios. Nevertheless, we do observe weak-to-moderate correlations between the isoprene emission flux and LAI in both El Niño and La Niña. Similarly, for monoterpenes, we also find moderate correlations with LAI in both El Niño and La Niña. This suggests that LAI is the primary factor driving BVOC emission fluxes in the southwestern USA during sustained El Niño and La Niña events.

Amazon Basin

In the Amazon region, BVOC emissions are higher during El Niño and lower during La Niña, although the effect during La Niña is relatively weaker compared to El Niño. We have observed that isoprene and monoterpene emission fluxes are moderately to strongly positively correlated with temperature and radiation and weakly to moderately negatively correlated with AI. However, the correlations with NPP and LAI are weak to negligible. These findings suggest that temperature, radiation, and AI strongly influence isoprene emissions in the Amazon region during both El Niño and, to a lesser extent, La Niña, with temperature and radiation playing particularly significant roles.

Central western Africa

In CWAfr, the signal in the BVOC flux anomaly is not very clear. During La Niña, there is an increase in the moderate scenario and a decrease in the very strong scenario. In El Niño, there is a slight decrease in the moderate scenario and an increase in the very strong scenario. In these regions, correlations with the driving variables were found to be insignificant. Only weak positive correlations between the BVOC emissions and NPP/LAI were found during La Niña. This suggests a potentially greater influence of vegetation on BVOC emissions in this region.

Southeastern Africa

In SEAfr, there are positive BVOC anomalies during El Niño and negative anomalies during La Niña. We have observed weak-to-moderate positive correlations between BVOC emissions and temperature and radiation, as well as weak-to-moderate negative correlations with AI. However, for monoterpenes, the correlation with radiation is negligible. Additionally, we have found moderate-to-strong positive correlations between isoprene/monoterpene fluxes and NPP. While moderate-to-strong positive correlations exist between monoterpene emissions and LAI, the correlation with isoprene is weak. These correlations suggest that all driving variables are important factors in modulating BVOC emissions in this region.

India

The signal in India from our simulations is not very robust. Although there is a slight increase in BVOC emissions during moderate La Niña, all other scenarios indicate a decrease in emissions. Regarding the correlations of the BVOC flux anomaly with the driving variables, only NPP and LAI show moderate-to-strong positive correlations. This suggests that BVOC emissions in India during sustained ENSO scenarios are likely to be driven by changes in vegetation.

Southeastern Asia

In SEAsia, we have observed a significant increase in isoprene emissions during very strong El Niño and a significant decrease during La Niña. Conversely, monoterpene emissions decrease during El Niño and increase during La Niña, although these changes are not statistically significant. We have found weak-to-moderate correlations between isoprene emissions and temperature (positive), radiation (positive), and AI (negative), while correlations with NPP and LAI are negligible. Monoterpene emissions correlate with temperature and radiation but not with AI, NPP, or LAI. These results indicate that temperature and radiation have a notable impact on BVOC emissions in SEAsia during ENSO.

Northern Australia

In NAus, BVOC emissions experience a slight decrease during moderate El Niño and La Niña and an increase during very strong El Niño and La Niña, with a stronger signal during La Niña. Both El Niño and La Niña scenarios show weak correlations between BVOC flux and temperature, radiation, and AI. However, strong positive correlations with NPP and LAI are observed during both El Niño and La Niña. These findings suggest that BVOC emissions in NAus are strongly influenced by NPP and particularly by LAI, regardless of the ENSO phase.

In some regions, e.g. SEAsia (all scenarios except moderate Niña) and the Amazon (moderate Niña scenario) (see Figs. 7 and S5), we notice asymmetry in the isoprene and monoterpene emissions. The isoprene and monoterpene parameterisations in ONEMIS only differ in the emission factors and the correction factor based on the number of carbon atoms per molecule. This means that the different emission factors can lead to variations in the overall emission rates between the two compounds, even when other variables are the same. For example, if the emission factor for isoprene is assigned a higher weight compared to monoterpene, the model will amplify the effect of the corresponding variable (e.g. temperature) on isoprene emissions, resulting in a larger increase in isoprene fluxes compared to monoterpene fluxes. Conversely, if the emission factor for monoterpene is given a higher weight, the model will prioritise the effects of that

variable, potentially leading to a larger decrease in monoterpene emissions compared to isoprene emissions.

From our PCA with the driving variables and BVOC fluxes, we conclude that in the northern latitudes, changes in surface radiation from ENSO correlate the best with changes in BVOC emissions, suggesting that in this region BVOC fluxes are mostly influenced by surface radiation anomalies, typically linked with cloud cover and aerosol changes (Scott et al., 2018; Petäjä et al., 2022). Boreal forests, found in higher latitudes, are typically characterised by colder climates with shorter growing seasons. In these regions, the availability of sunlight, represented by surface radiation, plays a crucial role in determining photosynthetic activity and plant growth. Changes in surface radiation, such as alterations in cloud cover or atmospheric conditions, can directly impact the amount of solar energy reaching the vegetation canopy. Increased surface radiation can enhance photosynthesis and, subsequently, BVOC emissions in boreal forests, where plants are sensitive to changes in light availability. However, modelling studies with similar vegetation models suggest that high-latitude NPP is primarily limited by cold and short growing seasons, i.e. temperature, not radiation (Lucht et al., 2002; Nemani et al., 2003). This implies that vegetation growth and BVOC controlling factors are not identical. Even though links between ENSO and European precipitation are evident, as pointed out by e.g. Martija-Díez et al. (2023), the effects of the precipitation changes are outweighed by the changes in radiation, resulting in less strong effects compared to the low latitudes. The high dependence of BVOC fluxes on changes in surface radiation over boreal forests also indicates potential links among emissions of secondary organic aerosol (SOA) precursors, cloud cover, and albedo. This model setup will be used to further investigate the aerosol–cloud–radiation interactions driven by BVOC emissions.

The dependencies highlighted in Fig. 9 are complex but can be associated with the magnitude of the anomalies of the driving variables and with how different plant species within a specific microclimate respond to such changes. In the tropics, especially in central Africa, changes in isoprene fluxes from ENSO are mostly driven by temperature anomalies. For example, in central Africa during El Niño, we have strong positive temperature anomalies, but anomalies in surface radiation and aridity are comparatively smaller (see Fig. 4). This observation potentially elucidates why temperature serves as the primary driver of BVOC anomalies in this particular area. On the other hand, in northeastern South America, we observe a substantial impact on temperature, accompanied by significant alterations in surface radiation and AI. These combined effects likely contribute to the robust signal in NPP and, more specifically, to the changes observed in LAI in this region.

Our findings indicate that in the southern USA, northeastern South America, southern Africa, central Asia, and Australia, BVOC anomalies are primarily influenced by changes

in the leaf area index (LAI) resulting from the adaptation of vegetation to new climate states. Although LAI is inherently influenced by atmospheric conditions, the prolonged alterations in LAI – resulting from changes in precipitation patterns, temperature, and radiation regimes associated with sustained ENSO conditions – have a significant impact on BVOC emissions. In the case of monoterpenes (Fig. S7), we see a similar response, but we notice that emissions are less driven by radiation anomalies even at higher latitudes. It is also important to note that the changes presented in this study do not account for anthropogenic influences, such as land use changes, deforestation, and increasing CO₂ concentrations, which would also influence the response of the biosphere and BVOC emissions.

The results presented here mostly focus on isoprene emissions; however, relevant plots for monoterpene emissions can be found in the Supplement. Monoterpenes' response to the factors driving emissions is similar to that of isoprene; however, the weights of the emission factors are responsible for the different responses. In terms of atmospheric chemistry, oxidation products from monoterpenes are more likely to partition into the particle phase but are emitted in much less quantities. Nevertheless, some plant types are more likely to emit isoprene, whereas others are stronger sources of terpenes.

5 Conclusions

This work sheds light on the complex interactions among atmospheric, oceanic, and vegetation processes during ENSO events. Compared to previous work with historical transient simulations (or observations), we used isolated and sustained ENSO scenarios to constrain BVOC emission flux anomalies resulting from ENSO without additional natural forcing from the climate system. The isolated ENSO event simulations suggest a global increase of 2.9 % in isoprene emission fluxes over 2 years with a very strong El Niño event. These simulations also show that a single ENSO event perturbs emission fluxes so profoundly that they do not return to baseline emissions even several years after the event.

We report potential vegetation changes as persistent ENSO conditions shift the vegetation productivity and structure (LAI) into a new quasi-equilibrium state, whereby changes in PFT composition are more transient. Over SEAsia, the fractional coverage of tropical broad-leaved evergreen trees could decrease by 8.6 % and increase by 4.9 % during persistent El Niño and La Niña scenarios, respectively. Our results show that if ENSO, particularly El Niño, becomes more persistent in the future we should expect amplification in BVOC flux changes as we enter this new climatic mode with a long-term influence on the biosphere. In this climate mode, isoprene emissions from the Amazon could increase by 19 %. We also show that ENSO-induced variations in BVOC emissions over the tropics are largely related to surface temper-

ature anomalies, whereas surface radiation predominates at higher latitudes. Variations in LAI were strongly linked to anomalies in BVOC emissions over the subtropics, indicating that ENSO-induced changes in plant phenology are an important driver of BVOC emissions.

In this work, we highlight that the impact of ENSO on the Earth system goes beyond changes in temperature and precipitation but also significantly impacts BVOC emissions, which can have far-reaching implications for the atmosphere and climate. ENSO-induced BVOC emissions changes contribute to the formation of SOA, potentially leading to radiation–chemical feedbacks. As BVOC emissions are projected to rise in a warming climate, it becomes imperative to understand and quantify these weather disturbances to accurately predict future BVOC emissions, SOA formation, and their climate feedbacks. Additionally, BVOCs are crucial players in the formation of tropospheric ozone and other harmful air pollutants, posing risks to human health and regional air quality, especially in communities located close to dense forests. Furthermore, BVOCs can act as precursors for greenhouse gases like methane, exacerbating the overall radiative forcing and contributing to climate change. Therefore, it is crucial to understand the intricate interplay among ENSO, BVOC emissions, atmospheric chemistry, and climate for accurately predicting and mitigating the far-reaching impacts of ENSO in the climate system.

Data availability. The datasets and analysis scripts used in this work are available here: <https://doi.org/10.5281/zenodo.8160152> (Vella et al., 2023b).

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3.4 Deforestation impacts on BVOC emissions and aerosols

This chapter includes a paper that will soon be submitted to the journal *Atmospheric Chemistry & Physics* as a research article. I am the lead author of this work, where I worked on the model setup, ran the simulations, analysed the data, generated the figures, and wrote the manuscript. The co-authors contributed to the model setup, interpreting the results and editing the manuscript. The manuscript includes an *Author contributions* detailing individual contributions. I look forward to the review process and am optimistic about the positive impact my research will have once published.

The impact of deforestation on biogenic volatile organic compound emissions and the atmospheric aerosol burden

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Abstract.

Biogenic volatile organic compounds (BVOC) are emitted in large quantities from the terrestrial biosphere and play a significant role in atmospheric gaseous and aerosol composition. Such emissions account for 90% of the total global volatile organic compound (VOC) emissions and exert a significant influence on the atmosphere's oxidation capacity. BVOCs are precursors for the formation of secondary organic aerosols (SOA), which affect the radiation budget both directly through scattering and absorption and indirectly through modifying cloud formation, properties and distribution. Human activities have extensively altered natural vegetation cover, primarily by converting forests into agricultural land. In this work, a global atmospheric chemistry-climate model with interactive vegetation was employed to study the impacts of perturbing the biosphere through human land use change, consequently exploring changes in BVOC emissions and atmospheric aerosol burden. Given that our vegetation model simulates potential natural vegetation (PNV), a land use scheme was implemented to constrain the Tree Plant Functional Type (PFT) cover based on deforestation fraction maps from the year 2015. The PNV scenario is compared with: (1) present-day deforestation, based on the deforestation of both crop and grazing land cover, and (2) an extreme reforestation scenario assuming that all grazing land is restored to natural vegetation (i.e., with only cropland deforestation). We find that, compared to the PNV scenario, present-day deforestation results in a 26% reduction in BVOC emissions, which decreases the global bSOA burden by 0.17 Tg (a decrease of 29%), while the total OA decreases by 0.16 Tg (a reduction of 9%). On the other hand, the extreme reforestation scenario, compared to present-day land cover, suggests an increase in BVOC emissions by 22%, which increases the bSOA and total OC burden by 0.10 Tg, an increase of 25% and 6%, respectively. The assessment includes changes in the cloud condensation nuclei (CCN) and cloud droplet number concentration (CDNC) in each scenario. In the present-day deforestation scenario, we estimate a positive total radiative effect (aerosol + cloud) of 87.9 mW m⁻² (warming) compared with the natural vegetation scenario, while in the extreme reforestation scenario, we report a negative forcing (cooling) of 38.2 mW m⁻² compared to the PNV scenario.

1 Introduction

Human activities have significantly modified the natural vegetation cover, primarily through the conversion of forests into agricultural land. It is estimated that approximately half of the Earth's land surface has been affected by human activities (Hurt et al., 2011). Land cover change (LCC) has a substantial impact on the Earth system as the biosphere plays a central role in major biophysical and biogeochemical cycles, as well as feedbacks with the atmosphere (Bonan, 2008). Forests store 45% of terrestrial carbon and can sequester large amounts of carbon annually (Field and Raupach, 2004). Furthermore, forests play a significant role in sustaining the hydrological cycle through evapotranspiration, which influences cloud formation, the onset of precipitation, and consequently surface temperatures (Betts et al., 2004; Vicente-Serrano et al., 2015). Land cover plays a crucial role in surface albedo, with dense forests capable of absorbing up to 90% of solar radiation (Forster et al., 2007a). In high latitudes, forests can mask the high albedo of snow, leading to planetary warming through increased solar heating of the land (Bonan, 2008).

