

# AERO-MAP: A data compilation and modelling approach to understand spatial variability in fine and coarse mode aerosol composition

Natalie M. Mahowald<sup>1</sup>, Longlei Li<sup>1</sup>, Julius Vira<sup>2</sup>, Marje Prank<sup>2</sup>, Douglas S. Hamilton<sup>3</sup>, Hitoshi Matsui<sup>4</sup>, Ron L. Miller<sup>5</sup>, P. Louis Lu<sup>1,6</sup>, Ezgi Akyuz<sup>7</sup>, Daphne Meidan<sup>1</sup>, Peter Hess<sup>8</sup>, Heikki Lihavainen<sup>9</sup>, Christine Wiedinmyer<sup>10</sup>, Jenny Hand<sup>11</sup>, Maria Grazia Alaimo<sup>12</sup>, Célia Alves<sup>13</sup>, Andres Alastuey<sup>14</sup>, Paulo Artaxo<sup>15</sup>, Africa Barreto<sup>16</sup>, Francisco Barraza<sup>17</sup>, Silvia Becagli<sup>18</sup>, Giulia Calzolari<sup>18</sup>, Shankaraman Chellam<sup>19</sup>, Ying Chen<sup>20</sup>, Patrick Chuang<sup>21</sup>, David D. Cohen<sup>22</sup>, Cristina Colombi<sup>23</sup>, Evangelia Diapouli<sup>24</sup>, Gaetano Dongarra<sup>12</sup>, Konstantinos Eleftheriadis<sup>24</sup>, Johann Engelbrecht<sup>25</sup>, Corinne Galy-Lacaux<sup>26</sup>, Cassandra Gaston<sup>27</sup>, Dario Gomez<sup>28</sup>, Yenny González Ramos<sup>29,16</sup>, Roy M. Harrison<sup>30</sup>, Chris Heyes<sup>31</sup>, Barak Herut<sup>32,33</sup>, Philip Hopke<sup>34,35</sup>, Christoph Hüglin<sup>36</sup>, Maria Kanakidou<sup>37,38,39</sup>, Zsófia Kertesz<sup>40</sup>, Zbigniew Klimont<sup>31</sup>, Katriina Kyllönen<sup>2</sup>, Fabrice Lambert<sup>41,42</sup>, Xiaohong Liu<sup>43</sup>, Remi Losno<sup>44</sup>, Franco Lucarelli<sup>18</sup>, Willy Maenhaut<sup>45</sup>, Beatrice Marticorena<sup>46</sup>, Randall V. Martin<sup>47</sup>, Nikolaos Mihalopoulos<sup>37,48</sup>, Yasser Morera-Gómez<sup>49</sup>, Adina Paytan<sup>50</sup>, Joseph Prospero<sup>26</sup>, Sergio Rodríguez<sup>51,16</sup>, Patricia Smichowski<sup>28</sup>, Daniela Varrica<sup>12</sup>, Brenna Walsh<sup>47</sup>, Crystal Weagle<sup>48</sup>, Xi Zhao<sup>43</sup>

<sup>1</sup>Department of Earth and Atmospheric Sciences, Cornell University, Ithaca, NY, 14853, USA

<sup>2</sup>Finnish Meteorological Institute, Helsinki, Finland

<sup>3</sup>Department of Marine, Earth and Atmospheric Sciences, North Carolina State, Raleigh, NC, USA

<sup>4</sup>Graduate School of Environmental Studies, Nagoya University, Nagoya, Japan 464-8601

<sup>5</sup>National Aeronautics and Space Administration, Goddard Institute for Space Studies, Columbia University, NY, NY 10025

<sup>6</sup>Earth and Climate Section, Nicholas School of the Environment, Duke University, Durham, NC, 27708, USA

<sup>7</sup>Eurasia Institute of Earth Sciences, Istanbul Technical University, 34467 Istanbul, Turkey

<sup>8</sup>Department of Biological and Environmental Engineering, Cornell University, Ithaca NY, USA

<sup>9</sup>SIOS Knowledge Centre, Postboks 156, 9171 Longyearbyen, Norway

<sup>10</sup>Cooperative Institute for Research in Environmental Sciences at the University of Colorado Boulder, Boulder, CO, USA

<sup>11</sup>Cooperative Institute for Research in the Atmosphere, Colorado State University, Fort Collins, CO, USA,

<sup>12</sup>Dip. Scienze della Terra e del Mare, University of Palermo, Italy

<sup>13</sup>Centre for Environmental and Marine Studies (CESAM), Department of Environment, University of Aveiro, 3810-193, Aveiro, Portugal

<sup>14</sup>Institute of Environmental Assessment and Water Research (IDAEA-CSIC), 08034, Barcelona, Spain

<sup>15</sup>Instituto de Fisica, Universidade de Sao Paulo, 05508-090, Sao Paulo, SP, Brazil

- 30 <sup>16</sup> Izaña Atmospheric Research Center (IARC), Agencia Estatal de Meteorología (AEMET), Santa Cruz de Tenerife, Spain
- 31 <sup>17</sup> Saw Science, Invercargill, New Zealand
- 32 <sup>18</sup> Department of Physics and Astronomy, Universita di Firenze and INFN-Firenze, 50019 Sesto Fiorentino, Italy
- 33 <sup>19</sup> Department of Civil & Environmental Engineering, Texas A&M University, College Station, TX 77843-3136, USA
- 34 <sup>20</sup> Dept. Environ. Sci. Engr. Fudan University Jiangwan Campus 2005 Songhu Road, Shanghai, China
- 35 <sup>21</sup> Earth & Planetary Sciences Department, Institute of Marine Sciences, University of California, Santa Cruz, CA, 95064 ,
- 36 USA.
- 37 <sup>22</sup> Centre for Accelerator Science, Australian Nuclear Science and Technology Organisation, New Illawarra Rd, Lucas
- 38 Heights, NSW, Australia
- 39 <sup>23</sup> Environmental Monitoring Sector, Arpa Lombardia, Via Rosellini 17, 20124 Milan, Italy
- 40 <sup>24</sup> Environmental Radioactivity & Aerosol Technology for Atmospheric & Climate impact Lab, INRaSTES, N.C.S.R.
- 41 Demokritos, 15341 Ag. Paraskevi, Attiki, Greece
- 42 <sup>25</sup> Desert Research Institute (DRI), 2215 Raggio Parkway, Reno, Nevada 89512-1095
- 43 <sup>26</sup> Laboratoire d'Aerologie, Universite de Toulouse, CNRS, Observatoire Midi Pyrenees, Toulouse, France
- 44 <sup>27</sup> Rosenstiel School of Marine and Atmospheric Science, University of Miami, Miami, FL, 33149, US
- 45 <sup>28</sup> Comision Nacional de Energia Atomica, Gerencia Química, Av. Gral Paz 1499, B1650KNA, San Martin, Buenos Aires,
- 46 Argentina
- 47 <sup>29</sup> Scientific Department, CIMEL, Paris, France.
- 48 <sup>30</sup> School of Geography, Earth and Environmental Sciences, University of Birmingham, Edgbaston, Birmingham B15 2TT,
- 49 United Kingdom
- 50 <sup>31</sup> Energy, Climate and Environment Program, International Institute for Applied Systems Analysis, 2361 Laxenburg,
- 51 Austria
- 52 <sup>32</sup> Israel Oceanographic & Limnological Research, Tel Shikmona, Haifa, 31080, Israel
- 53 <sup>33</sup> University of Haifa, Haifa, 3103301, Israel
- 54 <sup>34</sup> Clarkson University, Potsdam, NY, USA,
- 55 <sup>35</sup> Department of Public Health Sciences, University of Rochester School of Medicine and Dentistry, Rochester, NY, USA,
- 56 <sup>36</sup> Swiss Federal Laboratories for Materials Science and Technology(EMPA), CH-8600 Duebendorf, Switzerland
- 57 <sup>37</sup> Environmental Chemical Processes Laboratory (ECPL), Department of Chemistry, University of Crete, Heraklion, Greece.
- 58 <sup>38</sup> Center of Studies of Air quality and Climate Change, Institute for Chemical Engineering Sciences, Foundation for
- 59 Research and Technology Hellas, Patras, Greece.
- 60 <sup>39</sup> Excellence Chair, Institute of Environmental Physics, University of Bremen, Bremen, Germany
- 61 <sup>40</sup> HUN-REN Institute for Nuclear Research (ATOMKI), Debrecen, Hungary
- 62 <sup>41</sup> Geography Institute, Pontificia Universidad Catolica de Chile, Santiago, 7820436, Chile
- 63 <sup>42</sup> Center for Climate and Resilience Research, Santiago, Chile
- 64 <sup>43</sup> Department of Atmospheric Sciences, Texas A&M University, College Station, TX 77843
- 65 <sup>44</sup> Institut de Physique du Globe de Paris, Universite de Paris, Paris, France
- 66 <sup>45</sup> Department of Chemistry, Ghent University, Gent, Belgium
- 67 <sup>46</sup> Laboratoire Interuniversitaire des Systemes Atmospheriques (LISA), Universit«es Paris Est-Paris Diderot-Paris 7, UMR
- 68 CNRS 7583, Cr«eteil, France
- 69 <sup>47</sup> Energy, Environmental and Chemical Engineering, Washington University, St. Louis, MO, USA.
- 70 <sup>48</sup> Institute for Environmental Research and Sustainable Development, National Observatory of Athens, Pendeli, Greece

