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AERO-MAP: A data compilation and modelling approach to understand the fine and coarse mode aerosol composition

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Open Access Earth System Science Data Discussions

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Abstract. Aerosol particles are an important part of the Earth system, but their concentrations are spatially and temporally heterogeneous, as well as variable in size and composition. Aerosol particles can interact with incoming solar radiation and outgoing long wave radiation, change cloud properties, affect photochemistry, impact surface air quality, and when deposited impact surface albedo of snow and ice, and modulate carbon dioxide uptake by the land and ocean. High aerosol concentrations at the surface represent an important public health hazard. There are substantial datasets describing aerosol particles in the literature or in public health databases, but they have not been compiled for easy use by the climate and air quality modelling community. Here we present a new compilation of $PM_{2.5}$ and PM_{10} aerosol observations including composition, a methodology for comparing the datasets to model output, and show the implications of these results using one model. Overall, most of the planet or even the land fraction does not have sufficient observations of surface concentrations, and especially particle composition to understand the current distribution of aerosol particles. Most climate models exclude 10-30% of the aerosol particles in both $PM_{2.5}$ and PM_{10} size fractions across large swaths of the globe in their current configurations, with ammonium nitrate and agricultural dust aerosol being the most important omitted aerosol types. The dataset is available on Zenodo (https://zenodo.org/records/10459654, Mahowald et al., 2024).

1 Introduction

Recent Intergovernmental Panel on Climate Change (IPCC) reports and studies have highlighted the role of uncertainties in human-induced changes to aerosol concentration and composition in the atmosphere in limiting our ability to project future climate (IPCC, 2021; Gulev et al., 2021; Szopa et al., 2021). Aerosol particles are also a major contributor to air quality problems, which reduce life expectancy and quality of life (Burnett et al. 2018). Aerosol particles are suspended liquids or solids in the atmosphere originating from diverse sources and composed of a wide variety of chemicals (e.g., sea salts, dust, sulfate, nitrate, black carbon, organic carbon). Aerosol particles interact with incoming solar radiation, outgoing long wave radiation, change cloud properties and lifetimes, and modify atmospheric photochemistry (Mahowald et al., 2011; Kanakidou et al., 2018; Bellouin et al., 2020). Once deposited on the surface, they can modify land and ocean biogeochemistry, as well as the albedo of snow and ice surfaces (Mahowald et al., 2017; Hansen and Nazarenko, 2004, Skiles et al., 2018). New satellite remote sensing measurements provide important information about temporal and spatial distribution of aerosol particles, but challenges remain in quantifying the chemical composition of aerosol particles (Kahn et al., 2005; Tanré et al., 1997; Remer et al., 2005). In addition, the AERONET surface remote sensing network provides some information about loading, size and absorbing aerosol properties related to composition (Holben et al., 1991; Dubovik et al., 2002; Schuster et al., 2016; Goncalves et al., 2023; Obiso et al., 2023). Both the magnitude of the effects, and sometimes the sign of the aerosol effects are dependent on the composition and size of aerosol particles (Mahowald et al., 2011b, 2014a; Bond et al., 2013; IPCC, 2021). In addition, one cannot understand the impact of humans on aerosol particles without understanding the sources of aerosol particles, which is related to their chemical composition. Obtaining information about the composition and size of aerosol particles in many cases requires in situ observations, which are limited in space and time (Hand et al. 2017; Philip et al. 2017; Yang et al. 2018; Collaud Coen et al. 2020).

The climate and aerosol modelling community, especially under the auspices of AEROCOM, has compiled datasets and organized comparison projects that have provided substantial information to improve aerosol models (Huneeus et al., 2011; Textor and others, 2006; Dentener et al., 2006; Schulz et al., 2006; Gliß et al., 2021) or knowledge of the aerosol properties like cloud condensation nuclei (Laj et al., 2020; Fanourgakis et al., 2019). However, most of these comparisons include data only from North America and Europe (e.g., Szopa et al., 2021). In addition, previous compilation studies have focused primarily on understanding fine aerosol particles (here defined as particle with a diameter less than 2.5 µm) and improving model simulation of these aerosol particles, because of their importance for air quality, cloud interactions and short-wave forcing (Collaud Coen et al., 2020; Bellouin et al., 2020; Fanourgakis et al., 2019). Coarse mode aerosol particles (defined as those particles with a diameter larger than 2.5 µm) are important for long wave radiation interactions, cloud seeding and for biogeochemistry, and these interactions have received less attention (Jensen and Lee, 2008; Mahowald et al., 2011; Karydis et al., 2017; Chatziparaschos et al. 2023). In contrast to the many fine aerosol compilations and comparisons (usually considering particles with diameter less than 2.5 µm or $PM_{2.5}$), there are fewer studies focusing on aerosol compilations for both fine and coarse aerosol particles, and their comparison to models (Kok et al., 2014b; Albani et al., 2014b; Huneeus et al., 2011; Gliß et al., 2021, Kok et al., 2021). Nonetheless, there are many observations of the coarse particle mass included in the particles with diameter less than 10 µm (PM_{10}) (e.g Hand et al., 2017), and most climate models include these aerosol particles (e.g. Huneus et al., 2011). Compilations of in situ data are available for dust and iron aerosol particles (Kok et al., 2014b; Albani et al., 2014b; Mahowald et al., 2009) and for sea salts (Gong et al., 1997). Other studies have focused on the important topics of wet deposition (Vet et al., 2014) or trends in aerosol properties (e.g., AOD, surface PM) (Mortier et al., 2020; Aas et al., 2019). Observations of PM_{10} or coarse and fine aerosol particles are available for many regions and individual sites (e.g., Malm et al., 2007; Hand et al., 2019; Maenhaut and Cafmeyer, 1998; Artaxo and Maenhaut, 1990; McNeill et al., 2020) but have not previously been compiled into one database. For example, much of the air quality data used for deducing health impacts of aerosol particles is not publicly and easily available in a format appropriate for use in Earth system models (e.g., van Donkelaar et al., 2021, https://www.who.int/data/gho/data/themes/air-pollution/who-air-quality-database). Aerosol modelers need as much information as possible about the composition of the aerosol particles. Thus, there is a need to compile both $PM_{2.5}$ and PM_{10} in situ concentration data into one database to make it easy for modellers to compare model results with observations. One goal the aerosol community should work towards is making aerosol measurement datasets publicly available, while acknowledging the principal investigators who produced these datasets, which we hope this paper serves as a step towards achieving.

https://doi.org/10.5194/essd-2024-1
Preprint. Discussion started: 6 February 2024

The current generation of Earth system models used for the IPCC simulations tends to include the dominant aerosol particles (desert dust, sea spray, black carbon (BC), organic matter (OM) and sulfate) but not all aerosol particles. For example, some Earth system models ignore ammonium nitrate aerosol particles although these are known to be important for climate and biogeochemistry, and are impacted by human activities (Paulot et al. 2016; Adams et al., 1999; Thornhill et al., 2020). In addition, some models focus only on fine mode OM and BC aerosol particles, although there is evidence for coarse mode aerosol particles of both carbonaceous aerosol particles (Graham et al., 2003; Mahowald et al., 2005). Agricultural or land use sources of dust are not included in most models, although they could represent 25% of the anthropogenic sources (Ginoux et al., 2012), and significantly impact transported transhemispheric aerosol composition (García et al., 2017). In addition, fugitive, combustion and industrial dust emissions have traditionally been ignored as well, although emission datasets are available (Philip et al., 2017). In this study we use available observations to constrain a model estimate of the total PM_{10} and $PM_{2.5}$, and deduce the importance of these often-neglected aerosol particles. We propose a methodology for constraining aerosol particles that are not directly measured (dust or sea salts) using their elemental composition. Note that we exclude super coarse (>PM10) particles here because of the lack of available data, although studies have suggested their importance for climate interactions (e.g. Adebyi et al., 2023).

Harmonizing models with different types of measurements is critical (Huang et al., 2019). Models operate with the geometric or aerodynamic particle diameter, whereas in practise the measurements are done with a variety of particle equivalent diameter, e.g., optical, volume equivalent, projected-area equivalent, aerodynamic diameter or electrical mobility diameter, depending on the instrument used (Hinds, 1999; Reid et al., 2003; Rodríguez et al., 2012). In the inlets of the samplers used for the mass-measurements and collection of $PM_{2.5}$ and PM_{10} particles for subsequent chemical analysis, such size cut-off at 2.5 µm and 10 µm is defined in terms of aerodynamic diameter (i.e., geometric diametric weighed by the particle density; Hinds, 1999_/ The sharpness of the cut-off of such inlets influences the $PM_{2.5}$ and PM_{10} mass concentration (Hand et al., 2019; Wilson et al., 2012). The PM_{10} size cut-off aerodynamic diameter is equivalent to $PM_{6.3}$ geometric diameter for spherical dust particles (Hinds, 1999; Rodríguez et al., 2012) and to $PM_{6.9}$ in the case of dust aspherical particles (Huang et al., 2021). Similarly, PM2.5 (aerodynamic diameter) is equivalent to PM1.6 (geometric diameter) for dust. We consider the importance of this in global model simulations and comparisons to observations. Similar issues can occur for PM2.5 sized particles, which we do not yet consider in this study. In addition, there are issues about the temporal changes in aerosols. There have been substantial trends in emissions especially of anthropogenic aerosols over the last 40 years (Quass et al., 2022; Bauer et al., 2022). For this first study we include all aerosol data available, spanning 1970s to 2020s. Future studies will consider the temporal changes in the data in more detail.