The terrestrial biosphere is also the primary source of biogenic volatile organic compounds (BVOCs) emissions such as isoprene and various terpenes, accounting for around 90% of the total VOC emissions to the atmosphere (Guenther et al., 1995). Reported global isoprene emissions, the primary BVOC, span a range from 412 to 682 Tg yr⁻¹ (e.g., Sindelarova et al., 2014; Guenther et al., 2006; Vella et al., 2023a). BVOCs are highly reactive and short-lived, with lifetimes typically ranging from minutes to hours. Upon emission, they rapidly interact with tropospheric oxidant gases, thereby exerting a substantial influence on the oxidation capacity of the atmosphere (Lelieveld et al., 2008; Atkinson, 2000; Atkinson and Arey, 2003).

The short lifespan of BVOCs stems from their rapid oxidation upon release from the canopy. This oxidation primarily involves the OH radical as well as other key oxidizing agents like O₃ and NO₃ radicals (Shrivastava et al., 2017). These reactions produce various lower volatility oxidation products that tend to partition into the aerosol phase or condense onto existing particles, leading to the formation of biogenic secondary organic aerosols (bSOA) (Kavouras et al., 1998; Spracklen et al., 2011). Isoprene-derived oxidation products mainly contribute to the condensation onto existing aerosols, whereas the oxidation products from monoterpenes, despite being less abundant compared to isoprene, play a crucial role in generating new particles, thus influencing SOA formation significantly (Jokinen et al., 2015). Biogenic SOA can grow sufficiently, reaching a dry radius of approximately 50 nm, thereby contributing to the aerosol fraction involved in climate interactions. In other words, bSOA participates in the absorption and scattering of radiation (direct effect) and acts as condensation nuclei modifying cloud properties such as albedo and lifetime (indirect effect) (Forster et al., 2007b).

BVOC emission rates are inherently linked to land cover, and LCC can ultimately affect the climate system by influencing short-lived climate forcers like aerosols (Scott et al., 2014). In this study, we investigate the changes in BVOC emissions following crop and grazing land expansion on potential natural vegetation (PNV). We use the chemistry-climate model EMAC coupled with the global dynamic vegetation model (GDVM) LPJ-GUESS which simulates PNV. We apply a deforestation routine in LPJ-GUESS where crop and grazing land area is systematically deforested based on 2015 land cover data. Consequently, we present changes in BVOC emissions as well as the aerosol burden and radiation impacts using advanced descriptions in EMAC including dynamic vegetation representations that respond to changes in atmospheric states.

2 Model description and methods

2.1 The EMAC modelling system

The EMAC (ECHAM/MESSy Atmospheric Chemistry) model is a numerical chemistry and climate modelling system that contains submodels that represent tropospheric and middle atmospheric processes, as well as their interactions with oceans, land, and anthropogenic activities. It originally combined the ECHAM atmospheric general circulation model (GCM) (Roeckner et al., 2006) with the Modular Earth Submodel System (MESSy) (Jöckel et al., 2005) framework and philosophy, modularising physical processes as well as most of the infrastructure into submodels that can be further developed to improve existing process representations; new submodels can also be added to represent new or alternative process representations.

The configuration of the chemistry submodels in this study closely follows the approach outlined in Jöckel et al. (2016), but include aerosol processes for tropospheric applications. Aerosols are treated using the submodel GMXe (Pringle et al., 2010), where aerosol microphysics are characterised by seven interactive lognormal modes that span the typical size range of aerosol species. These modes are further categorised into four hydrophilic (nucleation, Aitken, accumulation, and coarse) and three hydrophobic (Aitken, accumulation, and coarse) aerosol modes. The representation of all aerosols assumes spherical particles. The properties of aerosols in each mode are fully determined by factors such as total mass (internal mixture of contributing species), density, number concentration, median radius, and width of the lognormal distribution. After each simulation step aerosols may transfer between modes depending on size changes.

Organic aerosol species are additionally described by the Organic Aerosol Composition and Evolution (ORACLE) submodel (Tsimpidi et al., 2014), taking into account the partitioning between aerosols and the gas phase based on the volatility basis set (VBS) framework (Donahue et al., 2006). ORACLE describes the following organic aerosols (OA): Secondary organic aerosols from the oxidation of anthropogenic (aSOA) and biogenic (bSOA) VOCs; primary organic aerosols from emissions from fossil fuel and bio-fuel combustion (fPOA) and biomass burning (bbPOA); and secondary organic aerosols from their subsequent photochemical oxidation (fSOA and bbSOA, respectively) (Tsimpidi et al., 2016, 2017). The submodel employs a simple photochemical ageing scheme that effectively models the combined impacts of fragmentation and functionalisation of organic compounds. The module not only predicts the mass concentration of organic aerosol (OA) components but also predicts their oxidation state (expressed as O : C), enabling their categorisation into primary OA (POA, chemically unprocessed), freshly formed secondary OA (SOA, with low oxygen content), and aged SOA (highly oxygenated). By explicitly simulating the chemical conversion of OA from initial emissions to a highly oxygenated state during photochemical aging, ORACLE facilitates tracking changes in OA hygroscopicity resulting from these reactions. This allows the computation of OA particle capability to serve as cloud condensation nuclei. Details on the implementation of ORACLE (v2.0) in EMAC, including an evaluation of the model, could be found in Tsimpidi et al. (2018). The output from the ORACLE model, based on the described setup, has been compared with observational data in tropical regions. It was found that ORACLE provides OA surface concentrations within 60% of the observed values, as documented in several studies (Hewitt et al., 2010; de Sá et al., 2019; Chen et al., 2009; Schmale et al., 2013; Tiitta et al., 2014; Zhang et al., 2010).

Heterogeneous and gas-phase chemistry are treated through the MECCA submodel (Sander et al., 2019), employing the Mainz Isoprene Mechanism (MIM1) as the chemical mechanism (Pöschl et al., 2000; Jöckel et al., 2006), which includes over 100 gas-phase species and more than 250 reactions. Dry deposition, sedimentation, and wet deposition processes are simulated using the submodules DDEP, SEDI (both Kerkweg et al., 2006), and SCAV (Tost et al., 2006a), respectively. Within this framework, convective cloud processes are taken into account based on the approach proposed by Tost et al. (2006b), utilising the convection schemes from Tiedtke (1989) and Nordeng (1994). However, convective cloud microphysics is solely based on temperature and moisture profiles, without accounting for the influence of aerosols on liquid droplet or ice formation processes. The vertical velocity distribution used for aerosol activation by grid-scale clouds in EMAC is calculated as the sum of the grid mean vertical velocity and the turbulent contribution, as detailed by Brinkop and Roeckner (1995). Large-scale stratiform clouds are described by the CLOUD submodel, which, in the applied configuration, incorporates a two-moment cloud microphysics scheme for cloud droplets and ice crystals, as detailed by Lohmann et al. (1999, 2007); Lohmann and Ferrachat (2010) and Lohmann and Kärcher (2002). CLOUD solves prognostic equations for specific humidity, liquid cloud mixing ratio, ice cloud mixing ratio, cloud droplet number concentration (CDNC), and ice crystal number concentration (ICNC).

For the evaluation of radiative forcing from clouds and aerosols, we employ the methods from Ghan (2013). The calculation of net direct forcing (RF_{aerosol}) involves determining the difference between the net top-of-the-atmosphere shortwave radiative flux (F) and the radiative flux, excluding the scattering and absorption of solar radiation by the aerosols (F_{clean}). F_{clean} is computed in a separate radiation call within the radiation submodel RAD. Similarly, the net cloud forcing (RF_{cloud}) is derived by assessing the difference between F_{clean} and the flux that disregards the scattering and absorption caused by both clouds and aerosols ($F_{\text{clear-sky, clean}}$).

110 LPJ-GUESS

The Lund-Potsdam-Jena General Ecosystem Simulator (LPJ-GUESS) (Smith et al., 2001, 2014) is a dynamic global vegetation model (DGVM) featuring an individual-based model of vegetation dynamics. These dynamics are simulated as the emergent outcome of plant growth and competition for light, space, and soil resources among woody plant individuals and a herbaceous understorey in each of a number (50 in this study) of replicate patches representing random samples of each simulated locality or grid cell. The simulated plants are classified into 12 plant functional types (PFTs) discriminated by growth form, phenology, photosynthetic pathway (C3 or C4), bioclimatic limits for establishment and survival and, for woody PFTs, allometry and life history strategy. LPJ-GUESS has previously been implemented in global ESMs (e.g., Weiss et al., 2014; Alessandri et al., 2017), and, more recently, coupled with EMAC (Forrest et al., 2020; Vella et al., 2023a). The LPJ-GUESS version used in this study (v4.0) currently provides information on potential natural vegetation (PNV), and it does not incorporate changes in land use. In this work, however, a custom deforestation routine was integrated to constrain the PNV using deforestation maps. The deforestation maps consist of values from 0 to 1, where a value of 1 signifies complete deforestation within the respective grid cell. The routine kills the tree PFTs after every simulated year and inhibits trees from establishing

in the specified areas. This implementation allows us to constrain the vegetation cover and address the research questions presented in this work. However, the latest version of LPJ-GUESS (v4.1) features a more advanced land cover scheme, which will be incorporated into our current LPJ-GUESS version in future developments.

2.2 EMAC/LPJ-GUESS configuration

In this work, we use the standard EMAC/LPJ-GUESS coupled configuration, where the vegetation in LPJ-GUESS is entirely determined by the EMAC atmospheric state, soil type, N deposition, and fluxes (Forrest et al., 2020), but there is no feedback from the vegetation to climate variables (e.g., changes in albedo and roughness length). After each simulation day, EMAC computes the average daily values of 2-meter temperature, net downwards shortwave radiation, and total precipitation and passes these state variables to LPJ-GUESS. Vegetation information (leaf area index, foliar density, leaf area density distribution, and PFT fractional coverage) from LPJ-GUESS is then fed back to EMAC for the calculation of BVOC emission fluxes using EMAC's BVOC submodules. In this study, the BVOC fluxes in EMAC are calculated using the Model of Emissions of Gases and Aerosols from Nature (MEGAN) version 2.04 (Guenther et al., 2006).

MEGAN is based on the work of Guenther et al. (1993, 1995), where the BVOC emission flux is calculated as a function of PFT-specific emission factors, and non-dimensional activity factors. These activity factors consider sensitivities to the canopy environment, including parameters such as leaf area index (LAI), temperature, light, and leaf age. Notably, the current setup does not incorporate sensitivity to soil moisture. The parameterised canopy environment emission activity (PCEEAA) algorithm is used, rather than the alternative detailed canopy environment model that calculates light and temperature at each canopy depth. The PCEEAA algorithm calculates the light sensitivity within the canopy as a function of the daily average above-canopy photosynthetic photon flux density (PPFD), the solar angle and a non-dimensional factor describing the PPFD transmission through the canopy. This setup activates BVOC-aerosol-vegetation feedbacks, making vegetation and BVOC emissions sensitive to changes in temperature and above-canopy radiation (excluding diffused radiation) resulting from aerosol interactions. BVOC emissions from this model setup were evaluated and applied in other studies (e.g. Vella et al., 2023a, b).

2.3 Experimental design

The deforestation scenarios were derived from the History database of the Global Environment (HYDE v3.2) (Klein Goldewijk et al., 2017). HYDE provides a wide range of land use products, encompassing both historical and projected data. To ensure an accurate representation, we rely on HYDE's "cropland" and "grazing land" products from 2015, derived from high-resolution satellite data. These products were transformed into deforestation fraction maps (Fig. 1) to constrain the vegetation in the model. Three experiments were conducted to assess the impact of human-induced deforestation on the natural land biosphere and atmospheric composition. The initial model run used simulated PNV without any deforestation. Additionally, two more model runs were conducted, incorporating deforestation. The first scenario aimed to represent present-day deforestation levels and employed deforestation based on cropland and grazing land. This scenario is referred to as "Deforested Crop and Grazing Land" (DCGL) - Fig. 1a. DCGL will sometimes be referred to as "present-day deforestation" scenario. The second scenario involved deforestation exclusively on cropland and is referred to as "Deforested Crop Land" (DCL) (Fig. 1b). We use the DCL

scenario to evaluate the potential impact of restoring all grazing land back to its natural state, essentially creating an extreme afforestation scenario, while maintaining the present-day croplands for agricultural food production.

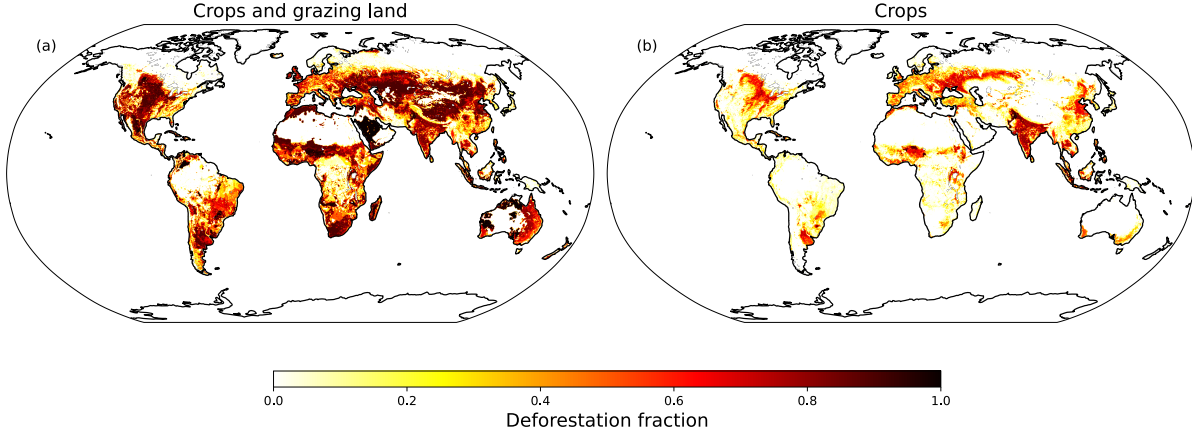


Figure 1. Deforestation maps used for the deforesting tree PFT's in LPJ-GUESS. Products derived from HYDE v3.2 based on the year 2015.