<sup>49</sup> Universidad de Navarra, Instituto de Biodiversidad y Medioambiente BIOMA, Irunlarrea 1, 31008, Pamplona, España

<sup>50</sup> Earth and Planetary Science, University of California, Santa Cruz, CA, USA

<sup>51</sup> Consejo Superior de Investigaciones Científicas, IPNA CSIC, Tenerife, Canary Islands, Spain.

*Correspondence to:* Natalie M. Mahowald (mahowald@cornell.edu)

**Abstract.** Aerosol particles are an important part of the Earth-climate system, and their concentrations are spatially and temporally heterogeneous, as well as variable in size and composition. Particles can interact with incoming solar radiation and outgoing long wave radiation, change cloud properties, affect photochemistry, impact surface air quality, change the albedo of snow and ice, and modulate carbon dioxide uptake by the land and ocean. High particulate matter 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<sub>2.5</sub> and PM<sub>10</sub> surface observations, including measurements of aerosol composition, focusing on the spatial variability across different observational stations. Climate modelers are constantly looking for multiple independent lines of evidence to verify their models, and in situ surface concentration measurements, taken at the level of human settlement, present a valuable source of information about aerosols and their human impacts that are complementary to the column averages or integrals often retrieved from satellites. We demonstrate a method for comparing the datasets to output from global climate models that are the basis for projections of future climate and large-scale aerosol transport patterns that influence local air quality. Annual trends and seasonal cycles are discussed briefly and included in the compilation. Overall, most of the planet or even the land fraction does not have sufficient observations of surface concentrations, and especially particle composition, to characterize and understand the current distribution of particles. Climate models without ammonium nitrate aerosols omit ~10% of the global average mass of aerosol particles in both PM<sub>2.5</sub> and PM<sub>10</sub> size fractions, with up to 50% of the particles not included in some regions. In these regions, climate model aerosol forcing projections are likely to be incorrect, as they do not include important trends in short lived climate forcers.

## 1 Introduction

Intergovernmental Panel on Climate Change (IPCC) reports (IPCC, 2021; Gulev et al., 2021; Szopa et al., 2021) and other community assessments have highlighted the role of uncertainties in human-induced changes to aerosol concentration and composition in limiting our ability to project future climate. Aerosol particles are also a major contributor to air pollution, which reduces life expectancy and quality of life (Burnett et al., 2018). Aerosol particles are suspended liquids or solids in the atmosphere originating from diverse natural and anthropogenic sources and composed of a wide variety of chemicals

(e.g., sea salts, dust, sulfate, nitrate, black carbon, organic carbon). 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). Satellite remote sensing retrievals provide important information about the temporal and spatial distribution of aerosol particles, but challenges remain in quantifying the aerosol size and chemical composition (Kahn et al., 2005; Tanré et al., 1997; Remer et al., 2005, Castellanos et al., 2024). In addition, the AERONET surface remote sensing network provides some information about loading, size and absorbing aerosol properties related to composition (Holben et al., 2001; Dubovik et al., 2002; Schuster et al., 2016; Gonçalves Ageitos et al., 2023; Obiso et al., 2023). Both the magnitude of the aerosol effects on climate, and sometimes their sign, are dependent on the composition and size of particles (Mahowald et al., 2011, 2014a; Bond et al., 2013; IPCC, 2021). In addition, one cannot understand the impact of humans on aerosol particles without identifying the sources of particles, which determine their chemical composition. Obtaining information about the composition and size of particles in many cases requires in situ observations, which are often 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; 2012; Gliš et al., 2021) or knowledge of the aerosol impacts like cloud condensation nucleation (Laj et al., 2020; Fanourgakis et al., 2019). However, most of the available data comes from North America and Europe (e.g., Szopa et al., 2021; Reddington et al., 2017). In addition, previous compilation studies have focused primarily on understanding fine aerosol particles (here defined as particles with a diameter less than 2.5  $\mu\text{m}$ ) and improving model simulation of these particles, because of their importance for air quality, respiratory health, cloud interactions and short-wave forcing (Collaud Coen et al., 2020; Bellouin et al., 2020; Fanourgakis et al., 2019; Reddington et al., 2017). Coarse mode particles (defined as those particles with a diameter larger than 2.5  $\mu\text{m}$ ) are important for long wave radiation interactions, cloud seeding and for biogeochemistry, but 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 aerodynamic diameter less than 2.5  $\mu\text{m}$  or  $\text{PM}_{2.5}$ ), there are fewer studies focusing on aerosol compilations for both fine and coarse particles, and their comparison to models (Kok et al., 2014b; Albani et al., 2014b; Huneeus et al., 2011; Gliš et al., 2021; Kok et al., 2021). Nonetheless, there are many observations of the coarse particle mass with diameter less than 10  $\mu\text{m}$  ( $\text{PM}_{10}$ ) (e.g., Hand et al., 2017), and most climate models include these particles (e.g., Huneeus et al., 2011). Compilations of in situ data are available for dust and iron 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  $\text{PM}_{10}$  or coarse and fine particles are available for many regions

136 and individual sites (e.g., Malm et al., 2007; Hand et al., 2019; Maenhaut and Cafmeyer, 1998; Artaxo and Maenhaut, 1990;  
137 McNeill et al., 2020) but have not previously been compiled into one database that would facilitate the evaluation of global  
138 climate models that are an important tool for projections of future climate change, air quality and their impacts upon human  
139 society. Aerosol modelers need as much information as possible about the observed composition of the particles and their  
140 transport. Thus, there is a need to compile both PM<sub>2.5</sub> and PM<sub>10</sub> in situ concentration data into one database to make it easy  
141 for modellers to compare global model results with observations. One goal the aerosol community should work towards is  
142 making aerosol measurement datasets publicly and conveniently available, while acknowledging the principal investigators  
143 who produced these datasets, which we hope this paper serves as a step towards achieving.

144 The current generation of Earth system models used for the IPCC simulations tends to include the dominant aerosol species  
145 (desert dust, sea spray, black carbon (BC), organic matter (OM) and sulfate) while omitting other potentially important  
146 aerosol constituents. For example, some Earth system models ignore ammonium nitrate particles although these are known  
147 to be important for climate and biogeochemistry, and are impacted by human activities (Paulot et al., 2016; Adams et al.,  
148 1999; Thornhill et al., 2020). In this study, we use available observations to compare to a global model estimate of the total  
149 PM<sub>10</sub> and PM<sub>2.5</sub>, and deduce the importance of these often-neglected aerosol species. We also propose a method for  
150 comparing species that are often not directly measured (such as dust or sea salts) using their elemental composition. Note  
151 that we exclude super coarse (>PM<sub>10</sub>) particles here because of the sparsity of available measurements, although studies have  
152 suggested their importance for climate interactions (e.g., Adebyi et al., 2023).

