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Climate Feedback on Aerosol Emission and Atmospheric Concentrations

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Abstract

Purpose of Review Climate factors may considerably impact on natural aerosol emissions and atmospheric distributions. The interdependencies of processes within the aerosol-climate system may thus cause climate feedbacks that need to be understood. Recent findings on various major climate impacts on aerosol distributions are summarized in this review.

Recent Findings While generally atmospheric aerosol distributions are influenced by changes in precipitation, atmospheric mixing, and ventilation due to circulation changes, emissions from natural aerosol sources strongly depend on climate factors like wind speed, temperature, and vegetation. Aerosol sources affected by climate are desert sources of mineral dust, marine aerosol sources, and vegetation sources of biomass burning aerosol and biogenic volatile organic gases that are precursors for secondary aerosol formation. Different climate impacts on aerosol distributions may offset each other.

Summary In regions where anthropogenic aerosol loads decrease, the impacts of climate on natural aerosol variabilities will increase. Detailed knowledge of processes controlling aerosol concentrations is required for credible future projections of aerosol distributions.

Keywords Natural aerosols · Atmospheric chemistry · Biogeochemical cycles · Climate forcing · Climate change

Introduction

Atmospheric aerosols are recognized as important climate factors. Much attention has been focused on the various direct and indirect effects of aerosol particles on the Earth's radiation balance and ecosystems. Such impacts—including the scattering and absorption of solar radiation, change of cloud properties, and modifications of atmospheric chemical reactions, as well as the role of aerosol particles in delivering nutrients to ecosystems—have been topics of many studies that are regularly summarized in the reports of the Intergovernmental Panel of Climate Change [1]. However, in addition to the aerosol effects on climate, climate variables in turn also influence processes that control atmospheric aerosol distributions, including emissions, transport, transformation, and deposition of aerosol particles. The various impacts of climate on aerosol

distributions were summarized in several reviews in recent years [2–4]. While those reviews were mainly motivated by surveying the role of climate variability on air quality changes and concentrations of cloud condensation nuclei (CCN), variability and trends in aerosol optical depth (AOD) ([5], Fig. 1a) are also an important factor for studies of the role of aerosol articles in climate [7].

Aerosols consist of mixtures of different particle species from specific natural and anthropogenic sources (e.g., primary particles like sea salt, mineral dust, biological particles, and soot; particles secondarily formed from precursor gases like ammonium sulfate, ammonium nitrate, and secondary organic aerosols (SOA)) (e.g., [8]). The high variability of the aerosol mixture in space and time makes quantification of global distributions of the different aerosol species difficult. Furthermore, understanding and disentangling the interactions of the different aerosol types and climate variables is very challenging.

Anthropogenic aerosols like sulfates and carbonaceous aerosol particles originating from burning of fossil fuels have received considerable attention as climate forcing factors, but variability and trends of aerosols originating from natural sources like mineral dust emitted from desert surfaces, marine aerosol emitted from oceans, and carbonaceous aerosols from wildfires must also be understood to correctly assess the