For this study we focus on the following: a) compiling available $PM_{2.5}$ and PM_{10} aerosol data, including aerosol composition into a new publicly available database for the modelling community (AEROMAP); b) presenting a methodology to compare these observations to an Earth system model, which has extended the normal sources of aerosol

https://doi.org/10.5194/essd-2024-1
Preprint. Discussion started: 6 February 2024

particles to include more types of aerosols such as agricultural dust, nitrates, etc.; c) identifying the measurement and modelling gaps from this comparison. In this paper, we focus on the annual average distribution of aerosol particles and some key chemical composition information. Future studies will focus on seasonal and daily scale variations and temporal trends as well as trends in other chemicals or elements within the aerosols.

2 Description of Methods

2.1 Observational data

PM observations are made by multiple networks, or during specific field campaigns, and for different size cut-offs, with and without a description of chemical composition. As expected, most of the observations are over North America or Europe, with much of the rest of the land areas and most of the ocean much more poorly observed (Fig. 1; supplemental dataset 1). For this study, we include both $PM_{2.5}$ and PM_{10} daily (or multiple day averages) data sets that were made available by the investigators or are available from public web sites (Fig. 1; supplemental dataset 1). Some measurement sites measure $PM_{2.5}$ and coarse ($PM_{2.5}$ to PM_{10}) aerosols. For those sites, we convert the latter to PM_{10} for comparison. Some measurement sites have only a few observations of composition or mass, while others have multiple years: we included less complete datasets at sites in regions with limited data. The time period for different datasets is included in the supplemental information.

Detailed studies have shown that PM_{10} and $PM_{2.5}$ samplers can differ in the sharpness of their size cut-off (Hand et al., 2019). As an example, comparisons between data from the U.S. Environmental Protection Agency (EPA) Federal Reference Method sites and data from the Interagency Monitoring of Protected Visual Environments (IMPROVE) network show that the coarse matter from collocated sites in both networks were offset by 28% (Hand et al., 2019). There was a bias when data were compared (slope of 0.9), but the correlation coefficient was high (0.9) suggesting overall a good agreement. We focus here on surface station measurements of PM_{10} and $PM_{2.5}$, since our model and most models only consider mass up to PM_{10}. For that reason, our model deposition is not directly comparable to observational bulk/total atmospheric deposition since larger particles may dominate the deposition close to the source areas (Kok et al., 2017; Mahowald et al., 2014b; Neff et al., 2013). Measuring absolute dry and wet deposition rates is also technically more challenging (especially dry deposition, since the particles can be re-entrained into the atmosphere), but worthwhile (Heimburger et al., 2012; Prospero et al., 1996). In regions with little data (e.g., outside of North America and Europe) we include measurements of total suspended particulates (TSP) with the PM_{10}, because of the lack of size-resolved data. Data from the Japanese air quality network use a different inlet for the PM10 cutoff as well, which will include a slightly larger size fraction (https://tenbou.nies.go.jp/download/).

In addition to particulate matter in the PM_{10} and $PM_{2.5}$ size fractions, we also compile the following observations to compare to the model: black carbon (BC), elemental carbon (EC), organic carbon (OC) (or particulate organic material, OM, that is

https://doi.org/10.5194/essd-2024-1
Preprint. Discussion started: 6 February 2024

here considered to be 1.8 x OC in mass), sulfate, nitrate, aluminum, sodium and chloride. To include both BC (based on light absorption measurements) and EC (based on thermal oxidation induced combustion measurements) data are also a source of uncertainty, both are proxies of the soot combustion particles since they are based on different measurements techniques, and there is no accepted equivalence between them (Mbengue et al., 2021) .Details on how the model is compared to data for different elements is in section 3.2.

For this paper, we focus on the annual means which are calculated for all values at each station that are above the detection limit and reported here. At some stations or times, concentrations can be below the detection limit, and excluding these data or time periods could bias our average values. We focus on the stations that have more than 50% of the data above the detection limit, and exclude other sites. For those included stations, if the values were reported as below the detection limit, we include in the average one-third of the minimum detection limit. The reported detection limits should bound the upper limit of aerosol mass and allow us to include sites, whose observations were otherwise too low to include, while reducing the potential biasing of our compilation towards higher values (doi: 10.5281/zenodo.10459654, Mahowald et al., 2024).

2.2 Model description

Simulations of aerosol particles were conducted using the aerosol parameterizations within the Community Atmosphere Model, version 6 (CAM6), the atmospheric component of the Community Earth System Model (CESM) developed at the National Center for Atmospheric Research (NCAR) (Hurrell et al., 2013; Scanza et al., 2015; Liu et al., 2012). The aerosol module in this version is closely related to the module used in the Energy Exascale Earth System Model (Golaz et al., 2019; Caldwell et al., 2019). Simulations were conducted at approximately 1°x1° horizontal resolution with 56 vertical layers for four years, with the last three years (2013-2015) used for the analysis (Computational and Information Systems Laboratory, 2019). The model simulates three-dimensional transport and wet and dry deposition for gases and aerosol particles based on MERRA2 winds (Gelaro et al., 2017).

The model included prognostic dust, sea salts, BC, OM, and sulfate aerosol particles in the default version, using a modal scheme based on monthly mean emissions for the year 2010 (Liu et al., 2012, 2016; Li et al., 2021). For this study, the coarse size mode (mode 3) was returned to the CAM5 size parameters (geometric standard deviation of 1.8) to better simulate coarse mode aerosol particles, and improve the dry deposition scheme and optics used in the model for simulating coarse mode aerosol particles like dust as described in Li et al. (2022b).

Desert dust is entrained into the atmosphere in dry, sparsely vegetated regions subject to strong winds. We use the Dust Entrainment and Deposition scheme (Zender et al., 2003) with the emitted size distribution given by the updated Brittle Fragmentation Theory (Kok et al., 2014b, a) with improved incorporation of aspherical particles for optics and deposition (Li

https://doi.org/10.5194/essd-2024-1
Preprint. Discussion started: 6 February 2024

et al., 2022b; Huang et al., 2021; Kok et al., 2017). Fossil fuel and natural emissions of sulfate, OM, and BCfollow the Climate Model Intercomparison Project 6 emission scenarios for present day (Gidden et al., 2019).

2.2.1 Modelling of additional aerosol sources and types

The model was modified to allow the addition of several new aerosol particles based on codes with expanded dust speciation (Li et al., 2022b) but here the extra dust tracers are used for the additional species as described below. The additional sources of aerosol particles use the same optical properties as bulk dust for this sensitivity study. The following aerosol particles were added, and the amount of emissions in each the $PM_{2.5}$ and PM_{10} sizes and contribution to surface concentration and aerosol optical depth are shown in Table 1. In addition, some of the base case aerosol emissions were modified to match observations, as discussed below.

Agricultural sources of dust are added to this version of the model using the same emission scheme as for natural sources (Kok et al., 2014b, a; Li et al., 2022b), but applied to the crop area, and each region is tuned to have the percentage amount of anthropogenic dust to match satellite based observations (Ginoux et al., 2012), except Australia, where other estimates (Bullard et al., 2008; Mahowald et al., 2009; Webb and Pierre, 2018) suggest a lower amount (see Table S1 for comparisons, based on Brodsky et al., 2023). Agricultural dust is separately considered by the model, so its importance can be isolated.

Coarse BC and O< as well as fine and coarse ash from industrial sources were added. Emissions estimated from the GAIN model are added to the model using the ECLIPSEV6_CLE base case (Klimont et al., 2017; Philip et al., 2017). Coarse BC and OM from biomass burning were assumed to be 20% of the fine mode mass (Mahowald et al., 2005).

Primary biogenic particles are released from ecosystems either as integral particles, such as bacteria, pollen or spores, or as accidentally entrained leaf pieces (Jaenicke, 2005; Mahowald et al., 2008; Despres et al., 2012; Burrows et al., 2009; Heald and Spracklen, 2009). These sources are poorly observed or understood, and thus looking at coarse mode organic material in this study could provide additional constraints on the budget. Assumptions about size are likely to be very important for the resulting distribution and impacts (e.g., compare P budgets from assuming one size versus five different bins (Brahney et al., 2015)). Four different types of primary biogenic particles were included: bacteria, spores and other miscellaneous emissions (leaf bits, pollen, etc.) from land ecosystems, as well as a marine organic aerosol. Included bacteria sources were read in from a monthly climatology (Burrows et al., 2009). Spore sources were calculated offline and read into the model based on observed leaf area index, temperature, and a source parameterization (Janssen et al., 2020; Heald and Spracklen, 2009). Other terrestrial emissions were estimated based on leaf area index following Mahowald et al. (2008). Marine organic aerosol emissions were included based on the physically based scheme OCEANFILMS (Burrows et al. 2014). Marine organics are externally mixed with sea spray, following Zhao et al. (2021). OCEANFILMS only estimates the fine mode

https://doi.org/10.5194/essd-2024-1
Preprint. Discussion started: 6 February 2024

organic mass, and here we assume that the coarse mode marine organic mass equals 1% of the seaspray mass, (Gantt et al., 2011). The assumptions about the mass and fraction in each size bin are shown in Table 1.