153  
154 Climate modelers are constantly looking for multiple independent lines of evidence to verify their models, and in situ surface  
155 concentration data presents a valuable source of information about aerosols often near human society. Understanding spatial  
156 variability in aerosols, and the composition of those aerosols is key to understanding how aerosols in different regions have  
157 evolved in the past, and how they will evolve in the future. Some regions are dominated by fossil fuel derived aerosols, which  
158 may have peaked in magnitude, even as greenhouse gas concentrations continue to increase, while in other regions aerosols  
159 are driven by agriculture or by natural aerosols (Bauer et al., 2016; Turnock et al., 2020; Kok et al., 2023). In addition, different  
160 aerosol species have different impacts on climate: for example, knowing whether aerosols are scattering or absorbing changes  
161 the sign of the interaction (Li et al, 2022). Some aerosols also serve as better cloud or ice nuclei than others, while  
162 biogeochemical impacts are very sensitive to composition (Mahowald et al., 2011). Knowing even the order of magnitude in  
163 regions with aerosols (e.g., contrasting 0.1 to 0.001) is important for aerosol-cloud interactions that can be non-linear especially  
164 at low aerosol levels (Carslaw et al., 2013). Having surface concentration observational dataset with large spatial coverage  
165 based on independent data can be valuable for aerosol model comparisons, especially for models with a global domain. We  
166 focus most of this paper on the spatial distribution of climatological mean, as that is easily obtained from models, and the most  
167 important variable for many climate impacts like radiative effects or aerosol-cloud interactions, except for aerosols dispersed  
168 by large infrequent events (e.g., Clark et al., 2015; Fasullo et al., 2022). Since aerosols are thought to cause between 2 and 10

million deaths per year (Landrigan et al., 2018; Lelieveld et al., 2019; Murray et al., 2020; Vohra et al., 2021), understanding and being able to model correctly the annual mean aerosol concentrations in the surface layer is vital and thus this dataset provides valuable information towards understanding aerosol contributions to mortality. Nonetheless, there have been trends in emissions especially of anthropogenic aerosols over the last 40 years (Quaas et al., 2022; Bauer et al., 2022), and we consider these as well.

For this study we focus on the following: a) identifying and compiling available PM<sub>2.5</sub> and PM<sub>10</sub> aerosol data, including aerosol composition, into a new publicly available database (AERO-MAP) for the modelling community across as much of the globe as possible; b) presenting a methodology to compare the spatial distribution of the climatological mean observations to the aerosols in an Earth system model; c) briefly present some temporal trends and comparisons available from this dataset and d) identifying the measurement and modelling gaps from this comparison. While our model evaluation is not exhaustive, we hope that the convenience of this observational compilation enables an expanding and more thorough set of comparisons by future investigators.

## 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. Datasets were identified by advertising at international meetings (Wiedinmyer et al., 2018), searching the literature, contacting principal investigators and accessing publicly available datasets. 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<sub>2.5</sub> and PM<sub>10</sub> 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<sub>2.5</sub> and coarse (PM<sub>2.5</sub> to PM<sub>10</sub>) aerosols. For those sites, we convert the latter to PM<sub>10</sub> 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 (e.g., field data: these are identified as station datasets with less than one year of data in supplemental datasets). In some poorly measured regions, we include total suspended particles (TSP) datasets (information on the size fraction measured is in the Supplemental dataset). The time period for different datasets is included in the supplemental dataset 1.

Detailed studies have shown that PM<sub>10</sub> and PM<sub>2.5</sub> 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<sub>10</sub> and PM<sub>2.5</sub>, since our model and most models only consider mass up to PM<sub>10</sub>. 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., 2014; 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 (Heimbürger 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<sub>10</sub>, because of the lack of size-resolved data. Data from the Japanese air quality network use a different inlet for the PM<sub>10</sub> 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<sub>10</sub> and PM<sub>2.5</sub> 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 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 the measurement methods and types are shown in Table 1 and vary between measurements of fine and coarse, versus PM<sub>2.5</sub> and PM<sub>10</sub>, with different measurement types for elemental and chemical analysis (Table 1). Details on how the model is compared to data for different elements are in Section 2.3.

For this paper, we focus on the climatological means for 1986-2023 and decadal means for 2010-2019. The first period is chosen as the full duration of the individual data sets comprising the compilation are available; the second is chosen to recognize decadal variations in anthropogenic emission within the longer period and isolate a particular decade when data is most plentiful. In addition, annual means for each year the data is available is also calculated, as well as the climatological monthly means. The temporal means 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 (Supplemental dataset 1).

Our goal is to create easy-to-use datasets for model-data comparisons. Included in this dataset are several files with different levels of description and analysis. One file provides traceability information, including a detailed citation, type and number of measurements included, as well as time period, climatological and decadal (2010-2019) means and standard deviations for each time period (Supplemental dataset 1). For each station dataset included in the database, there will be one line in this file. This means that for some stations (for example K-pushta), there are multiple lines in the supplemental file indicating the two different time periods where measurements were made as well as the two sizes that are measured during each time period. For each station dataset, there are latitude, longitudes, annual mean values, number of observations, year extent of the observations, standard deviations, etc, as well as the citation and where to obtain the data. There are also several netcdf files available at <https://zenodo.org/records/11391232> for this dataset. The most useful is likely to be the Allobervation.AEROMAP.nc file, which contains the same quantitative data for each station dataset as the supplement, except that the data is processed to be only PM<sub>2.5</sub> and PM<sub>10</sub> (with some TSP data in places with little data, as discussed above). That means PM<sub>2.5</sub> and coarse aerosol mass are added together if the station datasets are collocated to create a PM<sub>10</sub> dataset (e.g., see Table 1). In addition, this file contains climatological monthly means, and annual means for each year for each station dataset, so that temporal information is also easily available. Another file includes the climatological mean observations averaged up to a 2°×2° grid that is used for plotting the figures shown in the paper (Allobervation.AEROMAP.2x2.nc). As indicated in the data availability, only the time-means are available and the underlying data for some datasets cannot be openly published, but please contact the authors (identified by the citation) if other time periods are desired.

The location of each site is as accurate as possible and for most sites is accurate to less than 1km. Some datasets provided more limited information and those locations are accurate only to less than 10km (data downloaded from the following air quality networks: Mexico City: <http://www.aire.cdmx.gob.mx/default.php?opc=%27aKBh%27>, South Africa <https://saaqis.environment.gov.za/>, India: <https://app.cpcbcr.com/ccr/#!/caaqm-dashboard-all/caaqm-landing/data> and Chile: <https://sinca.mma.gob.cl/index.php/>).

## 2.2 Model description

Most of the 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°×1° 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 particles by nudging toward MERRA2 winds (Gelaro et al., 2017).

The model included prognostic dust, sea salts, BC, OM, and sulfate 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). The model includes separate primary and secondary organic species which are both emitted directly, but the primary organic and black carbon aerosols are allowed to age in the model from hydrophobic to hydroscopic, and their optical properties also change (Liu et al., 2016). The coarse mode is included for sulfate, dust and sea salts. For this study, the coarse size mode (mode 3) was returned to the size parameters used in the previous version of the model: CAM5 (geometric standard deviation of 1.8) to better simulate coarse mode particles, and improve the dry deposition scheme and optics used in the model for simulating coarse mode particles like dust as described in Li et al. (2022).

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 et al., 2022; Huang et al., 2021; Kok et al., 2017). Anthropogenic emissions of sulfate, OM, and BC follow the Climate Model Intercomparison Project 6 historical data for 2010 (Gidden et al., 2019). Emissions and mean concentrations for each of these constituents are included in Table 2.

### 2.2.1 Modelling of additional aerosol sources and types

Ammonium nitrate aerosol particles are not included in the standard CAM6, 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), so they are included in this study. Nitrate can also react with dust particles, for example, but that is ignored in this study (Dentener et al., 1996). Ammonium nitrate particles require tropospheric chemistry interactions because the nitrogen-containing particles are both a source and a sink for gaseous nitrogen species, which are key elements of tropospheric photochemistry and the 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  $2^\circ \times 2^\circ$  resolution and are linearly interpolated to  $1^\circ \times 1^\circ$  resolution used for the other modelled 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 model ammonium nitrate is compared to the observed nitrogenous aerosol optical depth. Ammonium nitrate is assumed to only form when there is surplus ammonium (and nitrate) after the ammonium sulfate is formed. 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 compared to nitrates, 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 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 (Vira et al. 2022).