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Fig. 1 **a** Average aerosol optical
depth from the Monitoring
Atmospheric Composition and
Climate (MACC) reanalysis for
the year 2010 [5]. **b** Aerosol
distribution from different source
types. Aerosol fractions of the
total AOD from different source
types were derived from the
global aerosol-climate model
ECHAM-HAM (Hamburg
Aerosol Model) [6] and applied to
the MACC AOD distribution.
The AODs for individual aerosol
types are normalized to the
respective maximum AOD
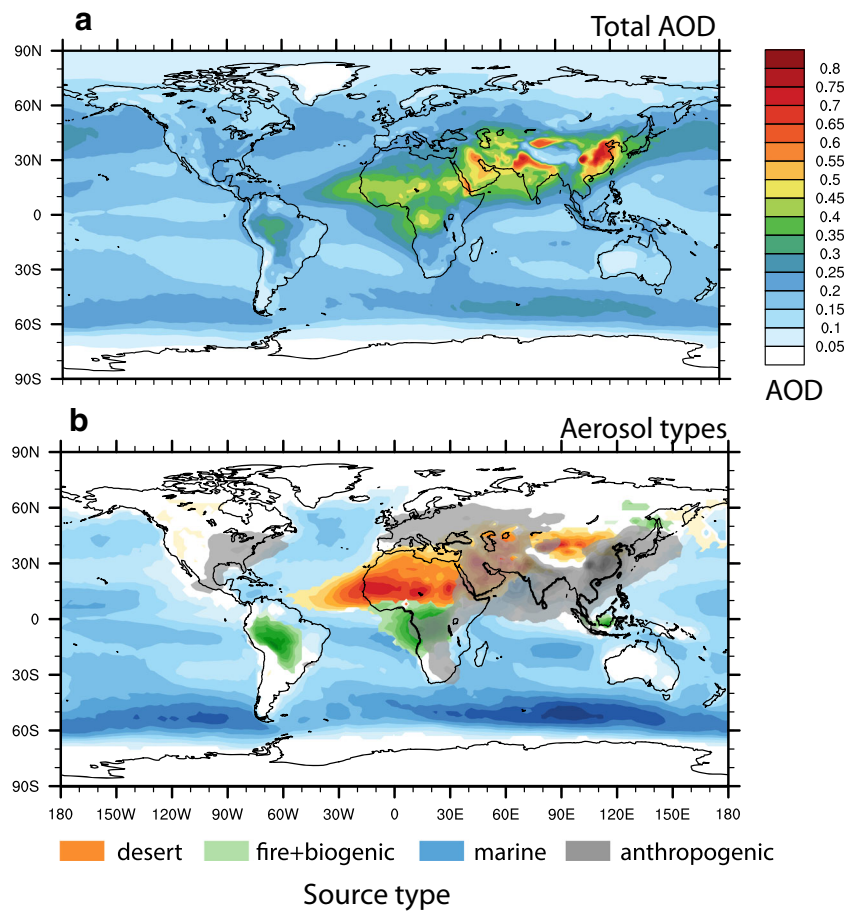
impact of anthropogenic aerosols within the climate system. The normalized regional distributions of aerosol AOD from different source types are indicated in Fig. 1b, where the aerosol source contributions were derived from the global aerosol-climate model ECHAM-HAM (Hamburg Aerosol Model) [6]. The shown individual aerosol contributions are normalized to their respective global maximum to highlight regions where these aerosol types are most prevalent. Anthropogenic aerosol emissions are changing in response to economic developments and environmental policies. In the past decades, anthropogenic aerosol concentrations have strongly increased in eastern Asia, but decreased in Europe and North America since the mid-1980s [7]. Not only fossil fuels but also changes in land use and agricultural emissions contribute to aerosol trends. While the anthropogenic emission changes mask possible climate effects influencing aerosol concentrations, in other regions like North Africa and Middle East, aerosol variations can be attributed to meteorological modifications of natural aerosol emissions [9].

The variability of aerosols from natural sources will become increasingly relevant for the aerosol mixture in regions where anthropogenic aerosol concentrations decrease. Evidence of possible significant changes in natural aerosol concentrations in changing climates is found in ice core and marine sediment records. Particularly dust concentrations have changed by as much as an order of magnitude between glacial and interglacial climate periods (e.g., [10]). Such variations may also be expected in a warming climate, but to assess the magnitudes or even the direction of the related changes with any confidence, the climate controls on aerosol processes must be understood in detail.

Processes describing emission and thus entrainment into the atmosphere, deposition processes, and atmospheric transformation rates determine the atmospheric concentrations and lifetimes of the different aerosol species. Atmospheric transformations include atmospheric chemical reactions and microphysical processes like secondary particle formation and growth. In turn, these processes depend to various degrees on meteorology and climate. Aerosol particles typically reside in the atmosphere few hours to about 2 weeks [11]. Changes in dust atmospheric residence time, e.g., due to wet removal processes or atmospheric static stability, will modify the distributions and concentrations of aerosol particles. Aerosol concentrations are a commonly used parameter for assessing and representing the variability of their life cycles.

The interdependencies of aerosols and climate can result in feedbacks between aerosol concentrations, aerosol effects, and climate variables. An example for such aerosol-climate

interactions is the suspected role of deposited dust in providing iron as micronutrient to the ocean ecosystem and thus affecting ocean biogeochemistry. A possible increase in plankton productivity by increased dust deposition may increase the CO_2 uptake by the ocean, causing feedback effects on climate and ultimately dust production [12].