Ammonium nitrate aerosol particles are not included in the standard CAM6 nor in E3SM, but are thought to be important for aerosol optical depth and surface concentrations (Paulot et al., 2016; Adams et al., 1999; Thornhill et al., 2020, Bauer et al., 2007, 2016). Nitrate can also be taken up onto dust particles, for example, but that is ignored in this study (Dentener et al., 1996). Ammonium nitrate aerosol particles require tropospheric chemistry interactions because the nitrogen based aerosol particles interact with tropospheric photochemistry and the aerosol particles are in chemical equilibrium with the gas phase (e.g. Nenes et al., 2021; Baker et al., 2021; Bauer et al., 2007; 2016), so simulations using the CAM-CHEM model with tropospheric photochemistry are used covering the same time period (Vira et al., 2022). Simulations with chemistry were conducted at 2°x2° resolution and are linearly interpolated to 1°x1° resolution used for the other modelled aerosol particles. Sulfate in the CAM6 is assumed to be in the form of ammonium sulfate and the nitrate is assumed to be in the form of ammonium nitrate for these studies, so as a rough approximation only the ammonium nitrate needs to be added to consider nitrogen aerosol optical depth. While aerosol amounts are simulated, ammonium nitrate aerosol optical depth is not calculated within the model but offline. The model does calculate sulfate aerosol optical depth, which has a roughly similar increase in size with humidity, and similar optical properties as long as the nitrates and sulfates are in similar size fractions (Paulot et al., 2016; Bellouin et al., 2020). Therefore the aerosol optical depth from ammonium nitrate (per unit mass) is assumed to be proportional to the sulfate aerosol optical depth per unit mass in each grid box at each time interval. Detailed comparison of the nitrate and ammonia aerosol particles, and other species was conducted in Vira et al. (2022). Overall, the model can simulate some of the spatial distribution, but overestimates the nitrate aerosol amounts. This is also seen in Vira et al. (2022), and as shown in Table 1, the calculated nitrate aerosol amounts are multiplied by 0.5 to best match the available observations.

2.3 Model-observation comparison methodology

Comparisons of the observations to model concentrations were done using BC, OC, SO_4^{2-}, Al, NO_3^-, NH_4^+, Na, and Cl composition measurements. Some of these elements/compounds map directly onto model constituents (BC, OC, SO_4^{2-}, NO_3^-, and NH_4^+), while others serve as proxies for modelled constituents (Al for dust and industrial ash, Na and Cl for sea salts, S for sulfate, etc.). We use non-sea-salt sulfate in ocean regions for estimating sulfate. Some observing networks like IMPROVE use a composite of elements to deduce dust amounts (e.g., Hand et al., 2017). We do not choose to do this for two reasons: first at some sites not all the elements are available, and secondly because these elements are not only from desert dust, but also from industrial sources, so we explicitly include industrial ash sources and Al in those sources to compare. Note that model values come from the midpoint of the bottom level of the model (~30 m) while the observations are usually taken at 2 or 10 m high. Modelled values of PM content, which assume dry particles, are used, while gravimetric

https://doi.org/10.5194/essd-2024-1
Preprint. Discussion started: 6 February 2024

measurements in some networks are equilibrated at 50% relative humidity, thus 5-25% of the mass of measured PM can be water (Prank et al., 2016; Burgos et al., 2020).

For the most part, we use model output for which there is a one to one relationship with what is being measured (BC, sulfate, etc). However, for dust this is not straightforward, as dust is composed of multiple elements. Here we use Al as a proxy for dust, as it is relatively constant (~7%) in dust (as opposed to Ca, which varies highly, or Fe which varies moderately) (Zhang et al., 2015). Al sources are primarily from dust, agricultural dust, road dust and industrial ash emissions; we ignore minor emissions from volcanoes, marine sea spray and primary biogenics for this study (Mahowald et al., 2018). Assumptions about the model composition and how they are compared to observations are shown in Table S2. Since we compare the sea salt variable to both Na and Cl (if Na is not available), we show the conversion of Cl to Na for sea salt calculations, and other assumptions for converting observations to be consistent with each other in Table S3. For example, OM is assumed to be 1.8 times OC.

Previous studies have pointed out that PM_{10} observations of dust represent an approximate cut-off of the aerodynamic diameter of 10 µm, which could be converted within the model to a 6.9 µm geometric diameter cut-off assuming spheroids, like dust (Huang et al., 2021). Using standard relationships between the modal aerosol particles used in the CAM6 (Liu et al., 2016) and the fraction of the aerosol particles below 6.9 µm (Seinfeld and Pandis, 2006) (here referred to as $PM_{6.9}$), a new diagnostic was added to the model, which shows that in regions with substantial coarse aerosol particles like dust, there can be a difference of about 30%, while in most places the differences are less than 5% (Fig. S1). These assumptions are less true for coarse particles like sea salts, but the differences are small in sea salt dominated regions (Fig. S1). For this study we use $PM_{6.9}$ from the model. Note that the inlet size discrimination for $PM_{2.5}$ measurements are also not a step function and also this might impact the comparisons for $PM_{2.5}$.

For ease of viewing the data in this paper in the densely sampled regions, observational records from different sites were combined into a mean within a grid cell that is two times the model resolution, or approximately 2° x 2°. This process averages the observations over a spatial scale appropriate for comparison with the chemistry model (Schutgens et al., 2016). We provide both the annual average data at each site as well as the averaged data (with the modelled data at doi: 10.5281/zenodo.10459654, Mahowald et al., 2024).

Notice that we include both urban regions and rural or remote sites into the same dataset. Some of the data is not resolved in location better than 0.25 degrees, so that the coordinates of the locations here provided with the gridded data should not necessarily be used for finer resolution studies. Because of the importance and size of megacities crossing multiple grid boxes including available data are more representative than excluding polluted regions for air quality and climate studies, but each application should make appropriate choices, and studies show expected differences between urban and rural concentrations and trends (e.g., Hand et al., 2019).

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3 Results

3.1 AEROMAP observational data set

First, we assess the amount of data and the number of stations within each ~ 2° x 2° gridded area (Fig. 1). The observational dataset provides coverage predominately over North America and Europe for $PM_{2.5}$ and PM_{10}, as noted by previous studies (e.g., Szopa et al., 2021), but in addition we provide here a synthesis of more air quality data in other regions, especially Asia (Fig. 1). This data set comprises most of the individual observations (at daily or higher time periods) of total $PM_{2.5}$ (Fig. 1a, 1e: blue bars) and most of the observing stations (Fig. 1e and blue line). Approximately 15,000 stations and over 20 million observations are included in this compilation as annual averages.

Notice that there are two to three orders of magnitude more daily observations for the total mass (PM) of aerosol particles compared to information about the composition of aerosol particles (Fig. 1e, which is shown also by contrasting the spatial distribution of measurements between $PM_{2.5}$ and measured amounts of OM (Fig. 1a versus 1b), as well as a large difference between the number of stations measuring the total mass versus the speciated aerosol particles like OM (Fig. 1c versus 1d). While this dataset presents a huge increase in the amount of data available to the aerosol modelling community, still most of the total $PM_{2.5}$ or PM_{10} data are clustered over a few regions, and there is little composition information over most of the globe (Fig. 1).

This dataset is presented as an annual mean, but the data span 1986 to 2023 (Fig. 1f). Over this time period there are statistically significant trends in different regions (Quass et al., 2022; Bauer et al., 2022). For this dataset we choose to present all the data averaged, but in future studies there will be datasets of seasonality and interannual variability of aerosols as available. Note that the model uses emission estimates for 2010, and meteorology for 2013-2015, so that there could be differences in the model-data comparison because of the time period discrepancies.

3.2 $PM_{2.5}$ model-data comparison

Modelled concentrations of $PM_{2.5}$ are more often compared against observations than for PM_{10} or other size fractions, and comprise an important portion of the particulate matter associated with human activities. Therefore, we describe first the observational synthesis and comparison to model results for $PM_{2.5}$. Because the high number of observations in some parts of the world would make the figures unreadable, the observations are gridded onto an approximately 2°x2° grid for comparisons with the model (Fig. 2a). As expected, in the model the highest concentrations are over the desert dust regions, such as North Africa, and over heavily industrialized regions in Asia. For the heavily industrialized regions in Asia, these high values are consistent with the observations, but the regions in North Africa with the highest modelled values do not have similar observational validation for high concentration values due to a lack of data (Fig. 2a).

Overall, the model is able to simulate much of the spatial variability in $PM_{2.5}$ over two orders of magnitude (Fig. 2a and 2c), however there is an overestimate in the $PM_{2.5}$ over India and China (Fig. 2b). These discrepancies over India and China could be due to errors in the input emissions datasets or the aerosol transport modelling, or to differences in the time periods covered: the observations are more recent while the assumptions for the emissions are for the year 2010. There could also be methodological and analytical differences due to which group or network did the observations or the exact locations of the different monitors. Much of the data in those regions are not usually included in compilations of data, so the fact that previous model studies have not been able to assess emission datasets in these regions could explain this discrepancy. Comparison between different observations in some cities (Fig. 3) shows that in these grid boxes there can be very large differences (~factor of 3) between the annually averaged values reported at nearby stations within 1° distance radially, and that the AirNow measurements (**https://www.airnow.gov/international/us-embassies-and-consulates/** on the US embassies) tend to be higher than those reported from government air quality networks. The sites compared are in large cities and thus are likely to have strong local sources and intense gradients in pollutants. For now, we keep in mind this large difference, but continue to use the observations. As indicated below, in these regions we do not have measurements of composition so we do not know which constituents are poorly simulated in our emissions or transport modelling. More statistics describing the model data comparisons are shown in Table S4.

The scatterplots show the comparisons of the model to the observations using the gridded data (Fig. 2c) and all original data (Fig. 2d), and the correlation coefficients are slightly better using the original data (Fig. 2d) (0.78versus 0.69 for the gridded data). Notice that most of the data in the Asian region (blue plusses) show the highest model bias.