### 2.3 Model-observation comparison methodology

Comparisons of the observations to model concentrations were done using BC, OC,  $\text{SO}_4^{2-}$ , Al,  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ , and Na composition measurements. Some of these elements/compounds map directly onto model constituents (BC, OC,  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ , and  $\text{NH}_4^+$ ), while others serve as proxies for modelled constituents (Al for dust, Na for sea salts, S for sulfate, etc.). We summarize the relationships used to obtain the values from the model (Table S1), and what observations are combined to include as much information as possible from the observations. (Table S2). We use non-sea-salt sulfate in ocean regions for estimating sulfate. We use the mean Na amounts in sea salt (31%; Schlesinger, 1997) to characterize the Na amounts and include the soluble Na measurements as well ( $\text{Na}^+$ ) if available when Na measurements are not available. Note that Cl cannot be used to evaluate sea salts as the Cl is degassed from aerosols, primarily due to sulfate interactions (e.g., Pio and Lopes, 1998). 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: 1) at some sites not all the elements are available, and 2) because these elements derive not only from desert dust, but also from industrial sources. 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. There are several sources of measurement differences between different networks as well as between model and observations. Modelled values of PM content, which assume dry particles, are used here, while gravimetric 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). In addition, comparisons of coarse mode composition at co-located sites in the US show that the inlet type can cause ~30% difference in measured mass (Hand et al., 2017). We include these differences in our error estimate in Section 3.2.

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 (Mahowald et al., 2018). Assumptions about the model composition and how they are compared to observations are summarized in Table S1. For example, OM is assumed to be 1.8 times OC if OC measurements are available but not OM measurements.

Harmonizing models with different types of measurements is critical, and yet a difficult task (Huang et al., 2021). 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 or aerodynamic 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<sub>2.5</sub> and PM<sub>10</sub> 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., Stokes diameter (involving size and shape) weighed by the square root of the particle density; Hinds, 1999). The sharpness of the cut-off of such inlets influences the PM<sub>2.5</sub> and PM<sub>10</sub> mass concentration (Hand et al., 2019; Wilson et al., 2002). The PM<sub>10</sub> size cut-off aerodynamic diameter is equivalent to PM<sub>6.3</sub> geometric diameter for spherical dust particles (Hinds, 1999; Rodríguez et al., 2012) and to PM<sub>6.9</sub> in the case of dust elliptical particles (Huang et al., 2021). Similarly, for dust, PM<sub>2.5</sub> (aerodynamic diameter) is equivalent to PM<sub>1.6</sub> (geometric diameter). These differences are important to keep in mind, but the information is not available for all networks, so we include the size cutoff as an uncertainty in the model/data comparisons as described in Section 3.2.

For ease of viewing the data in this paper in the densely sampled regions as well as to compare model output to more representative spatial scales, observational records from different sites were combined into a mean within a grid cell that is two times the model resolution, or approximately  $2^\circ \times 2^\circ$ . This process averages the observations over a spatial scale appropriate for comparison with the chemistry model (Schutgens et al., 2016). We provide both the climatological annual average data at each site as well as the  $2^\circ \times 2^\circ$  grid-averaged data (with the modelled data at doi: 10.5281/zenodo.10459654, Mahowald et al., 2024). In this dataset, the number of station datasets included in the average is included (stations) and the number of observations add up across all the station datasets included.

Notice that we include both urban regions and rural or remote sites into the same dataset. Some of the original metadata did not include the resolution of the location to better than 0.1 degrees, so that the coordinates of the locations here provided with the gridded data should not be used for finer resolution studies. Because of the importance and size of megacities, which cross multiple grid boxes, as well as the difficulty in separating urban vs. rural sites, we include urban and rural air quality data in the same dataset, and previous studies show the expected differences between urban and rural concentrations and trends (e.g., Hand et al., 2019).

Statistical comparison across the globe and different regions are included in the supplemental tables. These include model and observational averages, Kendall correlation coefficients (rank correlations), linear regression slopes and uncertainties, as well as root mean squared differences. We also include the fraction of the model/data comparison which is outside the error bounds defined in Section 3.2. These results are included in tables in the supplement and referred to in the text as appropriate.

350 There are multiple sources of uncertainties in the observations used in the model-data comparisons of PM concentrations at  
351 the global model grid scale: errors in the measurements, differences in measurement methods, variability in aerosol  
352 concentrations during events versus background conditions, spatial variability within a model grid box, and interannual  
353 variability. To assess the size of these uncertainties, we look at the normalized standard deviation (defined as the standard  
354 deviation over the mean) due to these factors in the observations for within year, within a  $2^\circ \times 2^\circ$  degree grid and for  
355 interannual variability. To evaluate within year and between year variability, we focus on stations that have more than 10  
356 years of data. To evaluate spatial variability within grid boxes, we use grid boxes that have more than 10 stations within  
357 them. Notice that these grid boxes are likely to lie close to cities and fossil fuel source regions, because the measurement  
358 network is more dense there, perhaps exaggerating the importance of spatial variability. In addition, different measurement  
359 methods (dry vs. moist aerosol mass, different inlet geometries) complicate the comparison of data. We assume here a  
360 measurement method uncertainty of 30% that is on the high side of previous studies (Prank et al., 2016; Burgos et al., 2020;  
361 Hand et al., 2017). Many of the measurements also include an assessment of their uncertainty or of the minimum detected  
362 limit: we use that to assess the average uncertainty of individual measurements (measurement errors).

363

## 364 **2.4 Temporal aerosol variability**

365 While the main goal of this study is to highlight and compile in one place the many surface concentration observational  
366 datasets available to compare against models, and we focus on the climatological annual mean, the datasets also include  
367 temporal variability. Annual means, standard deviations and the number of observations for each station for each year are  
368 included to allow for analysis of interannual variability or trends. In addition, the climatological monthly mean, standard  
369 deviation and number of observations is also available in order to assess the seasonal cycle. These values are all available in  
370 the Allobservvations.AEROMAP.nc file available at doi: 10.5281/zenodo.10459654.

371 To illustrate the included data, the trends in the PM<sub>2.5</sub> and PM<sub>10</sub> aerosols are calculated over 2000-2023, over 8 different  
372 regions: North America, South America, Africa, Europe and Asia. Only data after 2000 is included because there is much  
373 more data after 2000 than prior (see Section 3.1). All station datasets with more than 8 years of data are included in the  
374 calculation. In order to decrease the bias and uncertainty due to the large temporal and spatial variability (similar to Hand et  
375 al., 2024), we divide the annual mean at each station by the climatological annual mean over the two time periods, and  
376 average this with the other stations within the region. We then use a Theil regression which calculates the slopes excluding  
377 different datapoints and takes the median slope to reduce dependence on outliers (Hand et al., 2024). Median, 33 and 66  
378 percentile slopes are calculated to show the median and 1-sigma uncertainties for each region.

379 The seasonal cycle of aerosols can provide important information about the source strength and variability, as well as  
380 meteorological constraints (Gui et al., 2021; Rasch et al., 2000). To illustrate the value of the evaluation of the seasonal  
381 cycle in models, we calculate the climatological monthly mean in the observations and model and compare the correlation of  
382 these values, as well as the standard deviation of the 12 month means in the model versus the observations. This method  
383 allows us to separately evaluate the seasonal cycle from the spatial distribution. The correlation is only calculated at stations  
384 where the seasonal cycle is large enough: in math terms our criteria is where the observed standard deviation across months  
385 is larger than half of the average observed within month variability.

## 386 **3 Results**

### 387 **3.1 AEROMAP observational data set**

388 First, we assessed the amount of data and the number of station datasets within each  $\sim 2^\circ \times 2^\circ$  gridded area (Fig. 1). The  
389 observational dataset provides coverage predominately over North America and Europe for  $\text{PM}_{2.5}$  and  $\text{PM}_{10}$ , as noted by  
390 previous studies (e.g., Szopa et al., 2021), but in addition we provide here a synthesis of more air quality data in other  
391 regions, especially Asia (Fig. 1). This compilation data set comprises most of the individual observations (at daily or longer  
392 time periods) of total  $\text{PM}_{2.5}$  (Fig. 1a, 1e: blue bars) and most of the observing stations (Fig. 1e and blue line). Approximately  
393 15,000 stations and over 20 million observations are included in this compilation.