While the individual aerosol species are affected by transport, mixing, and deposition processes in similar ways, climate controls on natural aerosol sources vary considerably. Emissions of anthropogenic aerosol and aerosol precursors as well as volcanic aerosol emissions are unaffected by climate. In contrast, natural aerosol emissions may be directly influenced by atmospheric variables like wind speed and temperatures, or indirectly by climatic controls on source efficiencies, for example vegetation biomass or sediment availability. Typically, these dependencies are non-linear. The various main climate impacts on the major natural aerosol sources are described in the following section.

Climate Impacts on Aerosol Sources and Emission

Mineral Dust

Dust emission is determined by both surface characteristics and the state of the atmosphere: surface characteristics describe the supply and availability of sediments prone to wind erosion. The state of the atmosphere, in particular near-surface wind speeds and static stability, determines the forces acting on erodible soil elements necessary to mobilize and uplift dust particles, the so-called entrainment, and the capacity of the atmosphere to transport entrained dust particles off the source regions. Dust emission fluxes increase with wind speed, whereas no dust is mobilized for wind speeds below a local threshold, which is determined by the soil properties [13]. Thereby, knowledge on the surface characteristics is essential in order to determine spatial and temporal variabilities in dust emission fluxes. Recent studies illustrate the relevance of alluvial sediment deposits acting as dust source and emphasize the potential of this source type to modulate the interannual variability in atmospheric dust loading due to interannually changing sediment variability [14, 15]. Furthermore, alluvial dust sources potentially link the atmospheric dust life cycle to the atmospheric water cycle as precipitation drives the formation of fresh alluvial deposits. This way, variability in atmospheric dust burden is not only determined by atmospheric circulation [16–18], but also by sediment supply and availability.

Dust concentrations are a commonly used parameter for assessing and representing the variability of the dust life cycle. Impacts on one element of the dust life cycle consequently will alter atmospheric concentrations. A changing climate results in a changing environment, which has diverse influences on the nature of the dust life cycle in general as well as on individual elements

of the life cycle in particular. Changes in the current climate are commonly expressed as the energy available or conserved in the Earth system. Increases, respectively decreases, in temperature and atmospheric water vapor content or precipitable water are prominent examples. Although there is no direct impact of air temperature on dust entrainment and dispersion, the global temperature distribution stimulates global atmospheric circulation patterns, which for their part may affect local conditions enhancing or reducing processes characterizing the dust life cycle.

The atmosphere over the Sahara Desert, the world's largest dust source region in terms of both geographical extent and annual dust emission flux, is warming faster than the mean warming rate over the tropics. This amplification prompts to changes in atmospheric circulation affecting soil characteristics and atmospheric circulation ultimately resulting into changed dust emission fluxes and transport routes [19]. An amplified surface warming affects both summer and winter circulation regimes over the Sahara: during summer, it strengthens the Saharan heat low and the African easterly jet; during winter, it weakens the sub-tropical anticyclone and the Harmattan winds [20]. These results suggest that the amplified warming over the North African desert leads to a decrease in global atmospheric mineral dust loading.

Hoffman et al. [21] illustrate that global atmospheric teleconnection patterns controlled by sea surface temperature (SST) patterns drive variability in dust emissions. Processes and conditions controlling dust emission are modified by these teleconnection responses. In particular, via teleconnection patterns, SST anomalies alter near-surface atmospheric circulation over source regions and thus modify wind speed distribution, precipitation, vegetation, and soil moisture, which to their part affect dust emission, transport routes, and deposition fluxes. For the six examined dust source regions in northern and southern Africa, and Australia, the response is strongest to Indian Ocean (Indian Ocean Dipole, IOD) and Pacific (El Niño Southern Oscillation, ENSO) SST anomalies. Results from Hoffman et al. [21] suggest that with changing teleconnection patters, e.g., enhanced ENSO frequency and IOD or even superimposed teleconnections, dust source activity will change and consequently dust concentrations in the atmosphere will change as well. Tong et al. [22] analyzed a multi-year record of dust storm activity reports and found an increase in dust storm activity over the American deserts during 1988–2011. This positive trend correlates with the Pacific Decadal Oscillation (PDO), which was in a cold phase during the referring decade and promoted anomalously dry and strong wind conditions over the North American deserts. However, a quantification of changes in dust emission flux and dust concentration is still missing.