All simulations were conducted over 12 years, with the initial 2 years excluded from the analysis to ensure proper spinup and equilibrium state in the analysed data. The vegetation initial states for all simulations were taken from a previous non-chemistry 50-year run under similar atmospheric states. For this study, the simulations were performed in T63L31 resolution, i.e., approximately $1.9^\circ \times 1.9^\circ$ (or approx. 180×180 km at the Equator) with 31 vertical levels. The meteorological fields were nudged in the troposphere towards ERA-Interim reanalysis data (Dee et al., 2011) for the respective years 2000-2012. The corresponding forcing at the sea surface (Sea Surface Temperatures (SSTs) and Sea Ice Coverages (SICs)) is also inferred from the nudging data with continuous variation. Tracers were initialized using climatological data from previous simulations spanning 2000 to 2020, while the CO_2 concentrations in the radiation and vegetation schemes were kept fixed at 384 ppmv, representing the year 2015. All simulations employ surface offline emissions, based on the year 2015 forming a consistent set of boundary conditions.

3 Results

3.1 Present-day land use scenario

3.1.1 Changes in vegetation states

Fig. 2 shows the PNV scenario's spatial distribution of NPP and LAI for Tree (a-b) and Grass (e-f) PFTs. As illustrated in Fig. 2c-d, the deforestation routine implemented in LPJ-GUESS effectively prevents tree establishment in the transformed regions outlined in Fig. 1a, resulting in the dominance of grass PFTs (Fig. 2g-h) in these areas. Present-day deforestation decreases

the global LAI by $0.06 \text{ m}^2 \text{ m}^{-2}$, declining from an average of $1.52 \text{ m}^2 \text{ m}^{-2}$ in the PNV scenario. The simulations indicate a significant change in NPP patterns due to human-induced deforestation. Specifically, the global NPP originating from tree PFTs experiences a decline of 28.4 Pg (C) , while that from grass PFTs shows an increase of 26.7 Pg (C) . This implies that human deforestation activities effectively reduce the net annual carbon fixed by the terrestrial biosphere by 1.7 Pg .

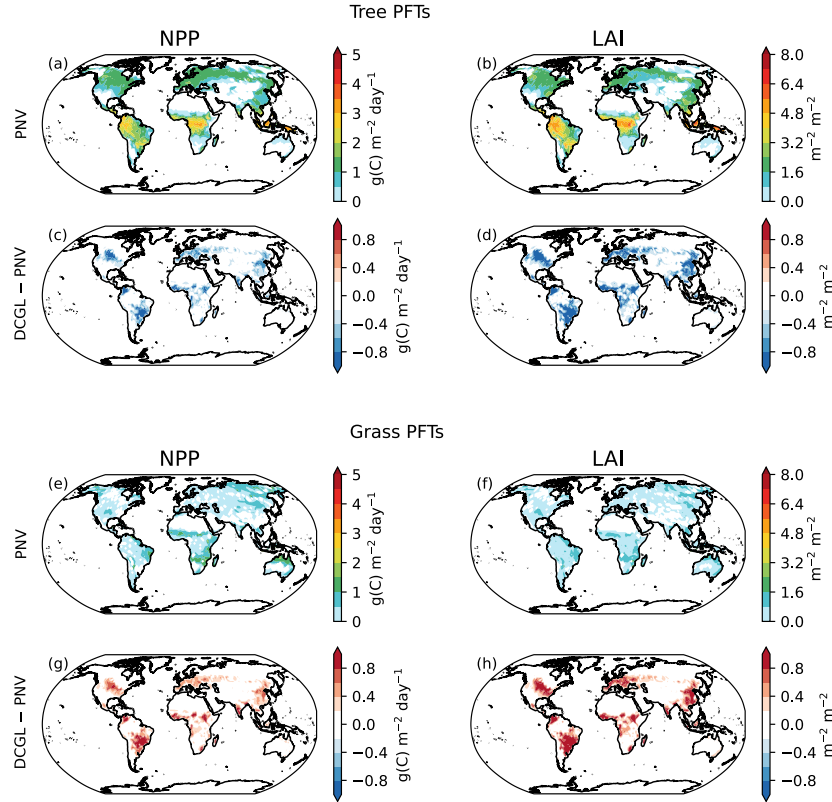


Figure 2. Changes in NPP and LAI for tree PFTs (a-d) and grass PFTs (e-h) in present-day deforestation i.e. DCGL - PNV.

3.1.2 BVOC surface emissions

The clearing of biomass from deforestation practices impacts the BVOC global emissions. Fig. 3 a-b shows the surface isoprene and monoterpene emissions in the PNV scenario. The model simulates fluxes of up to $100 \text{ g(C)} \text{ m}^{-2} \text{ day}^{-1}$ of isoprene, and $8 \text{ g(C)} \text{ m}^{-2} \text{ day}^{-1}$ of monoterpene in the tropics. Fig. c-d shows the spatial changes in the BVOC fluxes resulting from deforestation.

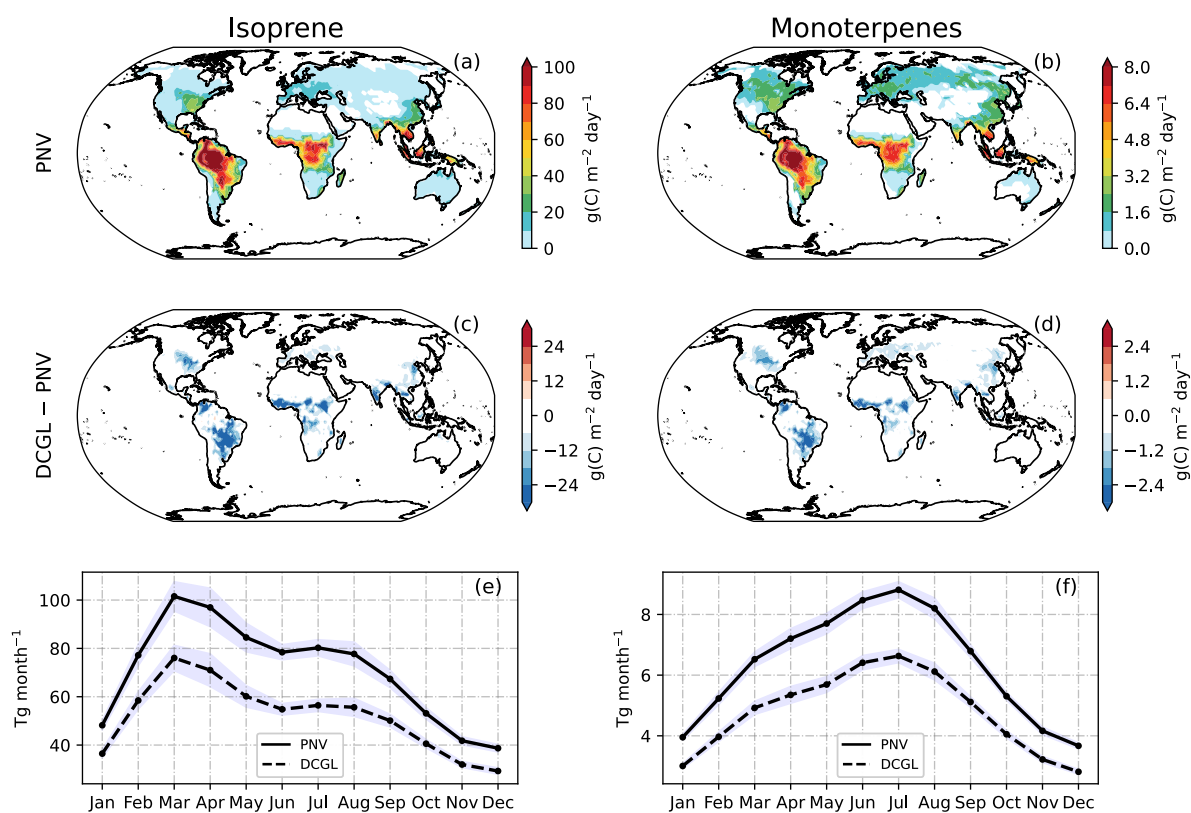


Figure 3. Isoprene and monoterpene surface fluxes for the PNV scenario (a-b). Subplots c and d show the spatial difference in isoprene and monoterpene emission fluxes (DCGL – PNV). Monthly emissions based on the 10-year average for isoprene (e) and monoterpenes (d). The shading represents 1 standard deviation derived from the monthly averages based on 10 simulated years. Fluxes in the Southern Hemisphere were shifted by 6 months to conserve the seasonal cycle.

In Fig. 3e-f, the temporal profile of global monthly emission totals is depicted, with shading indicating 1- σ variability based on 10 simulated years. To accurately capture the true seasonal cycle, values from the Southern Hemisphere were shifted by 6 months before combining fluxes from both hemispheres. In the PNV scenario, the global annual emission flux for isoprene totalled 845.7 Tg, which decreased to 620.9 Tg in the DCGL scenario, marking a reduction of 224.8 Tg (a 27% decrease relative to PNV). Similarly, for monoterpenes, the annual global emissions in PNV were 76.0 Tg and 57.3 Tg in DCGL, respectively, indicating a decrease of 18.7 Tg (a 25% decrease relative to PNV). Consequently, the global, annual source of BVOCs declined by 243.5 Tg. Isoprene emissions from MEGAN in EMAC are known to be relatively high compared to other global estimates in the literature, as shown in Vella et al. (2023a). Some atmospheric chemistry studies using EMAC, e.g., Pozzer et al. (2022b),

employ a global scaling factor to dampen the global emissions to desired values. However, in this study, no scaling factors were applied, which means that the values reported here may be slightly overestimated.

3.1.3 Aerosol burden, cloud interactions

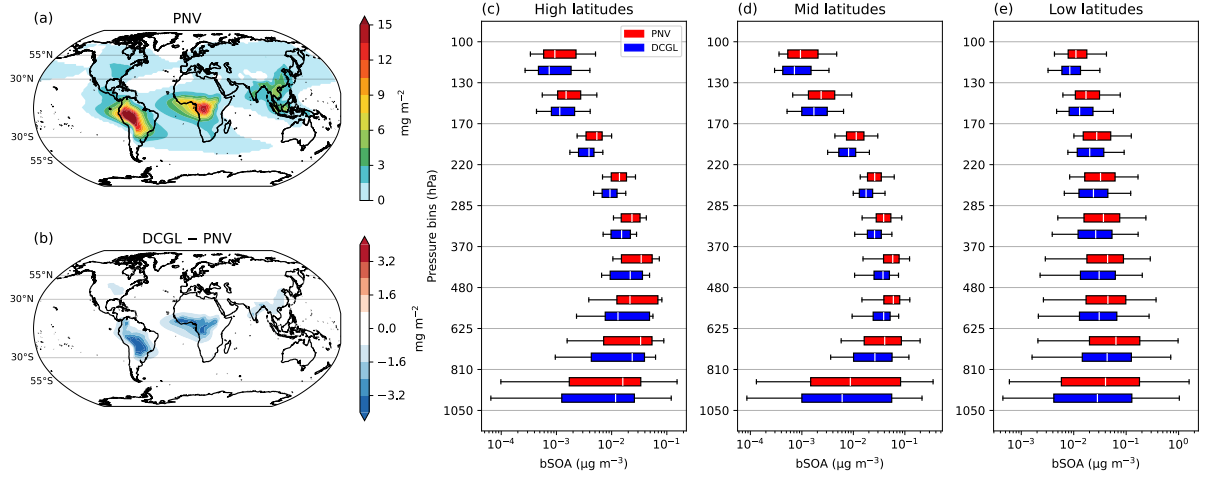


Figure 4. Total column bSOA from PNV (a) and changes in bSOA from deforestation (DCGL – PNV) (b). Panels (c), (d), and (e) show the vertical profiles of bSOA, represented by box-whisker plots for different pressure bins. The white line marks the median, the box corresponds to the lower and upper quarterlies, and the whiskers represent the 5th–95th percentile. The latitude ranges are defined as follows: High latitudes (90–55°S and 55–90°N), mid-latitudes (55–30°S and 30–55°N), and low latitudes (30°S–30°N). A log scale is used for the x-axis of (c), (d), (e).

Fig. 4 illustrates the bSOA burden, including the total column burden for the PNV depicted in Fig. 4a, as well as changes in the column burden resulting from deforestation (4b), and, the vertical profiles across three latitude bands (4c-e). The vertical profile is represented using box-whisker plots, showcasing variations at different pressure bins. In the PNV scenario, the bSOA load exhibits its highest concentration over the Amazon forest and the Congo basin, reaching a column mass of up to 15 mg m⁻². In the PNV scenario, we estimate a total annual OA atmospheric burden of 1.80 Tg, of which 0.56 Tg are bSOA, while in DCGL the total annual OA atmospheric burden reduces to 1.70 Tg, of which 0.40 Tg are bSOA. The bSOA burden decreased by 29% while the total OA decreased by 9%.