394 Notice that there are two to three orders of magnitude more individual observations for the total mass (PM) of particles  
395 compared to information about the composition of particles (Fig. 1e), which is shown also by contrasting the spatial  
396 distribution of measurements between  $\text{PM}_{2.5}$  and measured amounts of OM (Fig. 1a versus 1b), as well as a large difference  
397 between the number of station datasets measuring the total mass versus the speciated aerosol particles like OM (Fig. 1c  
398 versus 1d). While this dataset presents a huge increase in the amount of data available to the aerosol modelling community  
399 (for example, an eight-fold increase compared to the datasets included in Reddington et al., 2017), still the dominant  
400 proportion of the total  $\text{PM}_{2.5}$  or  $\text{PM}_{10}$  data are clustered over a few industrialized land regions, and there is little composition  
401 information over most of the globe (Fig. 1).

### 402 **3.2 Uncertainties in model-data comparisons**

403 Our goal in this study was to identify observational datasets and compile them together into one easy-to-use dataset for climate  
404 and air quality modelers. To do that we collect all available datasets, prioritizing long-term stations with composition data, but  
405 in regions with few measurements, we include only PM data, or data collected during field campaigns, which may last only a  
406 month or two. Previous studies have shown that even a 1-day average aerosol measurements, carried out on cruises, can  
407 constrain aerosol concentrations within a order of magnitude (1-sigma) for phosphorus in dust, which varies spatially by 4  
408 orders of magnitude (Mahowald et al., 2008). Other studies have highlighted that even for particles that have highly variable

sources, such as dust, that only a few months of observations are enough to characterize the mean and standard deviation in most places across the globe (Smith et al., 2017). However, that study highlighted that for places where dust events do not occur every year or occur with varying number, like near South America, several years are required to characterize the mean (Smith et al., 2017).

Uncertainties in the observation-model comparisons can include both uncertainties in the observations, as well as interannual variability in both the model and observations that are temporally averaged. Uncertainties used in the comparisons of aerosols at the global model grid scale come from multiple sources: errors in the measurements, differences in measurement methods, variability in aerosol concentrations during events versus background conditions, spatial variability within a model grid box, and interannual variability, as discussed in Section 2.3. To assess the size of the variability contribution to the uncertainties, we look at the normalized standard deviation (defined as the standard deviation over the mean) due to these factors in the observations for within year, within grid and interannual variability. Nonetheless, our estimate of spatial variability will underestimate the true value in the absence of sufficient spatial coverage. In addition, different measurement methods (dry vs. moist aerosol mass, different inlet geometries) complicate the comparison of data (Section 2.3 discusses sources of uncertainties in more detail). We assume here a measurement method uncertainty of 30% that is on the high side used in previous studies (Prank et al., 2016; Burgos et al., 2020; Hand et al., 2017). Many of the measurements also include an assessment of their uncertainty: we use that to assess the average uncertainty of individual measurements due to measurement errors.

We focus on the uncertainties in the PM<sub>2.5</sub> measurements first. The largest uncertainties are associated with within-year variability (0.53) (Figure 1f; Table S3). This is because most of the aerosol mass can sometimes come in a few pollution events. Uncertainty due to combining different measurement methods (0.3) and from spatial variability within a model grid cell (0.24) are also important (Figure 1g). Both interannual variability (0.18) and measurement errors (0.08) are smaller but important contributions to uncertainty. The importance of within year variability (which is similar to within month variability: see Table S4) is consistent with studies showing that in most places, there are a few pollution events carrying much of the mass, and with otherwise much lower background concentrations (Luo et al., 2003; Fiore et al., 2022). Obviously, interannual variability is important for secular trends (Gupta et al., 2022; Watson-Parris et al., 2020; Mahowald et al, 2010), but in this compilation the interannual variability is much smaller than the 2-4 orders of magnitude of the spatial variability across the globe, and thus can be neglected for understanding global spatial distributions (Figure 1f).

These sources of uncertainties occur simultaneously and if we sum them assuming orthogonality, we obtain an normalized uncertainty of ~0.68 (Table S3), which was interpreted as meaning that model/data comparisons within a factor or three should be considered adequate. To ease the visual evaluation of the comparison we show in the following scatter plots both the 1:1 line and the range within a factor of 3. We discuss an example of uncertainties in more detail in Section 3.3. Notice that if we

440 use the same metric (normalized standard deviation) to evaluate the variability across the climatological concentrations  
441 measured in the observations at different locations (Figure 3a) or across the grid averages in the model we obtain 1.0 and 2.2,  
442 respectively, much larger than the uncertainties (0.6): there is much more variability across different grid boxes (4-5 orders of  
443 magnitude: see Figure 2d) than across different years (up to 50% normalized standard deviation; Figure 2f). As expected, the  
444 model contains more spatial variability than the observations, as the model reports concentrations in very high (North Africa)  
445 and very low (Antarctica) aerosol regions where we have no data, although where we have data, the model simulates a similar  
446 range (Figure 3a). For composition measurements, there is larger uncertainty in some individual species (e.g., BC and Al)  
447 than for PM. However there are many fewer composition observations (Table S3). Since the statistics of the uncertainty  
448 calculations are likely more robust with the bulk PM measurements, as there are an order of magnitude more data for the bulk  
449 PM data, we use the uncertainty estimate derived for PM for all of the composition data in this paper.

450 There is time variation in how much data is available for both  $PM_{2.5}$  and  $PM_{10}$  data (Figure 2a and 2b), with the most data  
451 available between 2010 and 2020. Different regions have slightly different trends in the amount of data (Figure 2). For much  
452 of this paper we will discuss global and regional comparisons, and the regions we focus on are Africa, Asia, Australia, Europe,  
453 North America, South America and the high latitudes (Figure 2c).

454 Trends in aerosols are an important scientific question, although for most of this paper we use the climatological annual  
455 mean. What if there were strong trends in the aerosols; would that lead to differences between our climatological means and  
456 what we expect for some decades? In order to assess this, we look at the individual annual means for each station with more  
457 than 8 years of data and see if the individual annual mean is ever outside of the 3x uncertainty calculated here. Out of the  
458 13320 station datasets for  $PM_{2.5}$  or  $PM_{10}$  which have more than 8 years of data, only 175 (1.3%) have an annual average  
459 outside the uncertainty estimated here. Of those with a value outside the uncertainty, only 10 (<0.01%) have a statistically  
460 significant trend. This suggests that for the temporal interval we have chosen for the climatology, long term trends are not a  
461 significant source of differences in the spatial climatological dataset presented here. Nonetheless, we acknowledge that in  
462 regions where aerosol emissions increase and then decrease over our multi-decadal observational record (e.g. China), our test  
463 for trends will not reveal where the climatology over the full period is less representative of individual decades. We also  
464 supply in the compiled dataset a decadal mean for the time period of 2010-2019, which is made publicly available. A  
465 comparison of the climatological mean versus the decadal mean for the  $PM_{2.5}$  and  $PM_{10}$  concentrations show that for almost  
466 all locations, there is a small difference between the two values, and they lie on a one-to-one line (Figure 2d and 2e; Table  
467 S4). There are a few station datasets (<5%) which have a difference between the climatological mean and the decadal  
468 mean that is larger than 20%, and very few (<0.05%) have a difference which is larger than the uncertainties described in  
469 this section (factor of 3; Table S4). The biggest difference between the climatological and decadal average values is the  
470 number of station datasets and observations and thus spatial coverage: we lose between 20% and 100% of the station  
471 datasets, depending on the size and composition, when we use the decadal means (Table S5). This is because even though

472 this is the most observed decade, still some datasets are outside this time period. In order to emphasize the spatial  
473 distribution of the datasets, and because the climatological values are so similar to the decadal means, we will show just the  
474 climatological values in the next few sections, although both are available (Supplemental dataset 1 and  
475 <https://zenodo.org/records/10459654>).