The local wind speed distribution is a predominant control on local dust emission fluxes and transport pathways. Consequently, changes in wind speeds affect regional dust emission fluxes in particular and global dust distribution in

general [23]. Weaker winds result into lower dust emission fluxes and consequently lower dust concentrations in the atmosphere.

The nature of the atmospheric dust life cycle and thus the atmospheric dust load is determined by both surface characteristics and the state of the atmosphere. Thereby, the variability of sediment supply and availability, and atmospheric transport capacity in concert stimulate the variability in dust aerosol concentrations. The complexity of interactions among the impacting states and processes suggest that assessments on dust variability require a comprehensive view in order to identify first-order determinants and balancing feedbacks.

Biomass Burning Aerosol

Burning biomass (fires) emit various particulate and gaseous aerosols. Fire activity is part of different land ecosystems such as savannas, temperate and boreal forest, and agricultural regions. Prerequisites for fires are the supply and availability of fuel (biomass) and ignition, which can be natural or human-driven. An example is the strong change in boreal fires in recent decades [24].

The influence of climate on fires is strong but complex for the following reasons: it determines the primary productivity and vegetation and hence the supply and availability of bio-fuel, which is the major control for biomass burning aerosol emission ([25] and reference therein). The availability and supply of bio-fuel is additionally controlled by human activity, in particular agricultural practice, land management, and fire prevention [26]. Thus, climate-related controls on fire activity are beyond the supply/availability paradigm. Shifts in vegetation zones and predominant vegetation type (biomes) are natural consequences of a changing climate and thus an intrinsically driven factor impacting fire emissions. Following the fuel amount versus fuel moisture limitation paradigm, in a climate less favorable for plant growth like arid regions, low primary productivity will limit fuel supply and fires. In climate regions favorable for plant growth and thus with high primary productivity, unfavorable (wet) weather conditions will limit fire ignition and spread due to moisture. In the context of climate change, the occurrence of antecedent drought periods contributes to an increase fire activity. However, regions encompassing a variety of vegetation types may respond differently to climate change. The same is evident for areas which fire activity is controlled by more than one mechanism.

Changes in predominant weather regimes and thus precipitation rates have consequences on vegetation type and variety, which feedbacks on fire intensity and frequency. For future climate scenarios, Veira et al. [27] suggest fuel availability to be the major driver for changes in emissions in extra-tropical regions. Thus, the authors suggest a general increase in wildfire activity in the extra-tropics. In tropical regions, a general decrease in fire activity is expected due to land use change and a strong decrease in human-driven ignitions.

The injection height affects the transport route and thus the distribution within the atmosphere. It is controlled by the fire intensity, which depends on the type and supply of bio-fuel. For future climate scenarios, an increase in atmospheric static stability is predicted, which tends to dampen the fire plume height slightly. This is expected to slightly decrease the long-range transport of the biomass burning aerosol [27].

In conclusion, in order to estimate the impact of climate on the fire activity and vice versa, the human impact on ignition and spread of fire cannot be ignored. It is expected that human-driven ignitions in the low latitudes will decrease in future. However, land management practice and changes in fuel availability due to changes in bio-diversity are supposed to modulate fire activity as well, which requires examining fire activity in a broader context including processes impacting on the bio-productivity of land ecosystems.

Biogenic Aerosol

Biogenic Volatile Organic Gases and Secondary Organic Aerosols

The terrestrial biosphere is not only a source of carbonaceous particles due to biomass burning, but also for primary biological aerosol particles (PBAP) and biogenic volatile organic gases (BVOC) that partly oxidize to form condensable low volatility organic compounds that partition into secondary organic aerosols (SOA). Natural SOA is mainly produced from the plant-emitted BVOC species isoprenes and monoterpenes. BVOC emissions depend on environmental factors like temperature, light, leaf areas, and vegetation type [28, 29]. A fraction of BVOC is converted to SOA. BVOC emissions are estimated to lie in the range of 440–720 Tg/year for the sum of isoprenes and monoterpenes, while SOA production from BVOC is estimated to be between 20 and 380 Tg/year [1].