The rapid oxidation of BVOCs, yields oxygenated intermediate species at the surface, which act as precursors for the formation of bSOA. This explains the high abundance of bSOA close to the surface. As bSOA is transported in the atmosphere, concentrations typically decrease due to mixing and dilution. However, the vertical profile of bSOA is also governed by the vapour pressure of the oxygenated intermediates, which decreases with lower temperatures as they ascend in the troposphere. When they are transported upward, the reduced volatility at lower temperatures prompts the transition of oxygenated interme-

diates into the aerosol phase, contributing to the formation of bSOA. Consequently, this interplay between oxidation, transport, and vapour pressure-driven processes imparts a distinctive D-shaped vertical profile of biogenic SOA concentrations, with elevated concentrations near the surface, followed by an increase in bSOA in higher altitudes, and ultimately a decline in the upper atmosphere. In the low latitudes (tropics), the D-shape in bSOA concentration is only faint (Fig. 4e). The prevalence of warm air, larger boundary layer thickness, and the occurrence of frequently active deep convection transporting both aerosol particles as well as precursors into the upper troposphere lead to the maximum gas-to-particle partitioning occurring at higher altitudes, typically around 100-200 hPa, resulting in a second local upper tropospheric enhancement.

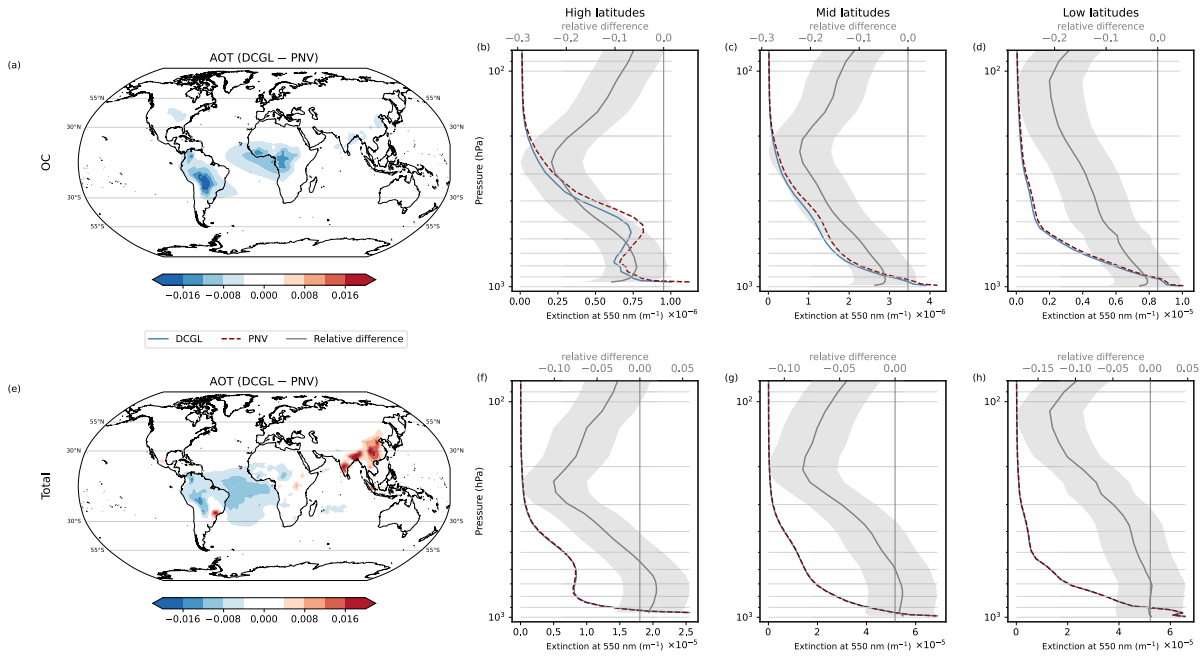


Figure 5. CAPTION NEEDED

Fig. 5 shows changes in the total column aerosol optical thickness (AOT) and extinction at 550 nm for OA (top panels) and for the total aerosols (bottom panels). The aerosol extinction refers to the fractional depletion of radiance per unit path length (m^{-1}). The aerosol optical thickness (AOT) is determined by integrating the aerosol extinction over the full atmospheric column, making it a dimensionless quantity.

Fig. 5a illustrates the absolute changes in AOT from OC and suggests a decrease of up to 0.02 over the Amazon forest and Congo basin in Africa. Panels b, c and, d, show that a consistent decrease in the OC extinction, with the highest relative difference peaking around 250 hPa in the high and mid-latitudes, and around 100 hPa in the tropics. Our calculations indicate a global decrease in the OC AOT of -7% , with corresponding decreases of -10% , -9% , and -5% in high, mid, and low latitudes. EMAC simulates six aerosol species, namely organic carbon (OC), black carbon (BC), water-soluble inorganic aerosols

(WASO), dust (DU), sea salt (SS), and aerosol-associated water (H₂O). The aerosol extinction at 550 nm from the total aerosol suggests a decrease over the tropics (Fig. 5e), however, this trend varies significantly across latitudinal bands. Notably, there is a marginal increase of 0.1% in the high latitudes with declines of -0.5% and -0.6% in mid and low latitudes, respectively. Over regional levels, the derived total AOT exhibits opposing effects, particularly, pockets of increased total AOT are evident in confined regions and notably in Southeast (SE) Asia. Here, the extinction from H₂O and WASO is found to be increasing, effectively masking the expected reduction in OC extinction from lower BVOC precursors in DCGL. This phenomenon is linked to the growth of aerosol particles into what is commonly referred to as the “Greenfield gap” - a range characterised by lower deposition velocities and scavenging values (Greenfield, 1957). This growth adds to the burden of WASO compounds and their associated water uptake, thereby amplifying aerosol extinction. Moreover, the absence of organics exacerbates this effect, leading to a disproportionate condensation on accumulation mode particles at the expense of Aitken mode particles. Consequently, the increased presence of WASO compounds, coupled with increased aerosol water content and a shift in size distribution towards larger particles, collectively contribute to the observed increase in extinction in these specific areas.

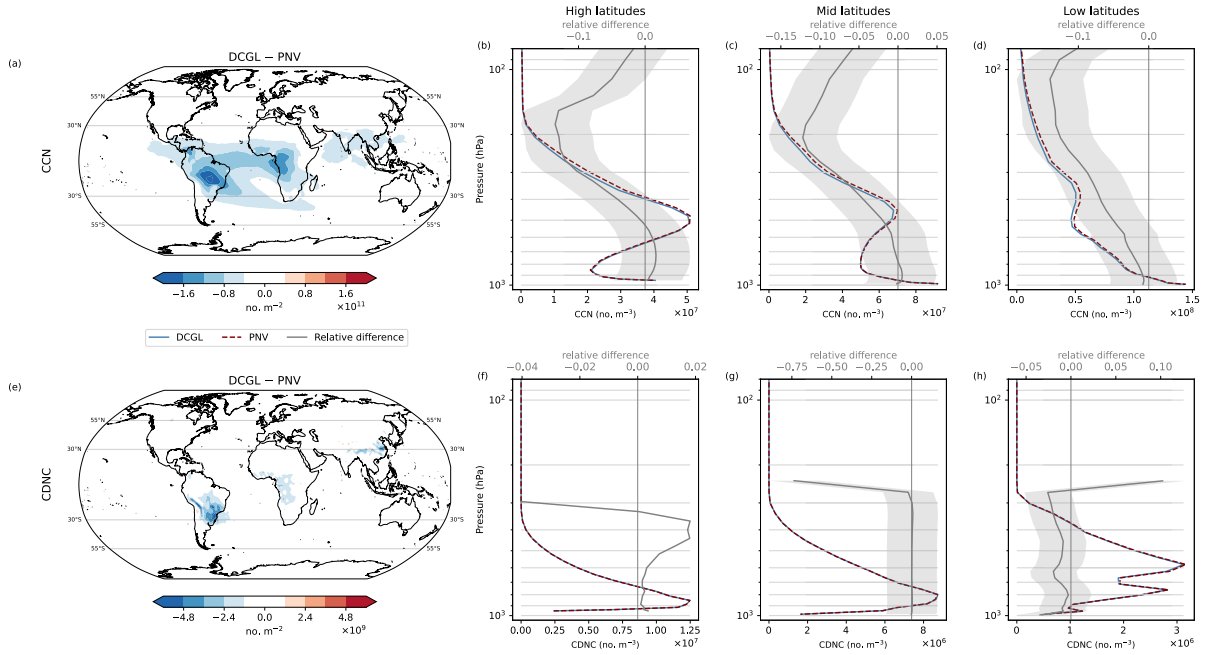


Figure 6. Changes in CCN at 0.2% supersaturation (top) and CDNC (bottom). The left panels, (a) and (e), show the difference in the total column burden (number of particles per square meter), while on the right the total-column vertical profiles from DCGL and PNV simulations and their relative difference are shown. Figure details are the same as Fig. 5.

Fig. 6 illustrates changes in the CCN at 0.2% supersaturation and CDNC. The reduced aerosol burden in DCGL leads to a CCN decrease of up to 2×10^{11} particles per m^2 over the tropical regions. Our calculations suggest a global decrease in CCN

of -4.8% , with respective declines of -3% , -5% , and -5% in high, mid, and low latitudes, respectively. Panels (b), (c), and (d) show the vertical profile of CCN for both DCGL and PNV. In high and mid-latitudes, the difference in CCN concentration peaks around 200 hPa, showing a decrease of approximately -12% in the DCGL scenario. In the tropics, the peak difference occurs higher up in the atmosphere (around 950 hPa), also with a decrease of around -12% .

Our results indicate that the lower availability of CCN (Fig. 6a) induces a decrease in CDNC in South America, Central Africa and Eastern China. Globally, deforestation leads to a decrease of -0.2% in CDNC. Spatial averages over high and mid-latitudes suggest a general increase of 0.2% and 0.3% , respectively. Conversely, over the tropics, CDNC decreases by -0.6% . Panels (f), (g), and (h), displaying the vertical profile of CDNC, affirm that the strongest influence on CDNC, with a consistent reduction over the full column, occurs in the tropics. This suggests that the shifts in aerosol loading directly impact cloud properties, particularly near the perturbed organic aerosols originating from the BVOC precursors, however, the effect appears to be small.

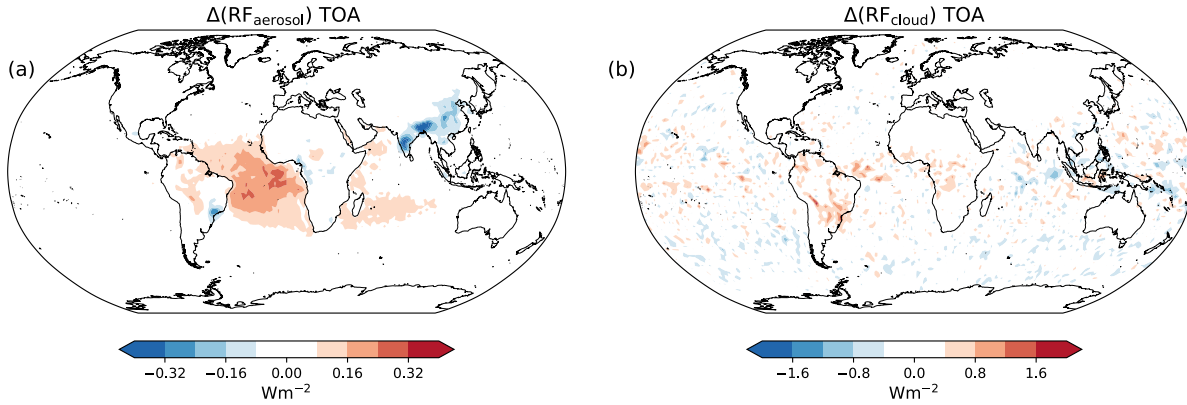


Figure 7. Aerosol and cloud radiative forcing mediated by aerosol changes from deforestation. Panels a and b show the top-of-the-atmosphere (TOA) direct (aerosol) and indirect (cloud) radiative forcing, respectively.

Biogenic SOA is known to have cooling effects on the Earth’s climate by scattering a portion of incoming solar radiation back into space (Tilmes et al., 2019). The changes in aerosol numbers and composition from deforestation result in net warming. We estimate a global increase in aerosol radiative forcing ($RF_{aerosol}$) of 60.4 mW m^{-2} , the tropics experiencing a particularly pronounced increase, averaging at 91.2 mW m^{-2} . The model indicates a less prominent signal in cloud radiative forcing (RF_{cloud}), with a noticeable uptick of $+27.5 \text{ mW m}^{-2}$ over the tropics, but a minor global cooling effect of -8.7 mW m^{-2} .

3.2 Grazing land restoration as a possible future reforestation scenario

In this section, we evaluate the changes attributed to a potential future afforestation scenario, assuming the restoration of all grazing land to natural vegetation. The DCGL scenario is compared with the scenario with only deforested crop cultivation (DCL), as shown in Fig. 1b. These analyses represent an extreme reforestation scenario. In DCL, the increased tree cover would

lead to an additional yearly fixation of 1.10 Pg (C) by the land biosphere. Moreover, the average global LAI would rise by $0.04 \text{ m}^2 \text{ m}^{-2}$. As a consequence, in DCL, annual isoprene and monoterpene emissions would increase by 140.47 Tg (+ 22.6%) and 11.55 Tg (+ 20.3%), respectively, compared to the present-day conditions.