### 476 **3.3 PM<sub>2.5</sub> model-data comparison**

477 Modelled concentrations of PM<sub>2.5</sub> are more often compared against observations than for PM<sub>10</sub> or other size fractions, and  
478 comprise an important portion of the particulate matter associated with human activities. Therefore, we describe first the  
479 observational synthesis and comparison to model results for PM<sub>2.5</sub>. Because the high number of observations in some parts of  
480 the world would make the figures unreadable, the observations are gridded onto an approximately 2°×2° grid for  
481 comparisons with the model, although individual data points are still difficult to read (Fig. 3a). The maps illustrate where the  
482 observational comparison in the scatter plot is made, and focused maps of each major region are available in the supplement  
483 (Figure S1) as well as global and regional statistics (Table S5). As expected, in the model the highest concentrations are over  
484 the desert dust regions, such as North Africa, and over heavily industrialized regions in Asia. For the heavily industrialized  
485 regions in Asia, these high values are consistent with the observations, but the regions in North Africa with the highest  
486 modelled values do not have similar observational validation for high concentration values due to a lack of data (Fig. 3a).

487 Overall, the model is able to simulate much of the spatial variability in PM<sub>2.5</sub> over two orders of magnitude (Fig. 3a and 3c),  
488 however there is a tendency to overestimate in the PM<sub>2.5</sub> over India and China (Fig. 3b), although the mean over all the  
489 regions is within the 3x uncertainty (Fig. 3c: bold symbols). In addition, there are some observations (globally ~6% Table  
490 S6) that are outside the 3x uncertainty estimates (Figure 3c and 3d). The scatterplots show the comparisons of the model to  
491 the observations using the gridded data (Fig. 3c) and all original data (Fig. 3d), and the correlation coefficients are similar  
492 (0.60 vs. 0.67 in Fig 3c and Fig 3d, respectively). It is interesting that the correlation using the ungridded data (Fig 3d) is  
493 slightly higher, perhaps because the model does better in regions with more data, although this is not a statistically  
494 significant result. The averages over different regions show that on average, the model is simulating the regions within the  
495 uncertainty (bold black symbols in Fig 3d; Table S5).

496 As an example of the source of the uncertainties discussed in Section 3.2, we discuss the differences over India and China in  
497 the Asia region in more detail. It seems likely that at least some of these errors are due to an overestimate in the emission  
498 databases, since satellite based remote sensing has suggested that models overestimate in SO<sub>2</sub> over China (Luo et al., 2020).  
499 In addition, these discrepancies could be due to an error in the aerosol transport modelling or the time period: the  
500 observations are more recent while the assumptions for the emissions are for the year 2010 (Quass et al., 2021). The  
501 comparison using the decadal averages (2010-2019) show similar biases (Figure S2) as expected since the decadal averages  
502 are so similar to the climatological averages (Figure 2d), which suggests the time differences may not be the most important



503 factor. In addition, notice that once averaged over the  $2^{\circ} \times 2^{\circ}$  grids more observations are within a factor of 3, our uncertainty  
504 bound (contrast 3c and 3d). However, there could also be methodological and analytical differences due to which group or  
505 network did the observations or the exact locations of the different monitors. Much of the data in those regions are not  
506 usually included in routinely used previous compilations of data (e.g., Reddington et al., 2017), so the fact that previous  
507 model studies have not been able to assess emission datasets in these regions could also partially explain this discrepancy.  
508 Comparison between different observations in some cities (Fig. 4) shows that in these grid boxes there can be very large  
509 differences ( $\sim$ factor of 3) between the annually averaged values reported at nearby stations within  $1^{\circ}$  distance radially.  
510 Notice that the AirNow measurements (<https://www.airnow.gov/international/us-embassies-and-consulates/> on the US  
511 embassies) tend to be higher than those reported from government air quality networks. The sites compared are in large cities  
512 and thus are likely to have strong local sources and intense gradients in pollutants. For now, we keep in mind this large  
513 difference, but continue to use the observations. As indicated below, in these regions we do not have measurements of  
514 composition so we do not know which constituents are poorly simulated in our emissions or transport modelling. More  
515 statistics describing the model data comparisons are shown in Table S5.

516 Next, we consider the composition of the  $PM_{2.5}$  aerosol in the model versus the observations. The model simulates high and  
517 low values of sulfate observed with a correlation coefficient of 0.64. Sulfate particles concentration are on the high side in  
518 the model in several regions: more so in North America, Africa, but less so for Europe and other regions (Fig. 5a and b;  
519 Figure S3; Table S5), although all of the regional means are within the 3x uncertainty (bold symbols in Fig. 5b). Previous  
520 studies have compared  $SO_4^{2-}$  aerosol observations to some model simulations and have not noted this bias (e.g., Barrie et al.,  
521 2001; Aas et al., 2019) but this bias was seen in this model and attributed to the simple chemistry included in the model (Liu  
522 et al., 2012; Yang et al., 2018). About 18% of the climatological mean model values are outside the 3x uncertainty, and a  
523 larger fraction is outside for Africa, Australia and South America, where there is less data (Table S5).

524 BC comparisons suggest the model results are roughly able ( $r=0.63$ , within the 3x uncertainty) to capture the spatial  
525 dynamics of this aerosol across more than 2 orders of magnitude, although in some regions model values are on the low side  
526 (Europe and Asia) (Fig. 5c and d; Fig. S4; Table S5). This is similar to previous model intercomparisons (Koch et al., 2009;  
527 Bond et al., 2004, 2013; Liu et al., 2012, 2016). About 18% of the model values are outside the uncertainty bounds, and  
528 many of these values come from Europe, where 36% of the values in Europe are outside the uncertainty bounds (Table S5).  
529 Simulations of OM in the default model (Fig. 5e) suggest that the model is within the uncertainty of most of the data, and the  
530 regional averages are close to the 1:1 line (Fig. 5f). Correctly modelling organic material is very difficult both due to the  
531 sparsity of data for comparison, as well as the importance of both primary and secondary OM in PM (Heald et al., 2010;  
532 Kanakidou et al., 2005; Olson et al., 1997; Tsigaridis et al., 2014), and previous studies with this model have noted an  
533 overestimate in comparison with surface observations (Liu et al., 2012).

534 As a proxy for sea salts, we use the elemental data of the major component, Na, and we see the highest values over oceans  
535 and lower values over land, as expected and seen in the observations (Fig. 5g). Although most of the data is within the  
536 uncertainties (30% is outside the uncertainties; Table S5), the model tends to be too high at low Na and too low at high Na in  
537 North America, where much of the data are available (Fig. 5g and h; also seen in slopes Table S5), which has been seen  
538 previously with this model (Liu et al., 2012). Notice that we do not include industrial emissions of Na, but the  
539 concentrations far inland include some Na, suggesting land-based natural or industrial sources. As a proxy for dust, we use  
540 Al amounts (Fig. 5i and j), which globally and over dust regions are dominated by dust, although there are few observational  
541 datasets in high dust regions. The comparisons suggest the model is able to simulate dust (correlation coefficient=0.5, Table  
542 S5) across 4 orders of magnitude, similar to previous studies (Liu et al., 2012; Albani et al., 2014a; Li et al., 2022; Huneus  
543 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  
544 regions, similar to sea salts (Fig. 5i and 5j; also seen in the slopes in Table S5). One reason for this overestimate of PM<sub>2.5</sub>  
545 aerosol mass for constituents like sea salt and dust that are predominantly in the coarse mode, is that the coarse mode in this  
546 model has a wide enough standard deviation that it contributes significantly to the PM<sub>2.5</sub> size fraction (Ke et al., 2022; Li et  
547 al., in prep.). Better resolution of the coarse mode aerosol may be required to better simulate these aerosols (Ke et al., 2022;  
548 Li et al., in prep.).