The understanding of the atmospheric processes leading to SOA formation is still incomplete. Investigations of climate impacts on SOA concentrations mostly focus on influences on BVOC emissions. Generally, BVOC emissions increase with rising temperature and decrease with higher atmospheric CO_2 concentrations [29]. Human-induced land use changes by, e.g., deforestation or agriculture influence BVOC emissions, as do variations in natural vegetation due to climate change impacts like droughts causing alterations in vegetation types and cover. Carslaw et al. [3] summarize several earlier publications that show that projections of future changes in isoprene emissions from climate effects (i.e., increasing temperatures) from year 2000 to 2100 range from an increase by 20 to 55%, increasing up to 90% when dynamic vegetation is included [30]. The response to land use changes ranged in different studies from − 20 to + 30%, while a decrease of 8%

was predicted in response to increasing CO_2 concentrations [31]. Monoterpene emissions were projected to increase from 20 to 60% considering the effect of changed climates [32–34].

Generally, plant stresses that may be induced by a warming climate increase BVOC emissions [35]. This includes the emission increase in response to temperature increase, but also to indirect consequences like insect infestations or droughts. A recent investigation that relates biogenic organic species concentrations in ice cores to the Arctic Oscillation supports the view that warmer temperatures lead to higher SOA concentrations [36]. Further recent publications highlight offsetting effects of the different influences but generally agree that rising temperatures lead to increasing BVOC emissions and SOA concentrations. Asmi et al. [37] analyze a multi-year aerosol record from an Arctic station in Russia. They also find from the measurements that warmer conditions in Siberia/Russia lead to higher emissions of organic species (as well as more wildfires) and thus cause increasing aerosol concentrations in the Arctic. Investigating the decadal trend of biogenic emissions with an interactive terrestrial biosphere model suggests that the CO_2 inhibition offsets the effects caused by warming climates despite the CO_2 fertilization effect on terrestrial vegetation [38]. Similarly, Lin et al. [39] project an increase by 18% in isoprene emissions by 2100 from an Earth system model, but while climate change alone would result in a 25% increase in SOA concentrations, projected changes in anthropogenic emissions would offset this trend leaving future SOA nearly unchanged. Also, natural and anthropogenic vegetation changes influence BVOC emissions such that changes may act in concert or cancel out [40, 41]. The wetting trend in the Tibetan Plateau since the 1980s is related to increased plant growth and higher BVOC emissions by Fang et al. [42] who hypothesize a positive feedback with warming trends. In contrast, Hantson et al. [41] find that monoterpene and isoprene emissions have decreased since the beginning of the twentieth century due to land use change, with these anthropogenic changes leading to decreasing isoprene emissions, and natural vegetation changes causing decreasing monoterpene emission trends. The authors point out that future emission trend projections strongly depend on land use and climate scenarios, but conclude that a future increase in global BVOC emissions is unlikely.

In addition to the effects on BVOC emissions, the atmospheric SOA production is also influenced by climatic conditions. Young et al. [43] compare conditions for the years 2010 and 2013 in San Joaquin Valley in California and find increased photochemical production of SOA when solar irradiance was enhanced. Zhao et al. [35] note that increases in BVOC concentrations under warming conditions increase particle sizes and thus CCN activities.

In summary, these studies show different climate impacts on BVOC emissions and SOA formation that may cancel each other. Future projections depend strongly on climate and land use scenarios. Investigations of the net effects of climate on SOA concentrations should focus on better understanding of the individual control mechanisms, but also on interpreting observed trends of BVOC and SOA concentrations at regional and global scales to separate the various underlying causes.

Primary Biological Aerosol Particles

Recent interest on atmospheric distributions of PBAP is motivated by their potentially important role in cloud ice formation and precipitation development (e.g., [44]). Number concentrations of biological particles were found to decrease by about two orders of magnitude from near-surface to the free troposphere [45]. Airborne biological particles can consist of pollen, fungal spores, and microorganisms such as bacteria, as well as fragments of biological organisms. Knowledge of the atmospheric amounts and emission processes of these particles is still incomplete, but it can be expected that PBAP emissions are related to characteristics of vegetation phenology that in turn depend on climate conditions. Understanding emission processes including release mechanisms of PBAP is needed to improve their emission and concentration estimates.

Fungal spores provide a considerable part of the PBAP [46]. Climate influence on the fungal spore release has been the subject of several recent publications [47]. However, since investigations focus on specific species and regions, generalizations remain problematic.