Next, we consider the composition of the $PM_{2.5}$ aerosol in the model versus the observations, starting with the aerosol components in the default version of the model. Sulfate aerosol particles tend to be overestimated in the model in North America, but not over Europe and other regions (Fig. 4a and b). Previous studies have compared SO_4^{2-} aerosol observations to simulations and have not noted this bias (e.g., Barrie et al., 2001; Aas et al., 2019) but this bias was seen in this model (Liu et al., 2012; Yang et al., 2018). BC comparisons suggest the model results are roughly able (r=0.25) to capture the spatial dynamics of this aerosol across more than 2 orders of magnitude (Fig. 4c and d). This is similar to previous model intercomparisons (Koch et al., 2009; Bond et al., 2004, 2013; Liu et al., 2012, 2016). Simulations of OM in the default model suggest that the model has difficulty in simulating all the variability in OM. Correctly modelling organic material is very difficult both due to the sparsity of data for comparison, as well as the existence and importance of OM in both primary and secondary aerosol particles (Heald et al., 2010; Kanakidou et al., 2005; Olson et al., 1997; Tsigaridis et al., 2014), and previous studies with this model have noted an overestimate in comparison with surface observations (Liu et al., 2012). In our study we include primary biogenic aerosol particles, which are usually not included in model studies (Mahowald et al., 2011, 2008; Jaenicke, 2005; Heald and Spracklen, 2009; Burrows et al., 2009; Myriokefalitakis et al., 2016), but these are a very small part of the $PM_{2.5}$ and occur mostly in the coarse fraction (Table 1) and thus are not causing the bias, which must be due to biomass burning and/or industrial emissions.

As a proxy for sea salts, we use the elemental data of the major components, Na and Cl, and there the model tends to be too high at low Na and too low at high Na in North America, where much of the data are available (Fig. 4g and h), which has been seen previously with this model (Liu et al., 2012). Notice that we do not include industrial emissions of Na or Cl as they have not been spatially estimated. As a proxy for dust, we use Al amounts (Fig. 4i and j), which globally and over dust regions are dominated by dust, although there are few observational datasets in high dust regions. The comparisons suggest the model is able to simulate dust across 4 orders of magnitude, similar to previous studies (Liu et al., 2012; Albani et al., 2014a; Li et al., 2022b; Huneeus et al., 2011) although there is a tendency for a high bias in the models over low dust regions and a low bias in high dust regions, similar to sea salts (Fig. 4i and 4.j).

Next, we consider the nitrogen aerosol ammonium nitrate which requires complicated gas-aerosol phase equilibrium to correctly simulate (e.g., Bauer et al., 2007; Thornhill et al., 2021; Adams et al., 2001; Regayre et al., 2018; Seinfeld and Pandis, 1998). To summarize these complicated interactions, because SO_4^{2-} is a stronger acid than NO_3^- in the atmosphere, the basic NH_4^+ is preferentially found with SO_4^{2-}. Thus NO_3^- aerosol particles will only form if there is sufficient NH_4^+ left over, therefore the ratio of NO_3^- to NH_4^+ can vary. As described in the methods, to include these aerosol particles we used simulations from a different version of the same model which include chemistry (Vira et al., 2022), and a more process-based source of ammonia (Vira et al., 2020) since the default CESM2 version used here for most aerosol particles does not include chemistry. Note that even in the chemistry version of the model for CESM2 the complicated gas-aerosol phase equilibrium is not included, which causes errors in the simulation of the amounts of nitrogen aerosol (e.g., Bauer et al., 2007; Thornhill et al., 2021; Adams et al., 2001; Regayre et al., 2018; Nenes et al., 2021); thus while the NH_3 emission scheme used in this model is state-of-the-art, the lack of an adequate gas-aerosol phase separation may lead to biases as discussed in Vira et al. (2022). NO_3^- aerosol particles compared against available observations show that over 2 orders of magnitude, the model results are able to simulate the spatial variability (Fig. 4k and l). Note that here, we have multiplied the simulations by a factor 0.5 in order to achieve a good mean comparison, as indicated by Vira et al. (2022). In addition, NH_4^+ results show the importance of NH_4^+ over agricultural regions especially (e.g., Vira et al., 2022), and that the NH_4^+ in the simulation used here compares well to available observations (Fig. 4m and n; Vira et al., 2022).

3.3 PM_{10} model-data comparison

PM_{10} used to be the standard air quality measurement until more studies showed that smaller particles ($PM_{2.5}$ or PM_1) were more relevant for health impacts (e.g., https://www.epa.gov/pm-pollution/timeline-particulate-matter-pm-national-ambient-air-quality-standards-naaqs, accessed October 4, 2023). However, there are still many PM_{10} measurements routinely made (Fig 1d; Fig. 5a). As discussed in the methods, what is described as measurements of PM_{10} (aerodynamic diameter) is probably closer to $PM_{6.9}$ (geometric diameter) as simulated in models (Huang et al., 2021), so here we use the $PM_{6.9}$ fraction as calculated in the model to compare to PM_{10} observations (Fig. S1 shows the fraction of PM_{10} that is $PM_{6.9}$: this is only important in regions with substantial coarse mode emissions like desert dust source regions. For marine coarse aerosols like sea salt, the distinction between geometric and aerodynamic diameter may be smaller.). The model is able to simulate PM_{10} concentrations across 2 orders of magnitude with some skill (Fig. 5a, c and d), although the region of East Asia, especially China and India are overestimated in the model similar to the $PM_{2.5}$ (Fig. 3a, c and d). Gridding the data before comparing to the model results in a lower correlation across space than including all data (fig 5c vs. d), suggesting the existence of significant sub-grid variability and that some oversampled regions (e.g., European or North American cities) may be better simulated by emissions or transport models. Again we are including new data in this study that have previously not been used, and the discussion in the $PM_{2.5}$ section is appropriate here (although we do not have AirNow measurements to compare against in the PM_{10} size) for both the variability with grid cells as well as the potential for model and data errors. PM_{10} aerosol particles have a shorter lifetime (Textor et al., 2006; Mahowald et al., 2011), allowing for more variability within a grid cell. More statistical comparisons are shown in Table S5.

There are fewer comparisons with PM_{10} composition data available in the literature: usually only sea salts and dust are compared to observations which include the coarse mode (Gong et al., 2003; Ginoux et al., 2001; Albani et al., 2014b; Mahowald et al., 2006). Comparisons for SO_4^{2-} (Fig. 6a and b.) suggest that within a large range the model does a good job of simulating the available observations. Similarly, for BC, the PM_{10} simulation captures the range of values, with less spread than SO_4^{2-} (Fig. 6c and d in contrast to a and b). Note that unlike many studies we include BC in the PM10 mode, since observations show that there is some contribution of BC to PM10 (compare figure 6c versus 4c). The model simulations for OM include primary biogenic particles and the limited available observations do not support larger sources of OM in the PM_{10} than included here (as suggested in e.g., Jaenicke, 2005): indeed the model may be overestimating the OM in PM10. Similarly, the limited Na and Cl (proxy for sea salt) data suggest the model in some places may overestimate Na even over continents (Fig. 6g and h), as discussed in the $PM_{2.5}$ section, as was seen previously (Liu et al., 2012). Comparisons with Al (proxy for dust) show that the mean is well simulated, but there are regions with poor agreement, consistent with previous studies with this model (Li et al., 2022b; Kok et al., 2014b; Albani et al., 2014a; Matsui and Mahowald, 2017; Zhao et al., 2022), and indeed across most dust models (Huneeus et al., 2011).

For the NO_3^- in aerosol particles, similar to the $PM_{2.5}$ size, the particles were multiplied by 0.5 to better match the observations following Vira et al. (2022) (Fig 6k and l). The model simulations suggest too high values in high NO_3^- areas, and too low in low NO_3^- regions (Fig. 6k and l). NH_4^+ shows a slightly better comparison to the limited available data (Fig. 6m and n) as seen in Vira et al. (2022). Note that as discussed earlier, the model does not include other forms of nitrate aerosols which may be important, such as the uptake of nitrate onto dust aerosols (Dentener et al., 1996; Xu and Penner, 2012;

3.4 Data and model coverage

The compilation shown here is the most comprehensive currently available for total mass and composition of in situ concentration data, and yet it highlights the lack of sufficient data to constrain the current distribution of aerosol particles and their composition (Fig. 7a and b). Only 3% of the grid boxes (2°x2°) have $PM_{2.5}$ data (about 10% of land grid boxes), and only 0.3% has sufficient data to constrain most of the composition (defined as having 90% of the variables considered here: total mass, SO_4^{2-}, BC, OM, Na or Cl, Al or dust, NO_3^- and NH_4^+). There are even less data available to characterize PM_{10}, which is less important for air quality and aerosol-cloud interactions but more important for aerosol-biogeochemistry interactions and long wave interactions (Mahowald et al., 2011; Li et al., 2022a; Lim et al., 2012; Kanakidou et al., 2018). Because of the high spatial and temporal variability and the lack of satellite or other remote sensing data to characterize the type of aerosol, this lack of data is a severe handicap in constraining aerosol radiative forcing uncertainties and other impacts of aerosol particles in the climate system.

In this simulation, we included several new aerosol sources and types that are not in the default model to investigate their importance. For the CESM this simulation includes agricultural dust, nitrogen aerosol particles and several other sources (see Table 1). As shown in Fig. 8, the default aerosol particles are the dominant aerosol particles over most of the planet, but in many regions for both $PM_{2.5}$ and PM_{10}, the default aerosol scheme includes less than 30% of the aerosol particles (Fig. 8a and c), with substantial contributions from the new added aerosol particles (Fig. 8b and d), especially nitrogen aerosol particles and agricultural dust. Many Earth system or climate models such as the CESM2 do not include nitrogen aerosol particles (NO_3^- and NH_4^+), because of the substantial complexity and computation load of chemistry and gas-aerosol equilibrium (Bauer et al., 2007; Thornhill et al., 2021; Adams et al., 2001; Regayre et al., 2018)). Previous studies have highlighted the importance of nitrogen aerosol particles for climate, air quality and ecosystem impacts (e.g., Adams et al., 2001; Bauer et al., 2007, 2016; Kanakidou et al., 2016; Baker et al., 2021). Changes in nitrogen aerosol emissions are likely to follow different future trajectories than SO_4^{2-}, BC or OC, whose anthropogenic sources are mostly fossil fuel derived and should decrease in the future as renewable energy resources expand (Gidden et al., 2019). Nitrogen aerosol particles have substantial sources from agriculture, which is likely to stay constant or expand (Gidden et al., 2019; Klimont et al., 2017; Bauer et al., 2016). This suggests there could be a substantial bias in both historical and future aerosol forcings due to the lack of inclusion of these important sources (e.g., Bauer et al., 2007; Thornhill et al., 2021; Adams et al., 2001; Regayre et al., 2018).