549 Next, we consider the ammonium nitrate that requires complicated gas-aerosol phase equilibrium to correctly simulate (e.g.,  
550 Bauer et al., 2007; Thornhill et al., 2021; Adams et al., 2001; Regayre et al., 2018; Seinfeld and Pandis, 2006; Wolff, 1984).  
551 To summarize these complicated interactions, because SO<sub>4</sub><sup>2-</sup> is a stronger acid than NO<sub>3</sub><sup>-</sup> in the atmosphere, the basic NH<sub>4</sub><sup>+</sup> is  
552 preferentially found with SO<sub>4</sub><sup>2-</sup>. Thus NO<sub>3</sub><sup>-</sup> particles will only form if there is sufficient NH<sub>4</sub><sup>+</sup> available. As described in the  
553 methods, to include these particles we added to the aerosol mass simulations from a different version of the same model  
554 which include chemistry (Vira et al., 2022), and a more process-based source of ammonia (Vira et al., 2020) since the default  
555 CESM2 version used here does not include chemistry. Note that even in the chemistry version of the model for CESM2 the  
556 complicated gas-aerosol phase thermodynamic equilibrium calculations are not included, which causes errors in the  
557 simulation of the amounts of nitrogen aerosol (e.g., Bauer et al., 2007; Thornhill et al., 2021; Adams et al., 2001; Regayre et  
558 al., 2018; Nenes et al., 2021). Thus while the NH<sub>3</sub> agricultural emission scheme used in this model is state-of-the-art, the  
559 lack of an adequate gas-aerosol phase separation may lead to biases as discussed in Vira et al. (2022). In addition, recent  
560 studies have suggested that emissions of NH<sub>4</sub> from vehicles should be 1.8x higher than previously estimated (Toro et al.,  
561 2024), highlighting the difficulty of adequate emission datasets for nitrogenous aerosol precursors. NO<sub>3</sub><sup>-</sup> particles compared  
562 against available observations show that over 2 orders of magnitude, the model results are able to simulate the spatial  
563 variability (correlation coefficient=0.55), but the model tends to overestimate the observations by about a factor of 2 (except  
564 in South America), similar to what was seen in Vira et al., (2022) (Fig. 5k, 5l, Table S5). The model surface concentration  
565 NO<sub>3</sub> values are with most of the data within the uncertainties (Fig. 5k and l; 46% are outside the uncertainty bounds in Table  
566 S6). The model and data distribution of NH<sub>4</sub><sup>+</sup> show the high values of NH<sub>4</sub><sup>+</sup> over agricultural regions especially (e.g., Vira et

al., 2022), like the mid-western US or central Europe (Fig. 5m and Fig. 5n; correlation coefficient=0.52). The  $\text{NH}_4^+$  in the simulation used here compares well to available observations across the different regions by having the regional averages being close to the 1:1 line (Fig. 5n), most of the individual model-data comparisons being within the uncertainties at most observational sites (Fig. 5m and n; and 16% of the data is outside the uncertainty bounds in Table S5).

How would these comparisons change if we used the decadal 2010-2019 averages instead of the climatological averages of the observations? As expected from the similarity between the observations averaged over these two time periods (Section 3.2; Table S4) the results do not substantially change (>20%) in most regions where there is a similar amount of data (Fig. S2a; Table S6). But for some regions and composition datasets, there is much less data (>25% less data), and in those cases, there can be large differences between using the decadal averages versus the climatological averages (Table S6). This suggests that using the climatological averages for our comparisons for  $\text{PM}_{2.5}$  allows us to include more data and evaluate more regions, without including much bias, since interannual variability is a small source of uncertainty compared to other uncertainties (Table S4).

### 3.4 $\text{PM}_{10}$ model-data comparison

$\text{PM}_{10}$  was the first size selective standard for particulate air quality until more studies showed that smaller particles ( $\text{PM}_{2.5}$  or  $\text{PM}_1$ ) were more relevant for health impacts and  $\text{PM}_{2.5}$  standards were added (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  $\text{PM}_{10}$  measurements routinely made (Fig 1d; Fig. 7a). The model is able to simulate  $\text{PM}_{10}$  concentrations across 2 orders of magnitude with some skill (correlation=0.55; Fig. 7a and 6b), as most of the data is within the uncertainties (Fig. 5a, b and c; 8% of data is outside the uncertainty Table S7). Gridding the data before comparing to the model results in a slightly higher correlation across space as including all data (0.55 vs. 0.72; Fig 5b vs. c). More statistical comparisons are shown in Table S7. The regional averages are all within the uncertainty bounds for all regions.

There are fewer comparisons with  $\text{PM}_{10}$  composition data available in the literature: usually only sea salts and dust are compared to observations that include the coarse mode (Gong et al., 2003; Ginoux et al., 2001; Albani et al., 2014b; Mahowald et al., 2006). Comparisons for  $\text{SO}_4^{2-}$  suggest that the model can estimate the distribution of the high and low concentrations (correlation coefficient=0.43), but tends to over predict  $\text{PM}_{10}$  values across most regions (Africa, Australia, Europe, North America and South America), as many observations are too high and outside the uncertainty bounds (Fig. 7a and b.; Table S7 indicates 48% of the model values are outside the uncertainty bounds). For BC, the  $\text{PM}_{10}$  simulation captures the range of values (correlation coefficient of 0.47), with most of the model results within the uncertainty bounds of the observations across all the regions (Fig. 7c and d; 16% outside the uncertainty bounds in Table S7). There is suggestion in the observations that there may be some fraction of BC in the coarse mode, since there is more BC in  $\text{PM}_{10}$  than in  $\text{PM}_{2.5}$ , but in the simulations used here there is no mass in the coarse mode (compare Fig. 7c versus 5c). The model-data

599 comparisons simulations for OM suggest a good spatial distribution of OM (correlation coefficient=0.43) and the modelled  
600 regional averages are similar to the observations. Again, the model does not simulate coarse mode OM currently, and does  
601 not include primary biogenics (Jaenicke, 1979; Mahowald et al., 2008), and yet can match the observations. The limited Na  
602 (indicating sea salt) data suggest the model can simulate the spatial distribution (correlation coefficient=0.49), but tends to  
603 overestimate and has many observations outside the error bound (Fig. 7g and h; 50% of the observations are outside the  
604 uncertainty bounds in Table S7), as was seen previously (Liu et al., 2012). Most of the regional averages, however, are just  
605 on the line of the uncertainty bounds (Fig. 7h). Comparisons with Al (used here as a proxy for dust) show that the spatial  
606 variability is correlated between model and observations (correlation coefficient of 0.46), but the model overpredicts the  
607 concentrations in high dust regions and underestimates in low dust region (Fig 7i and 7j; 54% of the observations are outside  
608 the uncertainty bounds in Table S7). The largest overestimates are in Asia and Africa (Fig 7i and 7j). Dust models are  
609 compared against aerosol optical depth, deposition and surface concentrations and it is currently not possible to simulate all  
610 of these different types of measurements at the same time, consistent with previous studies with this model (Li et al., 2022;  
611 Kok et al., 2014b; Albani et al., 2014a; Matsui and Mahowald, 2017; Zhao et al., 2022), and indeed across most dust models  
612 (Huneeus et al., 2011).

613  
614 The model simulations of  $\text{NO}_3^-$  suggest too high values in high  $\text{NO}_3^-$  areas, and too low in low  $\text{NO}_3^-$  regions, especially in the  
615 limited data for the South American region (Fig. 7k and l; Table S7 shows 69% of the data is outside the uncertainty  
616 bounds).  $\text{NH}_4^+$  shows a slightly better comparison to the limited available data (Fig. 7m and n) as seen in Vira et al. (2022).  
617 As discussed earlier, the model does not include other forms of nitrate aerosols which may be important, such as the reaction  
618 of nitrate with dust aerosols (Wolff, 1984; Dentener et al., 1996; Xu and Penner, 2012).

619  
620 If we compared instead to the decadal averages rather than the climatological averages, we would obtain similar results in  
621 many cases (Fig. 2b; Table S8), but being limited to decadal averages reduces substantially the amount of observations  
622 available for comparison. The few regions which lose less than 25% of the data sets when we temporally limit our  
623 comparison have similar statistics similar as in the  $\text{PM}_{2.5}$  comparisons. Again, this suggests that using the climatological  
624 averages includes more regions in the comparisons without evidence that it increases bias, because of the small amount of  
625 interannual variability in this data set (Section 3.2).