A clear enhancement of emissions of pollen and fungal spores during strong rainfall events has been observed [48]. Similarly, raindrops that impact on soils can disperse organic soil particles and soil bacteria into the air and thus provide an important emission mechanism [49, 50].

To study the impact of PBAP on ice formation, their transport by vertical mixing in altitudes where they can impact on clouds must be clarified. While in recent years the understanding of controls on PBAP emission and concentration has advanced, further field measurements are required. It has been noted that homogeneous sampling and analysis techniques for PBAP remain a challenge [51].

Marine Aerosol

Marine aerosol consists of sea salt, primary organics from surface films and secondary sulfate aerosols produced from DMS released by phytoplankton [52, 53]. Emission fluxes of primary marine aerosol species depend predominantly on surface wind speeds. The wind action leads to the formation of tiny air bubbles in the surface water, which form primary aerosol consisting of sea salt and organics when bursting at the surface [54]. DMS gas transfer from the ocean water to the atmosphere is enhanced by surface winds due to mixing of surface waters [55]. DMS emissions have received considerable attention as main player in the CLAW hypothesis [56],

which however has not been supported by observations [57]. Recent results related DMS emissions to the El Niño Southern Oscillation (ENSO), as consequence of surface wind speed patterns changes for the different ENSO phases [58]. Tesdal et al. [59] find that aerosol precursor DMS concentrations in the surface waters control its fluxes into the atmosphere, in addition to the surface winds.

Sea surface temperatures modify marine aerosol emissions. While phytoplankton productivity and thus, e.g., DMS production depend on the water temperatures (together with light and nutrient supplies), the temperature dependence of primary aerosol emissions was recognized only in the recent years. The temperature influence was detected in laboratory experiments and confirmed by field measurements [60–62]. At constant wind speeds, warmer temperatures lead to higher emissions and to larger particle sizes. The underlying physical cause of this dependency remains uncertain [63]. It may be related to the temperature dependence of water surface tension, viscosity or density, or to the solubility of air in water that influences the number and size of air bubbles in the surface water, which produce the primary aerosol when bursting. Oceanic surface wind speeds are not expected to change strongly in the future; therefore, it can be expected that marine primary aerosol emissions will increase due to increasing sea surface water temperatures [64].

The oceanic primary organic aerosol component dominates the submicron component of marine aerosol particles that may be transported over large distances and impact on clouds [65]. The contribution of primary organics has been related to marine microbiology, but the actual role of phytoplankton productivity for organic emissions is not yet clarified. Recent findings provide conflicting results about the relationship of organic marine aerosol emissions to phytoplankton photosynthesis as indicated by chlorophyll-a concentrations in surface water. Kasparian et al. [66] analyzed field measurements over the North Atlantic and relate organic ocean emissions directly to sea surface temperature and salinity. O'Dowd et al. [67] find that the organic aerosol content and phytoplankton productivity are better correlated on monthly rather than daily timescales. They suggest that the termination of a phytoplankton bloom releases large quantities of organic material such as transparent exopolymer particles, which form surface films and may be transferred into the marine aerosol. In contrast, strong correlation of organic aerosol and chlorophyll-a has been found in Mediterranean field measurements [68]. Since plankton productivity as well as species composition responds to changing climatic conditions, this connection should be clarified before believable conclusions on the potential change in organic marine aerosol for changing climates can be drawn.

Considering that large areas of the Earth are covered by ocean, marine aerosols have received less attention compared to their terrestrial counterparts. Dependencies of primary marine aerosol on temperature in addition to surface wind speeds have become increasingly evident. However, processes controlling emissions of aerosol species that are associated with oceanic biological activities are not fully understood and should therefore be studied in greater detail.

Climate Impacts on Aerosol Transport and Deposition

While the emission processes of the diverse aerosol species can vary considerably, all aerosol particle types are similarly affected by atmospheric transport and deposition. Boundary layer stability impacts vertical mixing of aerosols from the surface, controlling heights of aerosol layers, which in turn influence the impacts of aerosols on clouds and radiation (e.g., [69]). Aerosol particles are transported from their surface sources to higher altitudes by turbulent mixing and convection. Kipling et al. [70] emphasized the importance of convective mixing controlling the aerosol vertical profile for all types except dust. Simulations using a plume model to assess future changes in volcanic aerosol transport suggest that a future increase in tropopause height due to strengthened convection will lead to reduced transport of volcanic aerosol into the stratosphere reducing its lifetime and cooling effect [71].