4. Data availability: The data compiled here is available as a csv table with citations as well as gridded datasets with the modelled data in netcdf format at https://zenodo.org/records/10459654, Mahowald et al., 2024.

5. Code availability: The model used here is a version of the Community Earth System Model, and the modifications and input files to that code are available at https://zenodo.org/records/10459654, Mahowald et al., 2024.

5. Conclusions

In this study, we present a new aerosol compilation (AERO-MAP) specifically designed for evaluating Earth system and air quality models. This annual averaged dataset includes both total mass and composition, where available, including 12,000 station datasets and over 10 million daily averaged measurements. Here we expand beyond the usual biased coverage of only North America and Europe to present a more global data view for both $PM_{2.5}$ and PM_{10} (Fig. 1). Unfortunately, there are still very limited data characterizing both the surface concentration and composition of aerosol particles (Fig. 7). While satellite remote sensing can indicate the total atmospheric loading, it cannot yet provide substantial information about the size or composition of aerosol particles (Kahn et al., 2005; Tanré et al., 1997; Remer et al., 2005). Surface based remote sensing may provide more information about size and absorption properties (Holben et al., 1991; Dubovik et al., 2002; Schuster et al., 2016; Goncalves et al., 2023; Obiso et al., 2023). But knowing the size and the composition, which is key to their impacts on air quality and climate (Mahowald et al., 2011) as well as their potential for future change (Gidden et al., 2019) is important. We also present a methodology to use this dataset to evaluate both mass and composition for intercomparison projects and improvements in air quality and Earth system models. Future studies will use this compiled dataset for more detailed studies including temporal variability of different compositions. The fidelity of the annual means provided by this study will depend upon the ability of the measurement networks to capture the observed multi-decadal increases and decreases of emission that vary between source regions and sectors (Quaas et al., 2022).

This study also highlights the importance of including all aerosol components into the models, and shows that in the CESM2, in many places there is between 10-60% of the aerosol particulate mass missing, largely due to lack of the nitrogen aerosol particles (Paulot et al. 2016; Adams et al., 1999; Thornhill et al., 2020) and the poorly understood agricultural dust aerosol particles (e.g., Ginoux et al., 2012). Because these aerosol particles are largely driven by agricultural sources and not fossil fuels, their concentrations will be hardly impacted by the transition to renewable energy and may increase if agricultural production expands with population Therefore, these aerosol particles represent important air quality and climate impacts that should be represented more accurately in future studies.

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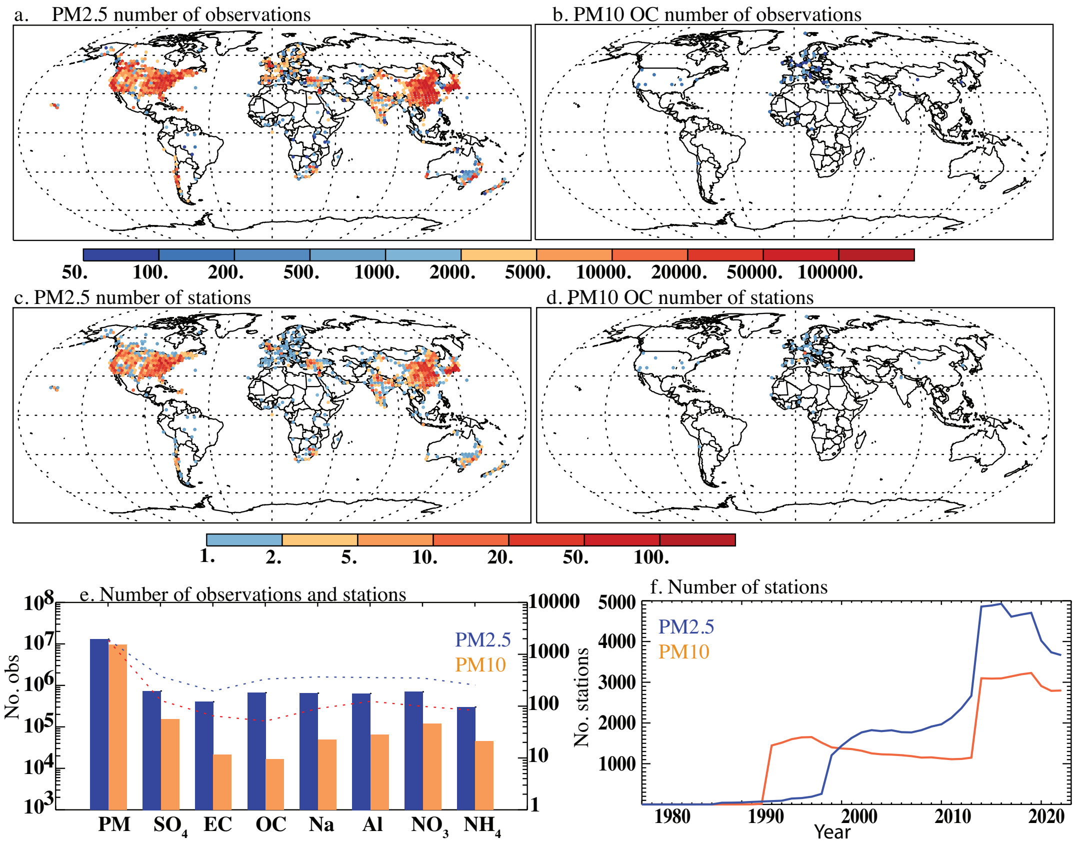

Figure 1: Distribution of observations in the data base, showing the number of observations of $PM_{2.5}$ (a) and PM_{10} Organic carbon (OC) (b) (with the colors indicating different numbers using the top color bar), as well as the number of stations within each 2x2 grid locations for $PM_{2.5}$ (c) and PM_{10} OC (d) (using the second color bar), showing that there is much more PM2.5 or PM_{10} data, in contrast to speciated data. e) The number of observations (bars) for total particulate matter (PM) or speciated data is summarized in for the $PM_{2.5}$ (blue) and PM_{10} (orange) fraction using the left hand side y-axis. The number of stations included in the study for each 2x2 grid box is shown as a dotted line (e) and uses the right hand size y-axis. f) The number of stations of PM2.5 (blue) and PM10 (orange) for each year is shown. The stations included derive from the following sources (see supplemental dataset for more details): (Alastuey et al., 2016; Almeida et al., 2005; Amato et al., 2016; Andreae et al., 2002;Arimoto et al., 2003; Artaxo et al., 2002; Barkley et al., 2019; Barraza et al., 2017; Bergametti et al., 1989; Bouet et al., 2019; 2021; Bozlaker et al., 2013; Chen et al., 2006; Chuang et al., 2005; Cipoli et al., 2023; Cohen et al., 2004; da Silva et al., 2008; Dongarrà et al., 2007, 2010; Formenti et al., 2003; Fuzzi et al., 2007; Hand et al., 2017; Heimburger et al., 2012; Herut and Krom, 1996; Herut et al., 2001; Hsu et al., 2016; Hueglin et al., 2005;

https://doi.org/10.5194/essd-2024-1
Preprint. Discussion started: 6 February 2024

Open Access Earth System Science Data Discussions

Furu et al., 2022, 2015; Gianini et al., 2012a, b; Kalivitis et al., 2007; Kaly et al., 2015; Kubilay et al., 2000; Kyllönen et al., 2020; Laing et al., 2014b, a; Lucarelli et al., 2014; 2019; Mackey et al., 2013; Maenhaut et al., 1996c, a, b, 1997a, b; 1999, 2000a, 2000b, 2002a,b,2005;, 2008, 2011; Maenhaut and Cafmeyer, 1998; Malm et al., 2007; Marticorena et al., 2010; Mihalopoulos et al., 1997; Mirante et al., 2009, 2013; Mkoma, 2008, 2009; Morera-Gómez et al., 2018, 2019; Nava et al., 2015, 2020; Nyanganyura et al., 2007; Oliveira et al., 2009, 2010; Pérez et al., 2008; Pio et al., 2023; Prospero et al., 1989, 2012, 2020; Prospero, 1996, 1999; Putaud et al., 2004, 2010; Rajot et al., 2010a, 2010b; Rodríguez et al., 2011, 2015; Salma et al., 1997; Savoie et al., 1993; Silva et al., 2010; Smichowski et al., 2004; Swap et al., 1992; Tørseth et al., 2012; Uematsu et al., 1983; Vanderzalm et al., 2003; Virkkula et al., 1999; Xiao et al., 2014; Zihan and Losno, 2016. Data from several online networks are also included (see Supplemental dataset for more details): e.g. https://www.airnow.gov/international/us-embassies-and-consulates/, https://quotsoft.net/air/, https://app.cpcbccr.com/ccr/#/caaqm-dashboard-all/caaqm-landing/data, https://sinca.mma.gob.cl/index.php/. https://tenbou.nies.go.jp/download/

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Preprint. Discussion started: 6 February 2024