260 Fig. 8 illustrates the changes in atmospheric conditions following the conversion of grazing land to reforestation (DCL – DCGL). The column mass of bSOA experiences a notable rise of over 3 mg m^{-2} across tropical South America and Central Africa (Fig. 8a). This rise leads to an elevation in the bSOA burden to 0.50 Tg (+0.10 Tg, +25%), consequently perturbing the total OA burden, which increases to 1.80 Tg (+0.10 Tg, +6%). The increase in aerosol burden perturbed the global AOT (+ 0.3%), CCN (+ 3.5%), and CDNC (+ 0.2%), as shown in Fig. 8b-d. The global aerosol direct radiative forcing is -38.2 mW
 265 m^{-2} , while the radiative forcing from changes in cloud properties is found to be insignificant.

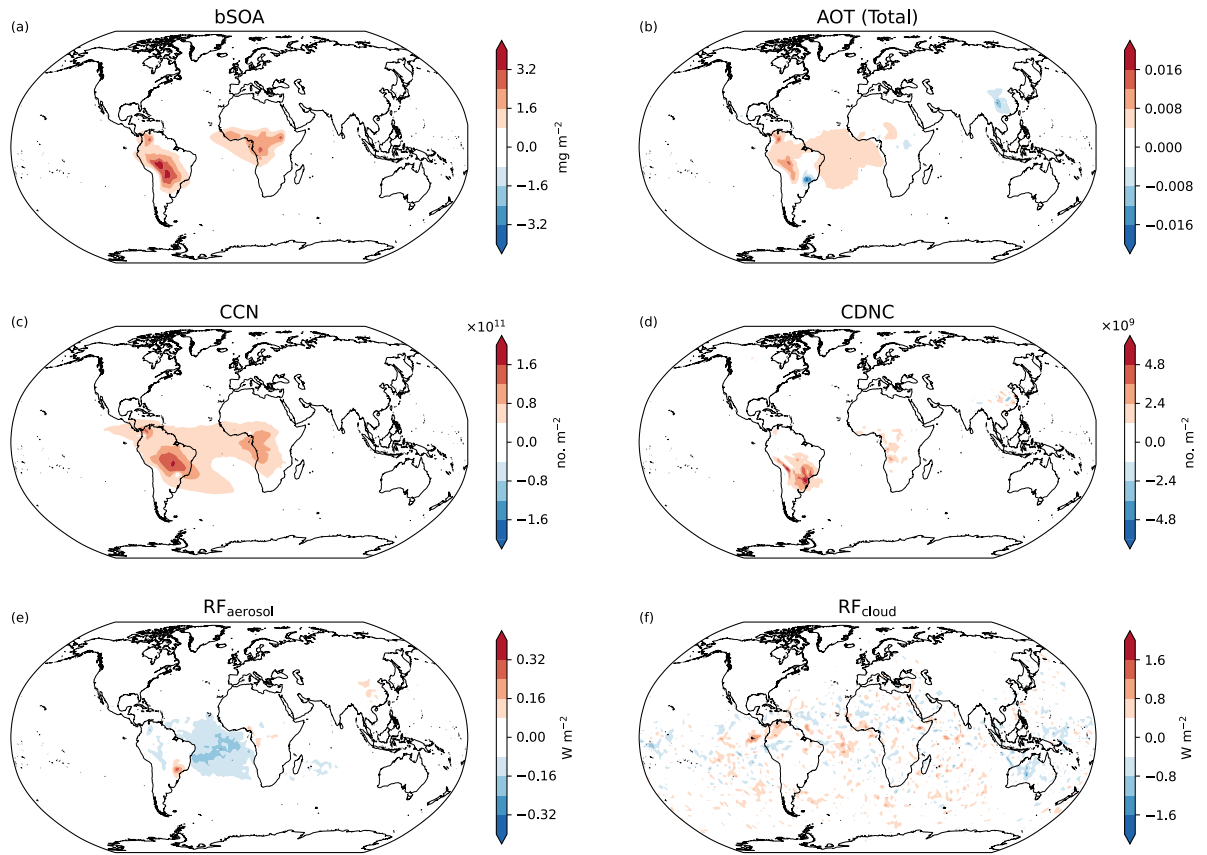


Figure 8. Changes in atmospheric states resulting from restoring present-day grazing land. Maps show variations in; (a) bSOA column mass, (b) total aerosol optical thickness (AOT), (c) cloud condensation nuclei (CCN), (d) cloud droplet number concentration (CDNC), (d) aerosol radiative forcing, and (e) cloud radiative forcing.

4 Discussion

The present study offers further insights into the ongoing impact of human-induced deforestation on potential reforestation on global carbon dynamics and atmospheric aerosols, building upon existing literature. In line with prior research documenting significant reductions in global vegetation due to human land use change, our simulations confirm a substantial decline in both net NPP and LAI resulting from deforestation. Our NPP calculations underscore the role of deforestation in diminishing the net carbon uptake of terrestrial ecosystems, thereby exacerbating the imbalance between carbon sequestration and anthropogenic emissions. Our results align with earlier findings by Haberl et al. (2007), who reported a reduction of 1.84 Pg C/yr from human land cover change. For context, the land carbon uptake averaged globally for the decade 2013–2022 amounts to 3.3 Pg (C), while anthropogenic emissions for the same period are estimated to be 9.6 Pg (C) (Friedlingstein et al., 2023).

Studies agree that land use changes predominantly affect BVOC emissions through the expansion of croplands in tropical regions such as the Amazon, central Africa, and Southeast Asia (Szogs et al., 2017). For example, Lathière et al. (2006) show a reduction of approximately 24% in isoprene emissions between 1901 and 2002. Additionally, Unger (2014) reports that land cover changes from the 1850s to the 2000s resulted in a global decrease of approximately 35% in BVOC emissions. In a comprehensive literature review exploring the influence of Land Use and Land Cover Change (LULCC) on atmospheric composition and climate, it was estimated that from the preindustrial era to the present, LULCC has led to a decrease of 15–36% in global isoprene emissions (Heald and Spracklen, 2015). Scott et al. (2018) reports a global decrease in isoprene and monoterpenes emissions of 87% and 94%, respectively, resulting from complete global deforestation. The 74% decrease in BVOC emissions reported in this study is in line with the findings reported in the literature, particularly because many studies analyse changes from preindustrial times, while our work focuses on changes from human land use compared to natural vegetation.

Several studies estimate the global mean burden bSOA at ~0.5–0.77 Tg (Henze et al., 2008; Pye et al., 2010; Hoyle et al., 2007; Tsigaridis and Kanakidou, 2007; Tilmes et al., 2019). This indicates that in the present-day deforestation scenario, ORACLE provides a lower bSOA burden (0.40 Tg) compared to literature values. Pozzer et al. (2022a) evaluated OC from ORACLE in EMAC and found that surface concentrations are reasonably well represented, however, OA is strongly underestimated in the free troposphere. The work from Pozzer et al. (2022a) employed the Mainz Organic Mechanism (MOM), which is a more complex chemical mechanism compared to the one used in this study (i.e., the Mainz Isoprene Mechanism, MIM), nevertheless, this indicates that even with a more complex mechanism EMAC still generally underestimates OC globally. Heald and Geddes (2016) reports a global annual mean tropospheric burden of bSOA to decrease by 13% due to land use change between 1850 and 2000, while Scott et al. (2018) estimates a 91% decrease in SOA from simulated global deforestation. Our model runs suggest a reduction of approximately 30% in bSOA from crop and grazing land deforestation compared to natural vegetation.

Studies focusing on changes in cloud properties due to perturbed BVOC precursors and SOA loading resulting from land use change are limited. Changes in cloud properties are often considered to evaluate the indirect radiative forcing from aerosol-cloud interactions. Scott et al. (2014) investigated the impact of bSOA on CCN and CDNC in the present-day atmosphere, and found that bSOA increases the mean annual concentration of CCN by 3.6–21.1%, and the global annual mean concentration of

CDNC by 1.9–5.2%. In this work, we show that present-day deforestation, when compared to the potential vegetation scenario, yields a decrease of 4.8% in CCN and only a marginal decrease of 0.2% in CDNC, with a reduction of 0.6% over tropical regions. Conversely, restoring all grazing land would lead to an increase in CCN of 3.5% and a slight rise of 0.2% in CDNC (+ 0.5% in the tropics). This suggests that the changes in CCN resulting from deforestation align well with findings from Scott et al. (2018). The changes in CDNC simulated by EMAC, however, seem to be very localised, in particular over the Amazon rain forest, but the global impact on CDNC is minimal. It is worth pointing out that the effect on cloud droplet numbers reported here only stems from aerosol-cloud interactions. In reality, reduced tree cover may lead to less evapotranspiration, which modifies the Bowen ratio (sensible to latent heat ratio) and potentially influences cloud forcing, however, this feedback is suppressed in our simulations.

Later on, Scott et al. (2018) went on showing that full deforestation leads to a radiative forcing of 117 mW m^{-2} attributed to aerosols and 200 mW m^{-2} attributed to clouds. However, deforestation projected for 2100 under the RCP8.5 scenario yields only 6 mW m^{-2} of radiative forcing attributed to aerosols, with negligible indirect effects from clouds. O’Donnell et al. (2011) shows a total SOA direct effect of 310 mW m^{-2} . The comparatively low direct aerosol forcing reported here from deforestation relative to the PNV scenario (60.4 mW m^{-2}) may stem from differences in methodology compared to other studies, which focus on the effects of full deforestation or total SOA (not bSOA). However, our $\text{RF}_{\text{aerosol}}$ estimates might be underestimating the actual impact due to lower bSOA budgets in our model runs.

In separate analyses assessing the statistical significance of atmospheric state changes resulting from deforestation, we found that changes in BVOC emissions and bSOA column mass exhibit statistically significant changes with 95% confidence (two-tailed Student’s t-test, i.e. $p < 0.05$). Similarly, variations in AOT attributed to OC and total aerosol also showed statistical significance. However, changes in CCN, CDNC, and cloud radiative forcing did not demonstrate statistical significance attributed to deforestation. The aerosol radiative forcing from deforestation did show statistical significance with 85% confidence, well over one sigma (68.27%) over the tropical Atlantic Ocean. While some studies, such as those employing the UK Earth System Model (UKESM) and the Community Earth System Model (CESM2), indicate statistically significant direct radiative effects from deforestation at a 95% confidence level (Weber et al., 2024). Other studies, such as the one from Unger (2014), employing the Yale-E2 global carbon–chemistry–climate model, suggest that the global-scale impacts of cropland expansion on bSOA are not statistically significant relative to interannual variability in climate. From this work, we can therefore conclude that deforestation significantly impacts biogenic aerosol budgets. However, it appears that the influence on global aerosol forcing and subsequent effects on cloud properties and radiative forcing outcomes on a global scale are likely to be relatively minor.

5 Conclusions

In this paper, we present an assessment of the atmospheric aerosol burden resulting from human deforestation based on 2015 land use data. Additionally, we explore potential large-scale reforestation scenarios by restoring all grazing land to its natural state. We use the chemistry–climate model EMAC coupled with the DGVM LPJ-GUESS, allowing for interactive BVOC emissions as well as biosphere–atmosphere feedbacks, i.e., the vegetation states respond to changes in temperature, precipitation

and surface radiation. While LPJ-GUESS simulates the PNV, a landuse scheme was implemented to constrain the tree PFT expansion based on deforestation fraction maps derived from HYDE v3.2 data.

335 Present-day deforestation results in a 73% decrease in isoprene emissions and a 75% decrease in monoterpene emissions. Consequently, the bSOA burden decreases from 0.56 Tg to 0.40 Tg, and the total OC burden decreases from 1.80 Tg to 1.70 Tg. As expected, this leads to a reduction in OC extinction, especially in the tropics near the source of bSOA and its precursors. Upon examining the impact on total aerosol extinction, the model indicates declines in AOT over South America and the tropical Atlantic Ocean. However, aerosol extinction increases over SE Asia in the present-day deforestation scenario
340 compared to PNV conditions. Particularly in SE Asia, the reduction in organic material results in the growth of water-soluble organic compounds (WASO). The absence of organics leads to more condensation on accumulation mode particles and less condensation on Aitken mode particles, resulting in increased aerosol water uptake and a shift in the aerosol size distribution towards larger particles. This effectively increases aerosol extinction.

The perturbations in aerosols resulting from deforestation lead to a positive radiative forcing in the tropics (warming),
345 stemming from the reduced aerosol burden. The increased presence of WASO compounds over SE Asia contributes to a negative radiative forcing (cooling) in this region. However, our findings indicate a global positive radiative forcing of 87.9 mW m⁻² (aerosol + cloud). In the reforestation scenario, isoprene and monoterpenes fluxes increase by 23% and 20% respectively, leading to an increase in the bSOA burden by 0.23 Tg and a negative radiative forcing of 38.2 mW m⁻².

Data availability. Will be made available on Zenodo

350 *Author contributions.* RV, HT, and MF implemented and tested the deforestation routine. RV prepared the model setup and performed the simulations with inputs from HT, AT, and AP. AT provided technical assistance for ORACLE and provided scripts to handle its output. RV evaluated the simulations, analysed the model results and wrote the article. The results were interpreted by all co-authors, with a special focus on vegetation analysis provided by MF and TH. JL and HT supervised the project. All authors discussed the results and contributed to the review and editing of the paper.