The distribution of aerosol particles is predominantly determined by wind regimes determined by the atmospheric circulation. Depending on the injection height and vertical mixing related to atmospheric stability, aerosols can be transported over long distances to remote continents before they are removed from the atmosphere by dry and/or wet deposition. Large-scale weather systems control transport patterns from aerosol sources to remote regions. Aerosols from biomass burning and mineral dust from desert sources are thus found to travel across oceans and to remote continents. In turn, weather systems may respond to climate modes like the North Atlantic Oscillation (NAO) or ENSO. In this context, the transport of aerosol to the Arctic region is of particular interest due to the suspected role of absorbing aerosol in Arctic climate warming [72].

Many publications have examined the role of aerosol direct and indirect forcing on monsoon circulations (e.g., [73]). Recent interest has also focused on the role of monsoon circulations on aerosol transport, particularly in East Asia. High pollution conditions have been associated with stagnant weather conditions, and wind stilling connected to a weakening monsoon has been related to an increase in aerosol concentrations [74]. These conditions imply less aerosol dispersion and shallow planetary boundary layers, which are both connected to higher aerosol concentrations of pollutants compared to better ventilated conditions [39].

For the USA, Tai et al. [75] find from a multi-model study that future projections of a slowing circulation in the east with stationary conditions and less frequent ventilation will lead to slightly increased aerosol concentrations. High aerosol concentrations

in the USA are related to positive phases of the Pacific-North America teleconnection by influencing boundary layer height and precipitation [76]. While changes in emissions will remain the main factor in aerosol trends, climate change may enhance aerosol particle concentrations. However, not only shifts in mean climate should be considered but the influence of variations of meteorological episodes is of great importance to understand climate impacts on aerosol [77].

Climate modes also influence precipitation patterns, which control removal of aerosol from the atmosphere by wet deposition processes. Cloud processing and photochemical aging of hydrophobic aerosols like soot or mineral dust particles may change their hygroscopic properties, in turn modifying growth and atmospheric lifetimes of the particles [78]. A multi-model study of climate impacts on aerosol finds that a global increase in most aerosol species is explained by a decrease of large-scale precipitation over land in a warmed climate. This would lead to less wet deposition, increasing aerosol lifetimes and burdens [79]. The importance of initial hygroscopicity and aerosol aging, e.g., by nitric acid, on particle for wet removal efficiency and thus the remote distribution of soot aerosol has been shown by [80, 81].

In polluted regions, projected emission changes due to changes in environmental policies are expected to dominate changes in aerosol concentrations in response to climate variations [82]. Further untangling of the roles of atmospheric processes like transport, chemical processes, and wet removal in contrast to changes in emissions that due to either anthropogenic changes or natural causes will require a detailed understanding of the atmospheric controls on particle processes together with long-term observations.

Conclusions

While aerosol impacts on climate remain a major uncertainty factor in the climate system despite having been extensively studied, the climate controls on aerosol concentrations contribute large uncertainties to the understanding of the climate system as well. However, these controls are less studied.

The interdependencies of aerosol and climate processes encompass the potential of climate feedbacks that so far are only partly explored. In regions that are strongly affected by anthropogenic pollution (Fig. 1), impacts of climate aerosol variability are masked by changes in anthropogenic emissions that are a consequence of economic developments or environmental policies. In addition to impacts of climate variations on transport and deposition of aerosol, emissions and production of individual natural aerosol species are strongly affected by climate in different ways. The non-linear interdependencies of aerosol and climate require a comprehensive understanding of the whole aerosol-climate system considering the individual linkages. Understanding aerosol changes in a changing

climate will require (1) improving the detailed knowledge of climate controls on individual aerosol processes; (2) not only considering individual processes but viewing the multi-species aerosol mixture as an integral part within the climate system that should be studied with coupled multi-scale models; and (3) using long-term aerosol records to interpret past aerosol changes in terms of climate variability. Such knowledge would be prerequisite for robust future projections of aerosol distributions and feedbacks.

Compliance with Ethical Standards

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