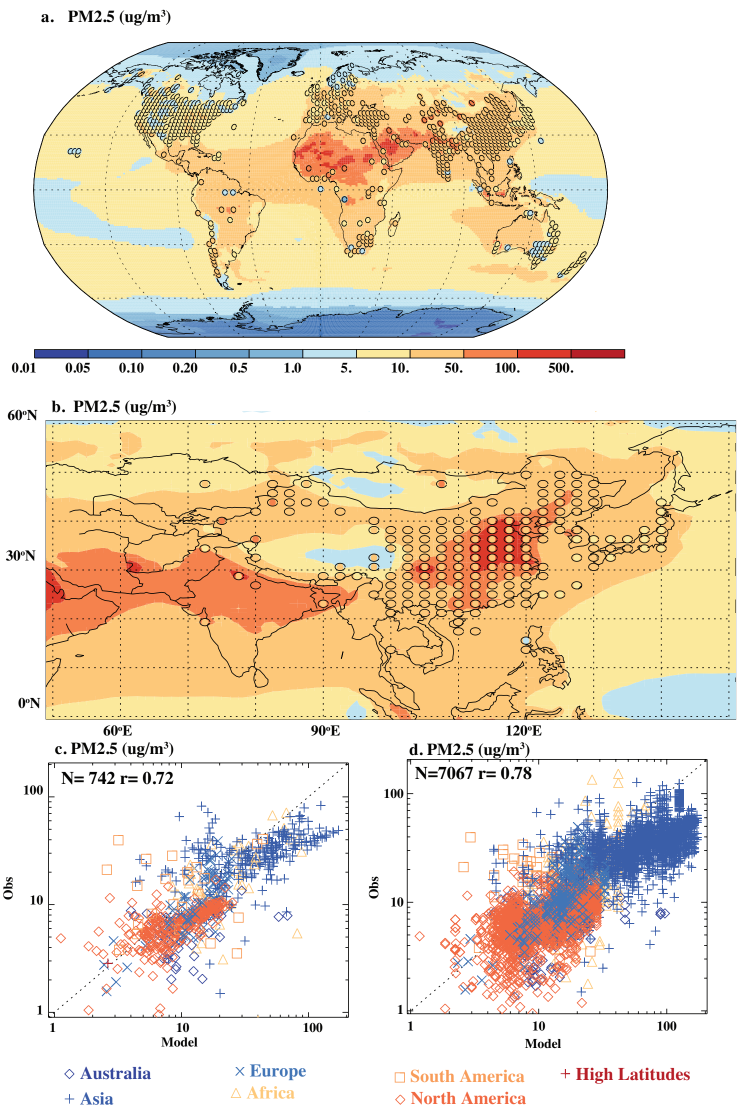


Figure 2: Model results and gridded observations for $PM_{2.5}$ in µg/m^3 spatially mapped globally (a) and focused on just East Asia (b) where the model is plotted as the background and the observations are circles with the colors indicating the amount of $PM_{2.5}$ using the same scale. A comparison of the model (x-axis) to the observations (y-axis) is shown for the gridded data (c) and including all stations (d). In the scatter plots, the colors and symbols indicate the regions. More statistics are shown in Table S4, and the model plotted alone is available in Figure S2.

https://doi.org/10.5194/essd-2024-1
Preprint. Discussion started: 6 February 2024

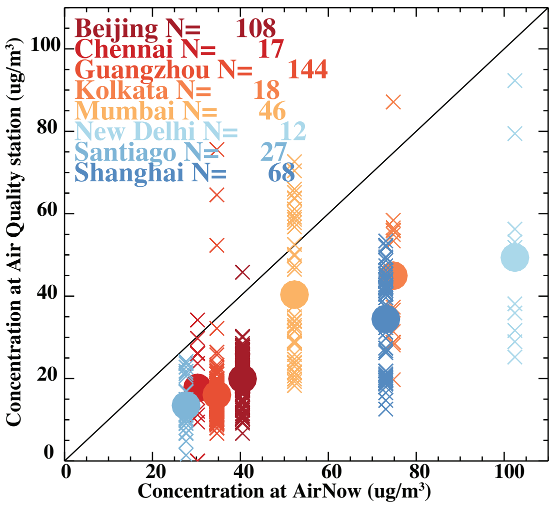
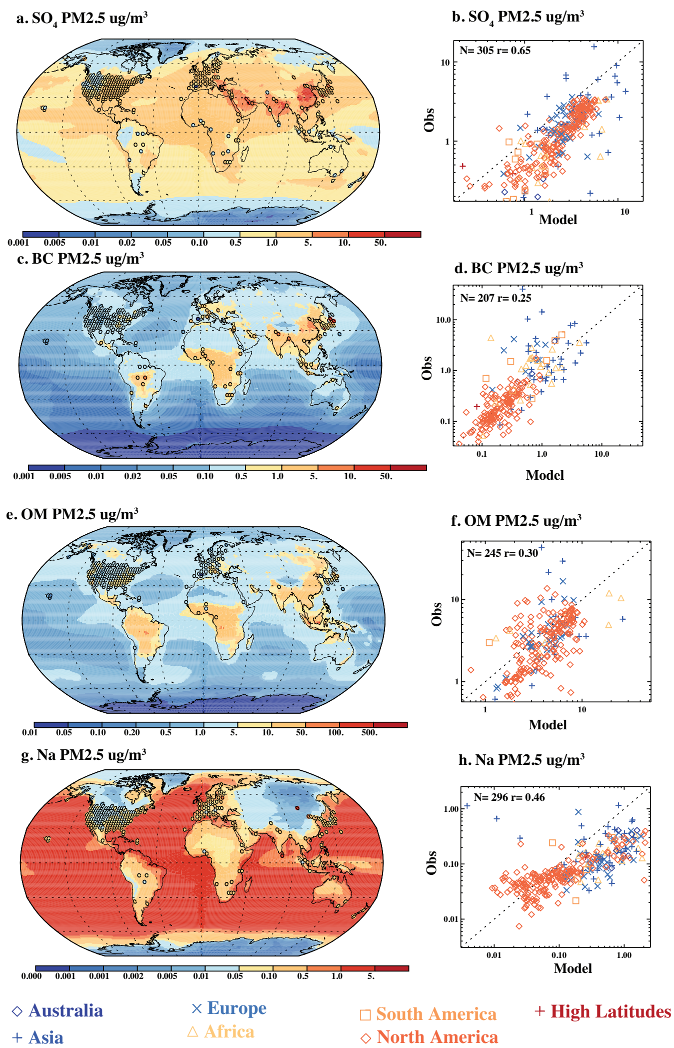


Figure 3: Comparison of $PM_{2.5}$ observations from the US Embassy's AirNow network (**https://www.airnow.gov/international/us-embassies-and-consulates/**) versus observations from the Chinese air quality network (downloaded from **https://quotsoft.net/air/**) (Beijing 39.9N 116.4E, Guangzhou 23N 113E, Shanghai 31N 121E) and the Indian (Chennai 13N 80E, Kolkata 23N 88E, New Delhi 27N 77E) network (**https://app.cpcbccr.com/ccr/#/caaqm-dashboard-all/caaqm-landing/data**); and observations (Barraza et al., 2017) from Santiago, Chile (23.7S 70.4W) against the Chilean air quality network (https://sinca.mma.gob.cl/index.php/). The numbers after each city name are the number of stations found within 1° distance of the AirNow (or Chile observations) station.

https://doi.org/10.5194/essd-2024-1
Preprint. Discussion started: 6 February 2024

Open Access Earth System Science Data Discussions



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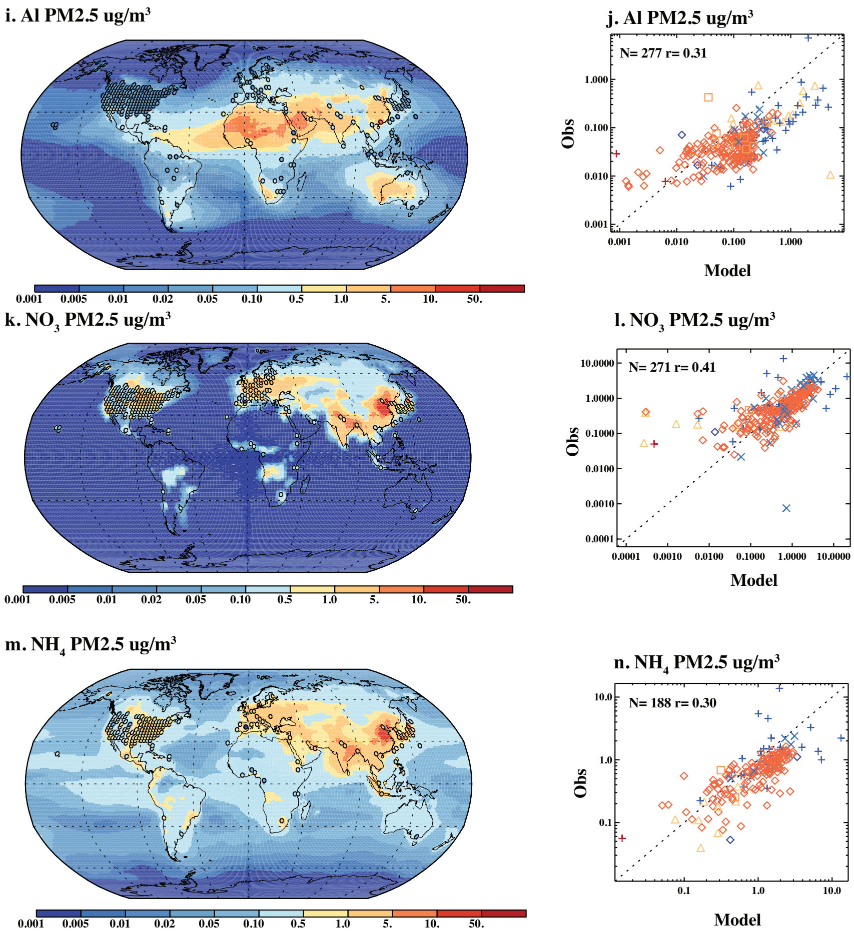

Figure 4: Model results and gridded observations for different types of $PM_{2.5}$ in μg/m^3 spatially mapped globally where the model is plotted as the background and the observations are circles with the colors indicating the amount $PM_{2.5}$ using the same scale for (a) SO_4^{2-}, (c) BC (black carbon), (e) OM (organic material=1.8 times organic carbon (OC)), (g) Na, (i) Al, (k) NO_3^-, (m) NH_4^+. A scatter plot comparison of the model (x-axis) to the observations (y-axis) is shown for the gridded observational data for for (b) SO_4^{2-}, (d) BC (f) OM, (h) Na, (j) Al, (l) NO_3^-, (n) NH_4^+. In the scatter plots, the colors and symbols indicate the regions using the same legend as Figure 2. More statistics are shown in Table S4, and the model plotted alone is available in Figure S2.