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

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3.5 BVOC-driven atmospheric aerosol burden and resulting feedbacks

This chapter offers insights into feedbacks driven by BVOC emissions. In the first part of this study, the impact of bSOA on atmospheric states is evaluated. The latter part explores the impact of bSOA on vegetation states, BVOC emissions, and subsequent bSOA yields from feedbacks. While the results regarding the impact of bSOA on atmospheric states are robust, the findings on the feedbacks are preliminary and require further investigation for definitive conclusions. However, these results are still pertinent to the overall scope and objectives of this thesis. These initial findings provide a foundation for further analysis and discussion, illustrating the direction and progress of my research thus far. I anticipate that subsequent refinements and additional data will strengthen and expand upon the insights presented here.

3.5.1 Introduction

The BVOC feedbacks studied here are discussed in Section 2.4. This work primarily focuses on analysing the influence of bSOA on atmospheric aerosol and radiation budgets. Additionally, it explores the role of bSOA in driving changes in vegetation, surface BVOC emissions, and consequent alterations in atmospheric aerosol concentrations through vegetation feedback mechanisms, as shown in Fig. 3 in Section 2.4. It is important to note that while BVOC emissions induce other indirect feedbacks by affecting methane and ozone concentrations, these are not accounted for in this study. This research provides further insight into the complex relationships between BVOC emissions, atmospheric aerosols, vegetation dynamics, and their collective impact on the Earth system.

3.5.2 Experimental design

Three model runs were conducted to investigate the aerosol burden stemming from BVOC emissions and the aerosol feedbacks with vegetation. The first experiment involves interactive BVOC emissions with activated bSOA (bSOAon), while the second experiment is identical except for deactivating bSOA in the aerosol scheme (bSOAoff). Comparing these two runs allows for an assessment of the specific impacts of the pure aerosol burden resulting from BVOC emissions. Since the second run lacks bSOA for evaluation, a third run (fixBVOC) was carried out utilising hourly BVOC emissions based on bSOAoff, but with bSOA formation activated. Hourly data was used to ensure that the diurnal cycle of BVOC emissions is preserved, incorporating both day and night emissions into the simulation. This is important for accurately capturing the full range of oxidation processes. By comparing bSOAon and fixBVOC, the changes in atmospheric aerosol resulting from vegetation-BVOC-aerosol feedbacks can be evaluated. All simulations were conducted over 10 years, with the initial 2 years excluded from the analysis. The simulation setup is summarised in Fig. 6. The vegetation initial states for all simulations were taken from a previous non-chemistry 50-year run under similar atmospheric states.

For this study, the simulations were performed in T63L31 resolution, i.e., approximately $1.9^\circ \times 1.9^\circ$ (or approx. 180×180 km at the Equator) with 31 vertical levels. Given that bVOC feedbacks are mostly driven by changes in surface temperature, the simulations employ a free atmosphere using climatological monthly average sea surface temperature (SST) and sea ice content (SIC) from the Atmospheric Model Intercomparison Project (AMIP) database from 1988 to 2007. Tracers are

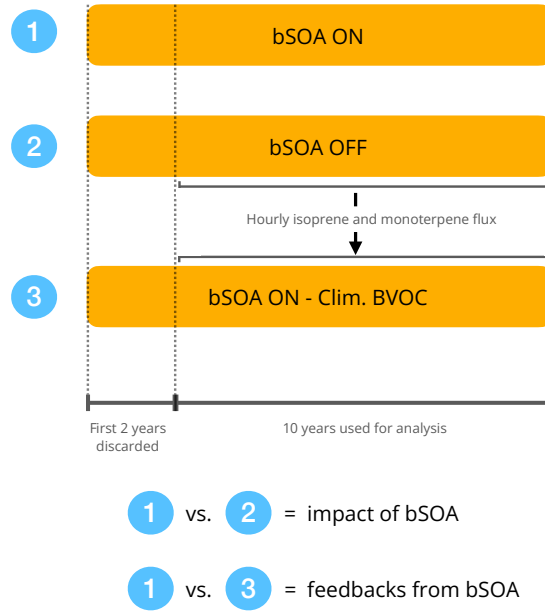


Figure 6: Experimental design. Three simulation runs: (1) baseline scenario with bSOA activated (bSOAon), (2) bSOA deactivated (bSOAoff), (3) bSOA activated but using BVOC emissions from bSOAoff (fixBVOC).

initialised based on a previous simulation by [Jöckel et al., 2016] for the year 2000, while the CO_2 concentrations in the radiation and vegetation schemes were kept fixed at 370 ppmv, representing the year 2000.

3.5.3 Preliminary results, discussion, and limitations

The impact of bSOA on atmospheric states

This section discusses the impact of bSOA on atmospheric states by comparing simulations 1 and 2 (Fig. 6). Fig. 7 illustrates the changes in aerosol optical thickness (AOT), cloud condensation nuclei (CCN), cloud droplet number concentration (CDNC), and radiative forcing (RF) resulting from the presence of bSOA (i.e., bSOAon – bSOAoff). The model runs indicate that bSOA increases the global organic carbon (OC) AOT by 13.1% (Fig. 7a), which consequently leads to a 1.3% increase in the total aerosol AOT (Fig. 7b). As shown in Fig. 7, the changes mostly occur in the tropics where elevated BVOC emissions from rainforests are found.

The presence of bSOA also increases the CCN count over the tropics, resulting in a global increase of 17.2% (Fig. 7c). This increase in CCN affects cloud properties, leading to a global rise in CDNC of 1.1% (Fig. 7d). Additionally, Fig. 7e and Fig. 7f illustrate the changes in direct aerosol radiative forcing ($\text{RF}_{\text{aerosol}}$) and cloud radiative forcing (RF_{cloud}) due to bSOA, respectively. The global $\text{RF}_{\text{aerosol}}$ from bSOA is estimated to be -145.2 mW m^{-2} , indicating a cooling effect, while the impact on RF_{cloud} appears to be insignificant.

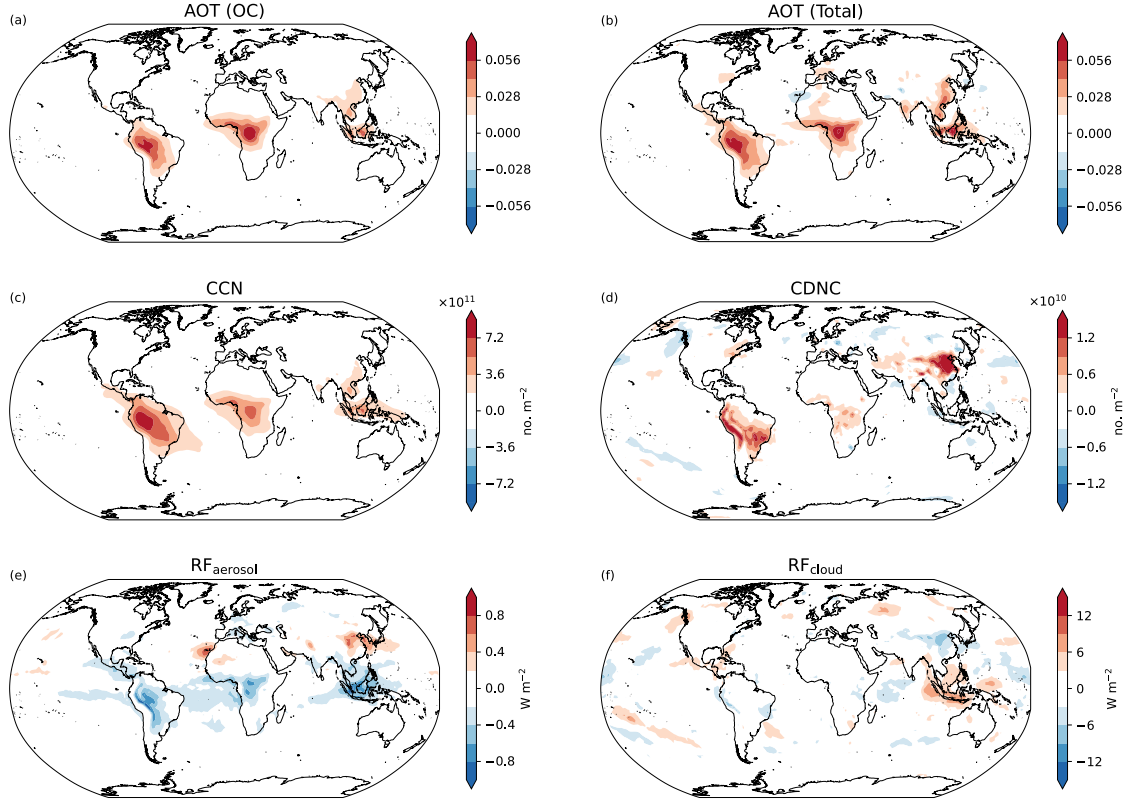


Figure 7: Changes in atmospheric states resulting from bSOA (bSOAon – bSOAoff). Maps show variations in; (a) aerosol optical thickness (AOT) from organic carbon, (b) AOT from total aerosol, (c) cloud condensation nuclei (CCN), (d) cloud droplet number concentration (CDNC), (e) aerosol radiative forcing, and (f) cloud radiative forcing.

BVOC-aerosol feedbacks

In this section, the influence of bSOA on vegetation states and BVOC surface fluxes is explored. This analysis aims to shed light on BVOC feedbacks, specifically how changes in atmospheric states due to bSOA formation propagate back to the biosphere and influence BVOC emission fluxes. Given that LPJ-GUESS is not sensitive to diffuse radiation, this section focuses solely on feedbacks through changes in direct above-canopy shortwave radiation and temperature effects, which are influenced by aerosol scattering and changes in cloud properties.

Fig. 8 shows changes in the NPP and LAI, BVOC emissions and resulting bSOA from feedbacks. The changes in NPP, LAI, isoprene, and monoterpene emissions are derived from simulations 1 and 2 in Fig. 6 (bSOAon – bSOAoff), while the consequent bSOA changes are obtained from simulations 1 and 3 in Fig. 6 (bSOAon – bSOAfixed). The changes in vegetation states appear to be small, with opposing effects observed in different regions. It is estimated that both NPP and LAI decrease by approximately 1% when bSOA is activated in the model. Similarly, for the subsequent BVOC emissions, the results indicate a global decrease in yearly isoprene and monoterpene emissions of less than 1% resulting from bSOA.

As depicted in Fig. 8e, the changes in the bSOA load emerging from feedbacks are small, with

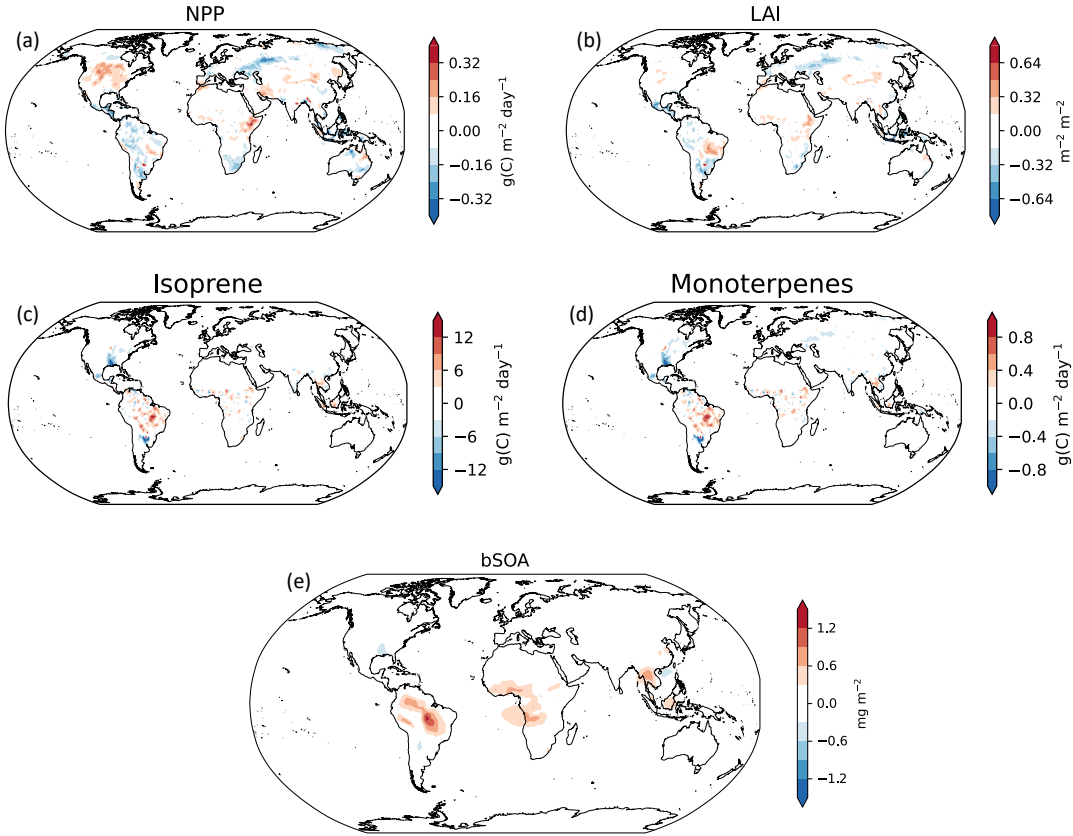


Figure 8: Changes resulting from bSOA. (a) net primary production (NPP), (b) leaf area index (LAI), (c) isoprene emissions, (d) monoterpene emissions, (e) resulting bSOA changes from feedbacks.

an increase of about 1 mg m^{-2} observed over some parts of the Amazon and Congo basins. These results suggest that the BVOC feedbacks via aerosol burden are very small and likely to fall within the internal variability of the model. However, further analysis is needed to explore these changes over seasonal cycles. Other limitations that might contribute to the low signal from feedbacks may be the fact that the simulations presented here run for only 12 years with a free atmosphere (i.e., no nudging), potentially requiring more years to be simulated to obtain a statistically robust result. Additionally, this setup employs a simplified chemistry mechanism (Mainz Isoprene Mechanism (MIM)). It would be possible to run this setup with a more complex chemical mechanism (Mainz Organic Mechanism, (MOM)) in EMAC, which resolves the oxidation paths of some BVOC individually. This could have an impact on the bSOA yields and thus the significance of such feedbacks. Nevertheless, such simulations would be very computationally expensive.

4 Overall conclusions and outlook

The work presented in this thesis improves the modelling capabilities of the chemistry-climate model EMAC, taking it a step closer to being a fully coupled earth system model (ESM). The development of ESMs allows for a far more detailed analysis of a fully coupled and dynamic system addressing many complex biosphere-atmosphere interactions governed by BVOC emissions and thus shedding more light on the significance of such processes on global climate change and air quality.

The results from Section 3.1 show the development towards producing a new atmospheric-chemistry-enabled ESM by coupling an atmospheric-chemistry-enabled atmosphere-ocean general circulation model (AOGCM) with a dynamic global vegetation model (DGVM). This work also explores, for the first time, partial bidirectional interactions between the two modelling systems via BVOC emissions, building on previous technical developments by [Forrest et al. \[2020\]](#) that enabled one-way coupled simulations (in which climate information generated by EMAC is used to force LPJ-GUESS, but no land surface information is relayed back to EMAC). The updated model version described in this work allows computations of isoprene and monoterpene emissions based on dynamic vegetation states running on the EMAC time step. This is a substantial improvement compared to the earlier set-up in which BVOC emissions were only based on offline vegetation information and not coupled with dynamic vegetation states. Global isoprene emission estimates from the coupled model configuration, using both ONEMIS and MEGAN, provide realistic global variability, corresponding to emissions with prescribed vegetation boundary conditions. Sensitivity runs with doubling CO₂ scenarios showed that BVOC fluxes are sensitive to temperature increases from BVOC routines (ONEMIS and MEGAN) and changes in plant productivity simulated by LPJ-GUESS.

Section 3.2 highlights the complex interactions among atmospheric, oceanic, and vegetation processes during ENSO events. Despite the numerous studies focusing on the effects of ENSO perturbation on the biosphere, only a limited number of studies have specifically investigated the sensitivity of BVOC emissions [e.g., [Lathière et al., 2006](#), [Müller et al., 2008](#), [Naik et al., 2004](#), [Pfannerstill et al., 2018](#), [Wells et al., 2020](#)]. These studies either use observations or historical transient simulations. While this approach employs realistic scenarios, it is difficult to assess the impact of ENSO alone given the multiple types of climate forcing, e.g., from anthropogenic emissions of CO₂ and aerosol. This study evaluates the changes in atmospheric and vegetation states from ENSO and links these changes to BVOC emissions. It is found that a very strong El Niño event increases global isoprene emission fluxes by 2.9% and that an isolated ENSO event perturbs BVOC emission fluxes so profoundly that they do not return to baseline emissions even several years after the event. More focus is given to the sustained ENSO simulations, where ENSO conditions from a single event are simulated continuously over several years. This approach offers the advantage of reducing internal variability, thereby enhancing the robustness of statistical analyses when correlating atmospheric and vegetation variables with BVOC flux anomalies. It was found that persistent ENSO conditions shift the vegetation productivity and structure (LAI) into a new quasi-equilibrium state, whereby changes in plant functional types (PFT) composition are more transient. Over South East Asia, the fractional coverage of tropical broad-leaved evergreen trees could decrease by 8.6% and increase by 4.9% during persistent El Niño and La Niña scenarios,

respectively. It was also shown that if ENSO, particularly El Niño, becomes more persistent in the future we should expect amplification in BVOC flux changes as we enter this new climatic mode with long-term influence on the biosphere. In this climate mode, isoprene emissions from the Amazon could increase by 19%. The study also links BVOC emission changes with changes in the driving variables (i.e., temperature, light, and vegetation dynamics) during ENSO. It was shown that ENSO-induced variations in BVOC emissions over the tropics are largely related to surface temperature anomalies, whereas surface radiation predominates at higher latitudes. Variations in the LAI were strongly linked to anomalies in BVOC emissions over the subtropics, indicating that ENSO-induced changes in plant phenology are an important driver of BVOC emissions.

The work following the above focuses more on how changes in BVOC emissions propagate to the Earth system via the atmosphere. BVOCs influence atmospheric chemistry with global implications for the atmospheric oxidation capacity and the lifetimes of key gaseous species, including O_3 , CH_4 , and NO_x . However, this work focuses on the link between BVOCs and aerosols.

Section 3.4 presents an assessment of the atmospheric aerosol burden resulting from human deforestation, based on 2015 land use data. The current version of LPJ-GUESS in EMAC simulates potential natural vegetation (PNV). To simulate land cover scenarios, a land use scheme was implemented to constrain the Tree Plant Functional Type (PFT) cover, using deforestation fraction maps from the year 2015. The PNV scenario is compared with (1) present-day deforestation, based on the deforestation of both crop and grazing land cover, and (2) an extreme reforestation scenario assuming that all grazing land is restored to natural vegetation (i.e., with only cropland deforestation). The study finds that, compared to the PNV scenario, present-day deforestation results in a 26% reduction in BVOC emissions. These estimates align with the findings of [Lathière et al. \[2006\]](#) and [Unger \[2014\]](#), which also report changes in BVOC emissions resulting from deforestation.

The findings of this thesis suggest that the transition from natural vegetation to present-day land use has led to a reduction in BVOC emissions, resulting in a global decrease in bSOA burden by 0.17 Tg (equivalent to a 29% reduction) and a total organic carbon (OC) reduction of 0.16 Tg (a 9% decrease). Under the present-day deforestation scenario, the total radiative effect (aerosol + cloud) is estimated to be a positive 87.9 mW m^{-2} , indicating warming compared to the natural vegetation scenario. Conversely, the extreme reforestation scenario demonstrates a negative forcing (cooling) of 38.2 mW m^{-2} compared to the scenario with primary natural vegetation. The extreme reforestation scenario indicates an increase in BVOC emissions by 22%, which increases the bSOA and total OC burden by 0.10 Tg, an increase of 25% and 6%, respectively, compared to present-day land use cover. However, it appears that the influence on global aerosol forcing and subsequent effects on cloud properties and radiative forcing outcomes on a global scale are relatively minor.

This setup was also used to investigate biosphere-atmosphere feedbacks via BVOC emissions (Section 3.5). Changes in global aerosol burdens from BVOCs modify atmospheric states. These changes manifest through the interactions of aerosols with surface radiation and cloud properties, resulting in alterations in surface temperatures and potential changes in precipitation patterns. These shifts influence vegetation and BVOC emission fluxes, setting off feedback loops [[Kulmala et al., 2004, 2013](#), [Rap et al., 2018](#), [Sporre et al., 2019](#)]. BVOCs are involved in other feedbacks by affecting methane and ozone concentrations, though these feedbacks are not accounted for in this work. Results presented in Section 3.5 show that BVOC emissions increase the global organic carbon and the total aerosol AOT by 13%. However, the model runs from this setup suggest that

these perturbations in the atmospheric states do not significantly impact vegetation and BVOC emissions. Consequently, it is concluded that the changes in BVOC emissions resulting from bSOA are small and fall within the internal variability of the model.

Outlook

This work improved the representation of BVOCs in EMAC, providing valuable insights into BVOC impacts on the Earth system and their interactions with the biosphere. Moving forward, there are exciting opportunities to refine the coupling between LPJ-GUESS and EMAC, which would enable more precise investigations and address emerging research questions with greater accuracy.

While the current configuration utilised a simplified deforestation scheme within LPJ-GUESS (v4.0) (implemented in this work), the adoption of the latest model version (v4.1) offers a more sophisticated land cover representation, enhancing the realism of studies involving future or historical land cover scenarios.

Moreover, recent advancements in LPJ-GUESS, such as the incorporation of sensitivity to diffuse radiation [[Martín Belda et al., 2022](#)], and a more sophisticated version of MEGAN, present promising avenues for improvement. Implementing these changes would not only enhance the accuracy of studies but also broaden the scope of analysis by capturing finer-scale processes and interactions within the biosphere, such as BVOC-aerosol feedbacks via the diffuse radiation fertilisation effect as proposed by [Rap et al. \[2018\]](#).

The current EMAC/LPJ-GUESS configuration includes feedbacks in the hydrological cycle, although this aspect was not explored in this work. Consequently, this setup presents an opportunity to investigate whether observed changes in aerosols resulting from vegetation changes primarily arise from aerosol-cloud interactions or shifts in evapotranspiration.

Additionally, integrating new parameterisations for aerosol-cloud interactions in convective warm clouds, for example, as suggested by [Miltenberger et al. \[2018\]](#), holds the potential to significantly enhance the accuracy and relevance of findings, especially in regions dominated by convective cloud systems where BVOC emissions are prominent.

Looking ahead, further analyses utilising the configured setup, as discussed in Section 3.4 but focusing on changes in the lifetime of key species such as O_3 , CH_4 , and NO_x , as well as changes in the oxidation capacity will deepen our understanding of biosphere-atmosphere interactions using the EMAC model. Moreover, evaluating the combined effects of LPJ-GUESS and the newly implemented land surface model JSBACH [[Martin et al., 2024](#)] within EMAC offers a unique opportunity to assess BVOC emissions and sensitivities across different temporal scales, providing a more comprehensive perspective on biosphere-atmosphere dynamics.

5 Supplement

5.1 Supplement of Section 3.3

This chapter provides the Supplement of *Changes in biogenic volatile organic compound emissions in response to the El Niño–Southern Oscillation*, available at:

<https://doi.org/10.5194/bg-20-4391-2023-supplement>

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Supplement of

Changes in biogenic volatile organic compound emissions in response to the El Niño–Southern Oscillation

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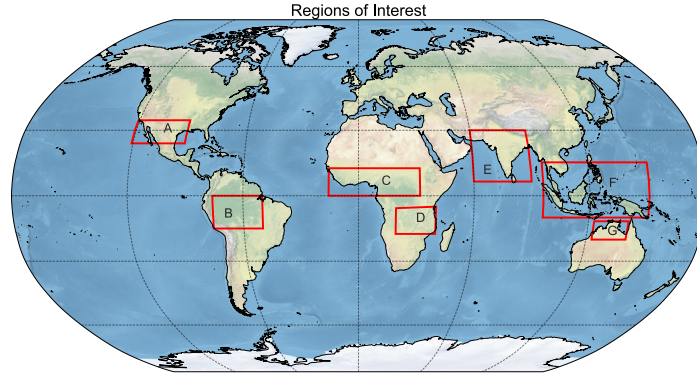


Figure S1. Regions of interest. The extent of areas is presented in Table 1.

Region	Label on figures	Acronym	Latitude range	Longitude range
South West USA	A	SWUSA	24, 34.6	−120.8, −92.4
Amazon Basin	B	Amazon	−15, 0	−75.8, −50
Central West Africa	C	CEAfr	0, 12.6	−15.54, 32
South East Africa	D	SEAfr	−14.6, −5.7	19.5, 40.3
India	E	India	6.5, 30	60, 90
South East Asia	F	SEAsia	−10, 15.3	96, 151
North Australia.	G	NAus	−20.1, −11.7	123, 141

Table S1. Details on the regions considered.

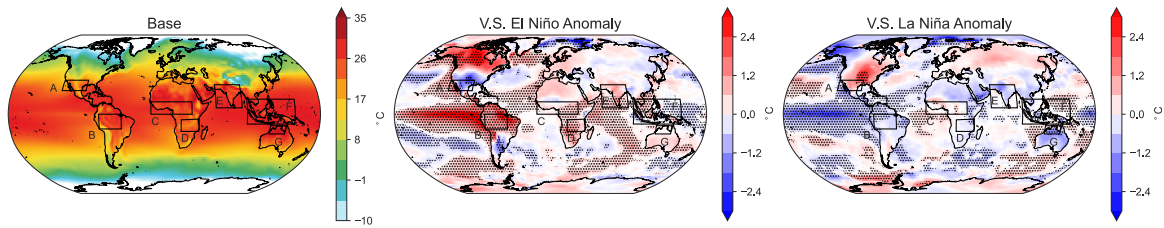


Figure S2. Same plot for surface temperature as in Fig. 4 but also showing anomalies over the ocean. This confirms that our model captures well the ENSO scenarios.

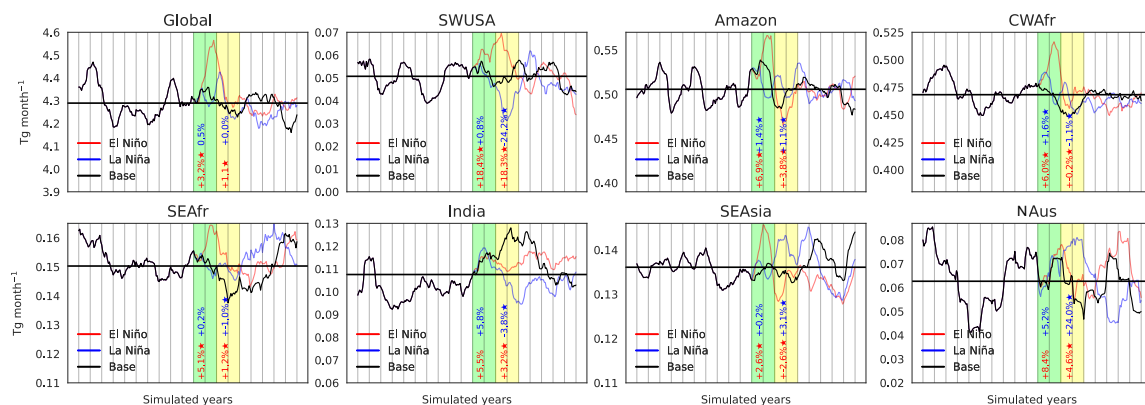


Figure S3. Time evolution of global and regional monoterpene fluxes with El Niño (red) and La Niña (blue) events over two years (green columns). The black line illustrates base conditions throughout all simulations. Statistically significant changes ($p < 0.01$) are marked with a star.