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Preprint. Discussion started: 6 February 2024

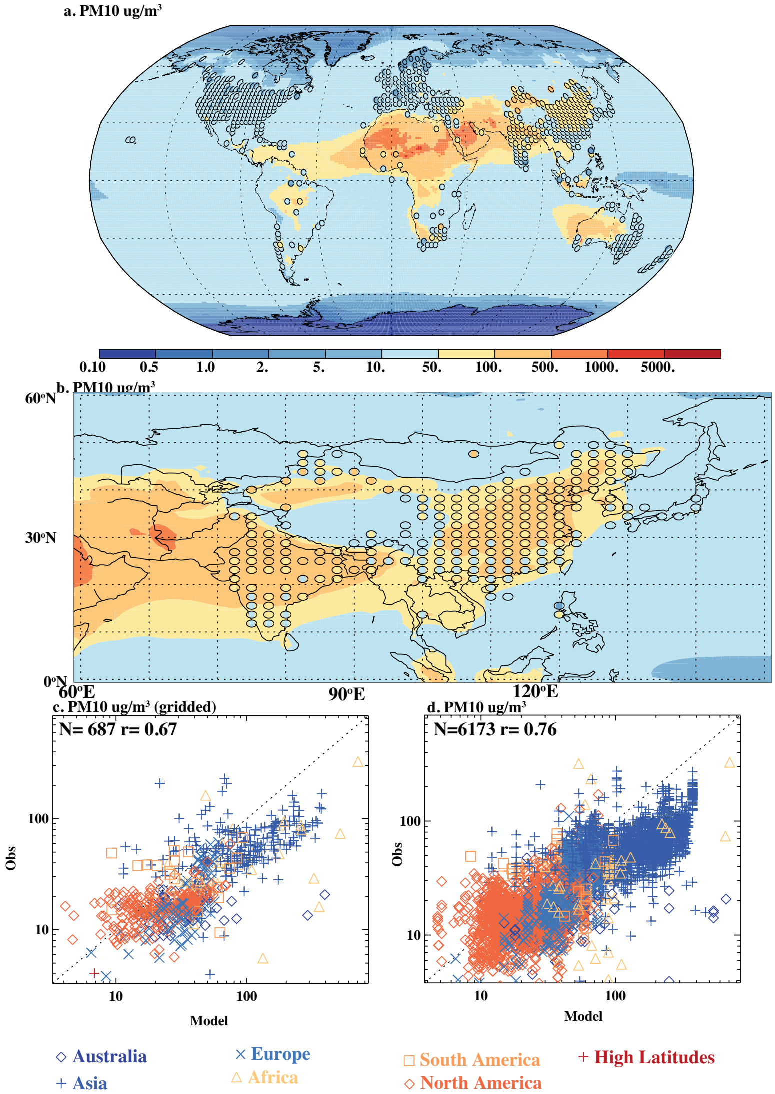
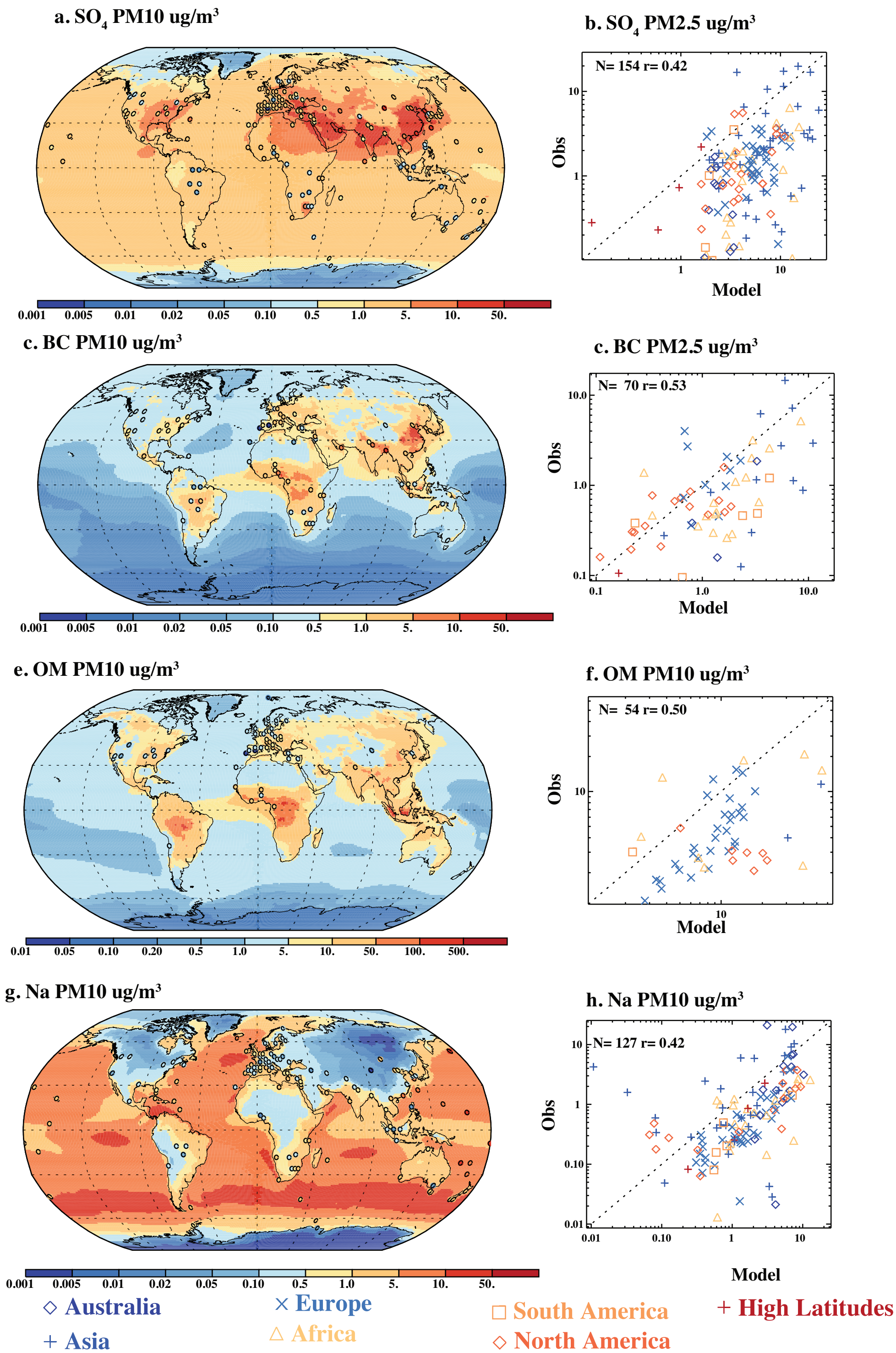


Figure 5: Model results and gridded observations for PM_{10} in µg/m^3 spatially mapped globally (a) and focused on just East Asia (b) where the model is plotted as the background and the observations are circles with the colors indicating the amount of $PM_{2.5}$ using the same scale. A comparison of the model (x-axis) to the observations (y-axis) is shown for the gridded data (c) and including all stations (d). In the scatter plots, the colors and symbols indicate the regions. More statistics are shown in Table S5, and the model plotted alone is available in Figure S3.

https://doi.org/10.5194/essd-2024-1
Preprint. Discussion started: 6 February 2024

Open Access Earth System Science Data Discussions



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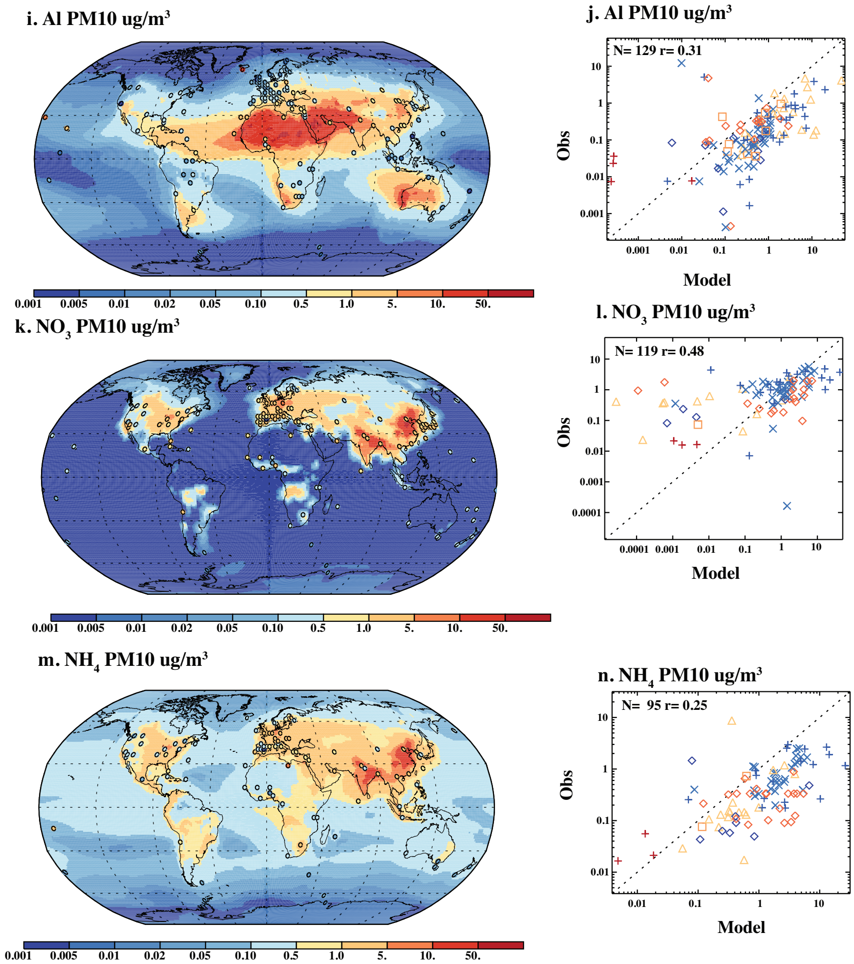

Figure 6: Model results and gridded observations for different types of PM_{10} in µg/m^3 spatially mapped globally where the model is plotted as the background and the observations are circles with the colors indicating the amount PM_{10} using the same scale for (a) SO_4^{+2}, (c) BC (black carbon), (e) OM (organic material=1.8times organic carbon (OC)), (g) Na, (i) Al, (k) NO_3^-, (m) NH_4^+. A scatter plot comparison of the model (x-axis) to the observations (y-axis) is shown for the gridded observational data for (b) SO_4^2, (d) BC (f) OM, (h) Na, (j) Al, (l) NO_3^-, (n) NH_4^+. In the scatter plots, the colors and symbols indicate the regions using the same legend as Figure 2. More statistics are shown in Table S5, and the model plotted alone is available in Figure S3.

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Preprint. Discussion started: 6 February 2024

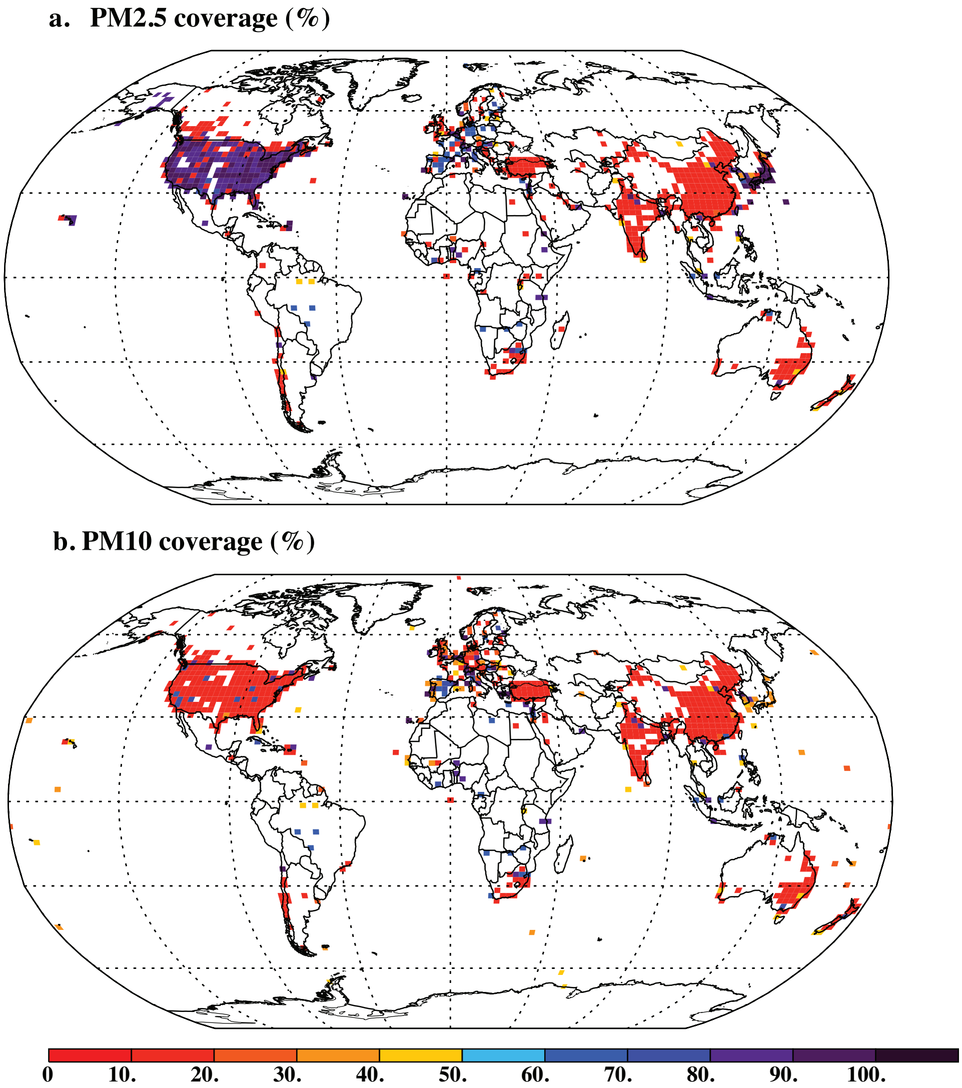

Figure 7: Observational coverage (%) for gridded observations, showing within each grid box (2x2) the % of the constituents that are measured assuming that PM, SO_4^{2-}, BC, OM, Na, Al, NO_3^-, and NH_4^+ are required to constrain the PM distribution for (a) $PM_{2.5}$ and (b) PM_{10}.

https://doi.org/10.5194/essd-2024-1
Preprint. Discussion started: 6 February 2024

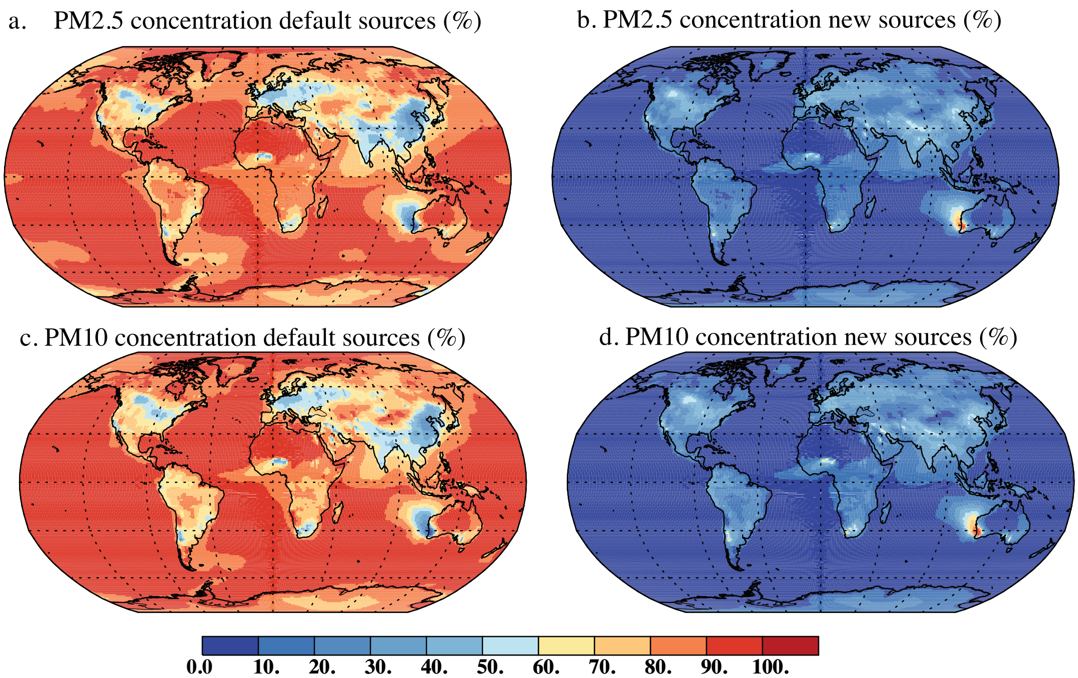

Figure 8: Modelled estimates of what percent of the surface concentration of $PM_{2.5}$ is considered in the default CAM (a) or is new in this study (b). Similarly PM_{10} is shown for the default model (c) and new sources in this study (d).

https://doi.org/10.5194/essd-2024-1
Preprint. Discussion started: 6 February 2024

Open Access Earth System Science Data Discussions

Table 1: Global Aerosol Budgets

Global deposition (Tg/year), percentage of aerosol that is $PM_{2.5}$, and globally and annually averaged surface concentration (µg/m^3) and aerosol optical depth for each of the sources used in the model. An asterisk indicates that there are additions to the model from the default CAM6.

	PM_{10}	$PM_{2.5}$		
	Deposition (Tg/year)	%	Conc (µg/m^3)	AOD (unitless)
Sulfate	121	100	2.1	0.018
Black carbon	10	100	0.5	0.009
Primary organic aerosol	34	100	1.6	0.008
Secondary organic aerosol	37	100	1.0	0.007
Sea salts	2520	3	13.0	0.045
Dust	2870	1	19.4	0.030
NH_4NO_3*	20	100	0.4	0.013
Agricultural Dust*	585	1	3.7	0.006
Road*	0.43	79	0.02	0.0000
Coarse organic carbon*	4	0.0	0.04	0.0000
Coarse black carbon*	0.35	0.0	0.00	0.0000
Fine and coarse inorganic industrial matter *	56	46	1.2	0.0018
Bacteria and Fungi spores from land*	4	0	0.04	0.0000
Other primary biogenic	54	3	0.4	0.0005

https://doi.org/10.5194/essd-2024-1
Preprint. Discussion started: 6 February 2024

particles from land*				
Marine organic aerosols	44	99	0.6	0.0008