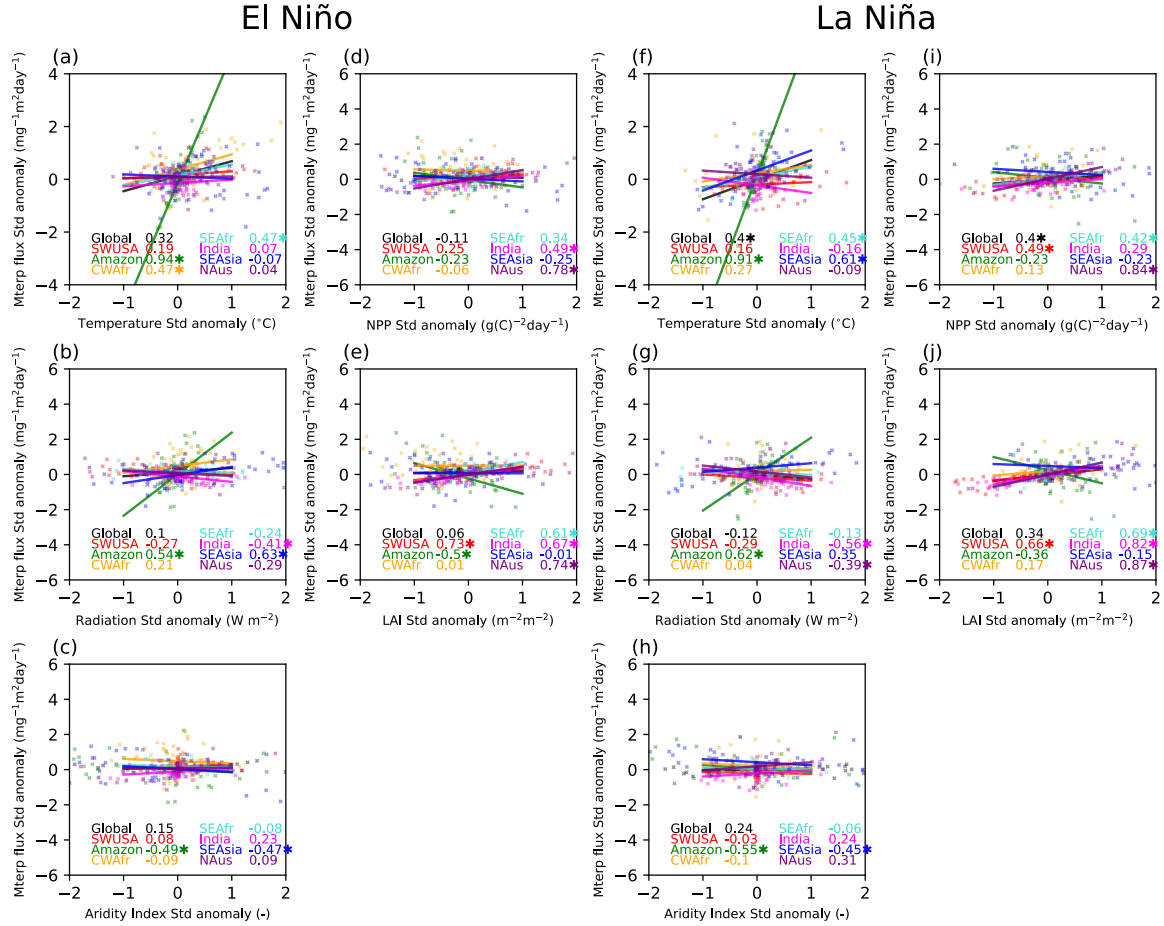


Figure S4. Scatter plots with a linear regression fit including the Pearson's correlation (r) between the standardized monoterpene flux anomaly and standardized temperature, radiation, AI, NPP, and LAI anomaly at different regions for the event years and the following two years (four years in total). Very strong El Niño and La Niña events are shown in the top and bottom panels, respectively.

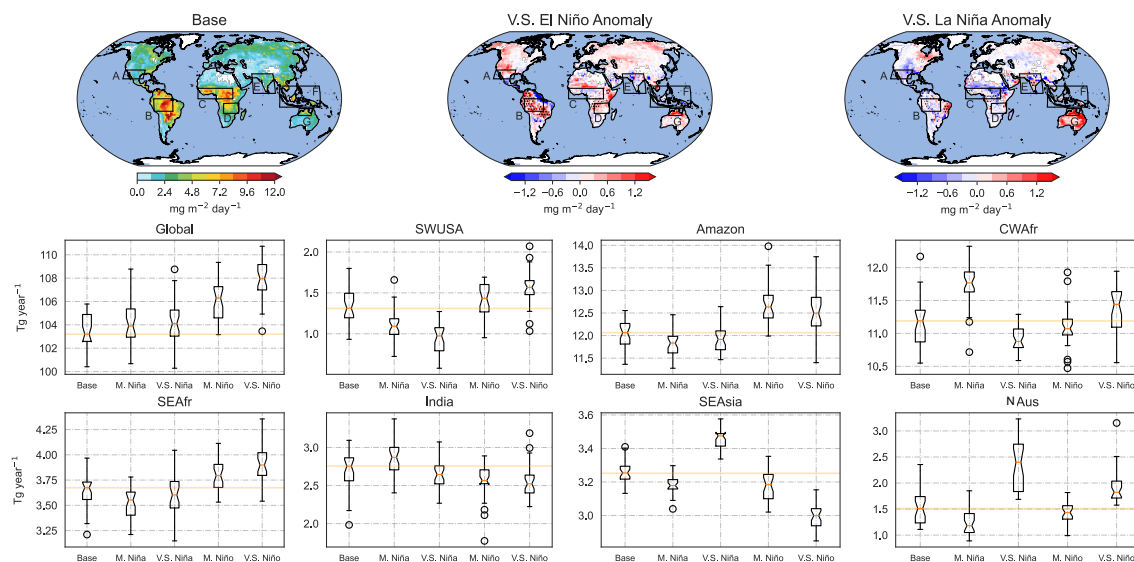


Figure S5. Global monoterpene fluxes for the base scenarios as well as changes in emissions during very strong El Niño and La Niña events. The bar plots show emission changes for the different scenarios. The median of the base scenario is indicated in orange.

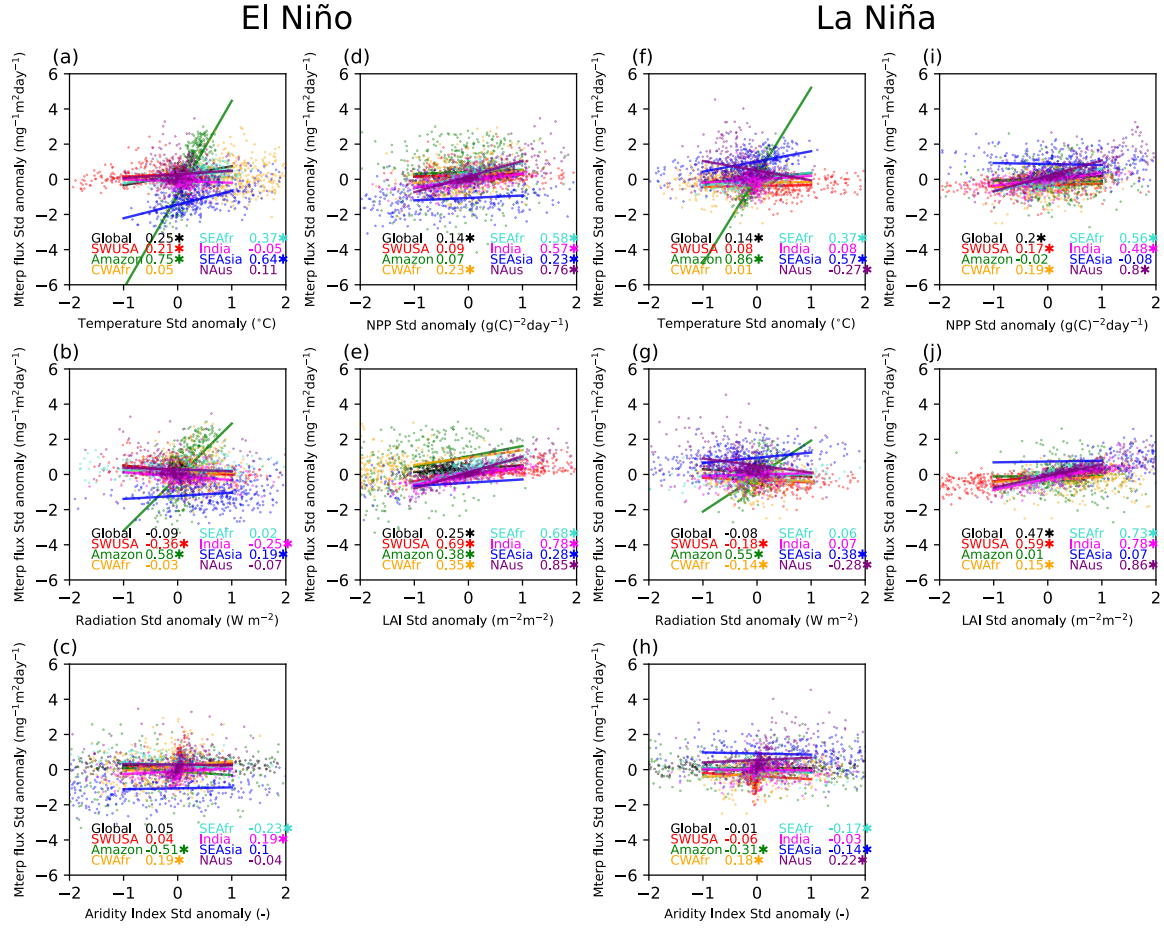


Figure S6. Scatter plots with a linear regression fit including the Pearson's correlation (r) between the standardized monoterpene flux anomaly and standardized temperature, radiation, AI, NPP, and LAI anomaly at different regions for very strong El Niño (top panels) and La Niña (bottom panels) events. Correlations with $p < 0.01$ are marked with a star.

Main variables driving monoterpene emissions during El Niño

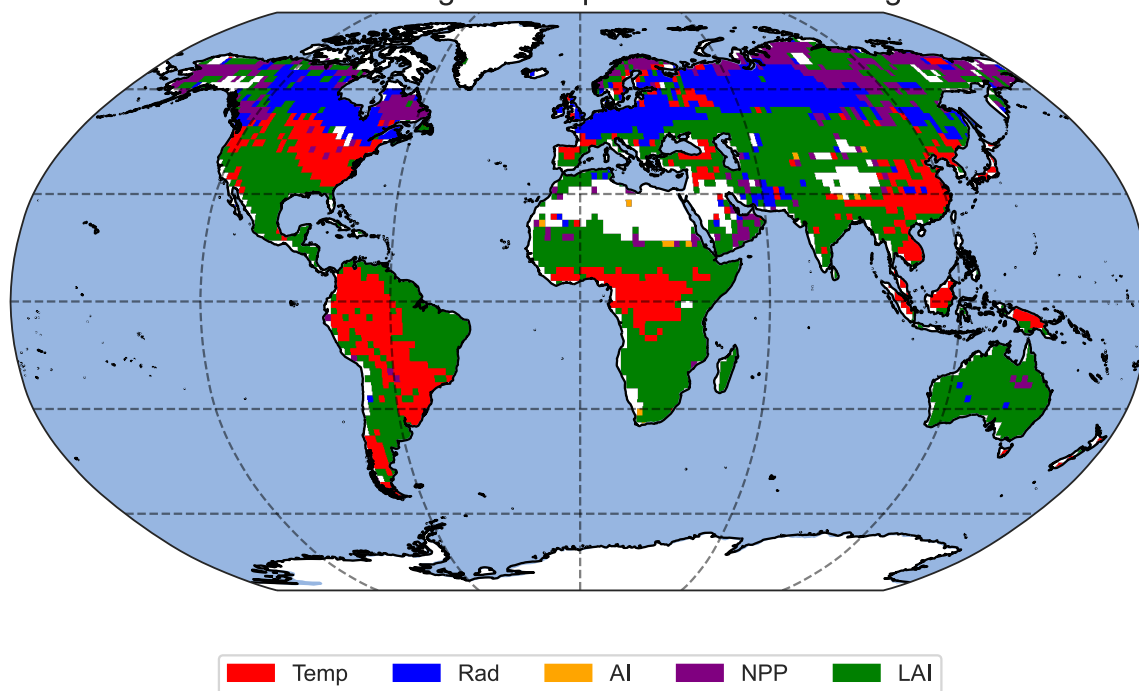


Figure S7. Map showing the variables whose anomalies correlate most strongly with the monoterpene flux change for every pixel of the model output. The correlations between the monoterpene flux and the temperature, radiation, AI, NPP, and LAI anomalies are classified using PCA.

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