### 626 627 **3.5 Temporal variability**

628 This paper emphasizes the expanded spatial coverage in this compiled dataset with the spatial comparisons in Section 3.2-  
629 3.5, but the dataset also contains temporal variability as well. To illustrate the type of temporal data within this dataset we  
630 present briefly some common metrics. First, we consider what trends this data suggests in the surface concentrations for  
631  $\text{PM}_{2.5}$  and  $\text{PM}_{10}$ . Because most of this data comes from after 2000 (Figure 2a and 2b), we focus on the trends between 2000-  
632 2023. We also average by region in order to obtain a large-scale trend in surface concentrations (see details of methods in

Section 2.5). Overall, the observations suggest that there is a statistically significant (1-sigma) decrease in aerosols over this time period of about 1% per year for PM<sub>2.5</sub> for North America, South America, Africa, and Europe, but not a statistically significant change over Asia and Australia (Figure 8a). These downward trends are similar to those seen in other studies including North America and Europe (Hand et al., 2024; Gui et al., 2021; Gupta et al., 2020; Mortier et al., 2020) and South America (Mortier et al., 2020), and the more ambivalent signals over Asia and Australia have also been seen (Gui et al., 2021; Gupta et al., 2020; Mortier et al., 2020). For PM<sub>10</sub>, there are different trends: North America and Europe have a statistically significant downward trend of about 1%/year while Asia has a larger downward trend of about 3% per year, but the error bar overlaps the 0 line for the south American, Africa and Australian regions, indicating that those regions do not have statistically significant downward trends. There are no other studies we know of that looked at trends in PM<sub>10</sub> specifically. Note that we do not compare against the model results here, as our example model simulation does not include emission trends, but these datasets include each station's annual average so that more detailed comparisons could be conducted. In addition, apparently these trends do occur long enough to cause a large bias in the climatology (Section 3.2)

Next, we use the climatological monthly mean data for PM<sub>2.5</sub> and PM<sub>10</sub> and compare against the model to see how well the models simulate the seasonal cycle. There are many ways to evaluate the seasonality in the literature (Gleckler et al., 2008; Henriksson et al., 2011; Huneus et al., 2011; Rasch et al., 2000). We chose one way here, but this dataset could be used in other ways as well. The models can simulate the timing of the seasonal cycle well across most regions as seen in correlations between the climatological monthly mean in the model and observation (Figure 9a and 9b), but there are several regions where the model is not capturing the timing of the seasonal cycle (e.g., northern India, Turkey, New Zealand). The spatial distribution of the size of the seasonal cycle (defined here as the standard deviation in the climatological monthly mean) is less well simulated than the annual mean (contrast Fig. 8d with Fig. 3c and 8f with 6c: the correlation coefficients are smaller and there is more spread in the comparisons with the scatter plot). Examining whether this is a model-specific result or more generally occurs in the models would help discriminate between errors in the input emission datasets or meteorological errors in the model (e.g., Huneus et al., 2011).

### 3.6 Data and model coverage

The compilation shown here is the most comprehensive currently available for describing the spatial variability of the total mass and composition of in situ particulate surface concentration data, and yet it highlights the lack of sufficient data to constrain the current global distribution of particles and their composition (Fig. 10a and b). Only 3% of the grid boxes (2°×2°) have PM<sub>2.5</sub> data (about 10% of land grid boxes), and only 0.3% have sufficient data to constrain most of the composition (defined as having 90% of the variables considered here: total mass, SO<sub>4</sub><sup>2-</sup>, BC, OM, Na or Cl, Al or dust, NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup>). There are even less data available to characterize PM<sub>10</sub> (Fig. 10b), which is less important for air quality and aerosol-cloud interactions but more important for aerosol-biogeochemistry interactions and long wave interactions

665 (Mahowald et al., 2011; Li et al., 2022a; Lim et al., 2012; Kanakidou et al., 2018). Because of the high spatial and temporal  
666 variability of coarse aerosols and the lack of satellite or other remote sensing data to characterize coarse sizes, this lack of  
667 data is a severe handicap in constraining aerosol radiative forcing, its uncertainties and other impacts of particles in the  
668 climate system. Indeed, many of these regions have also been identified as regions lacking sufficient remote sensing data for  
669 climate and air quality purposes (Millet et al., 2024).

670 In this paper, we included nitrate aerosols, which are not included in the default CESM simulations conducted for climate,  
671 and represent about 10% of the globally averaged surface concentration mass (Table 2; Fig. S18 and S19). When we look  
672 spatially, the default particles are the dominant particles over most of the planet (Fig. 11), but in many regions for both PM<sub>2.5</sub>  
673 and PM<sub>10</sub>, the default aerosol scheme includes less than 50% of the aerosol particles (Fig. 10a and c), with substantial  
674 contributions from the nitrate particles that we add to the simulation (Fig. 10b and d). The CESM2 (and some other climate  
675 models) do not include nitrogen particles (NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup>), because of the substantial complexity and computation load of  
676 chemistry and gas-aerosol equilibrium (Bauer et al., 2007; Thornhill et al., 2021; Adams et al., 2001; Regayre et al., 2018)).  
677 Previous studies have highlighted the importance of nitrogen particles for climate, air quality and ecosystem impacts (e.g.,  
678 Adams et al., 2001; Bauer et al., 2007, 2016; Kanakidou et al., 2016; Baker et al., 2021). Changes in nitrogen aerosol  
679 precursor emissions are likely to follow different future trajectories than SO<sub>4</sub><sup>2-</sup>, BC or OC, whose anthropogenic sources are  
680 mostly fossil fuel derived and should decrease in the future as renewable energy resources expand (Gidden et al., 2019).  
681 Ammonia has substantial sources from agriculture, which will likely to stay constant or expand (Gidden et al., 2019;  
682 Klimont et al., 2017; Bauer et al., 2016). This suggests there could be a substantial bias, especially regionally, in both  
683 historical and future aerosol forcings due to the exclusion of these important sources (e.g., Bauer et al., 2007; Thornhill et al.,  
684 2021; Adams et al., 2001; Regayre et al., 2018).

#### 685 4. Conclusions

686 In this study, we collect aerosol surface concentration datasets and present a new aerosol compilation (AERO-MAP)  
687 designed to evaluate the spatial and temporal variability of particulate matter in Earth system and air quality models. The in  
688 situ surface measurements complement the column totals typically retrieved by satellites. This dataset includes both total  
689 mass and composition, where available, including 15,000 station datasets and over 20 million daily to weekly averaged  
690 measurements. Climatological and decadal averages (2010-2019) are presented, and we recommend that the climatological  
691 averages be used, because they include more datasets, and multi-decadal and decadal means are extremely similar when  
692 compared (Section 3.2). Spatial variability of aerosols (Figure 1f and Section 3.2) is important to simulate accurately in  
693 models, as a prerequisite to identifying their human impacts. In addition, we make available annual means across time, and  
694 the climatological monthly means so that temporal trends can be assessed. Here we expand beyond the usual limited  
695 coverage of North America and Europe to present a more global view for observations of both PM<sub>2.5</sub> and PM<sub>10</sub> (Fig. 1).

Unfortunately, there are still very limited data characterizing both the surface concentration, size and composition of aerosol particles (Fig. 10), and the locations where we lack data have also been identified as lacking sufficient remote sensing data as well (Millet et al., 2024). While satellite remote sensing can indicate the total atmospheric loading during cloud free conditions, it cannot yet provide information about the size or composition of 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., 2001; Dubovik et al., 2002; Schuster et al., 2016; Gonçalves Ageitos et al., 2023; Obiso et al., 2023), but single scattering albedo, for example, is only available under very high ( $>0.4$  AOD) aerosol loading conditions, and thus is not available most of the time and space (Dubovik et al., 2002). Knowing the size and the composition of aerosols is key to their impacts on air quality and climate (Mahowald et al., 2011). Knowing what particles are dominant in a region is required, as fossil fuel derived aerosols will likely be reduced, while agriculturally based aerosols may well increase (Gidden et al., 2019). We also present a method that is generalizable to other models that use this dataset to evaluate both mass and composition for intercomparison projects and improvements in air quality and Earth system models. The novel aspect of this paper is to present this compilation in an easy to use netcdf format and some example comparisons that can be used in the future to evaluate and improve model simulations for individual models or for AEROCOM intercomparisons. The underlying data could also be used for data assimilation efforts or for estimating from the observations what the contributions are from different aerosols (e.g., similar to Prank et al., 2016).

This study has highlighted the value of surface concentration data by showing that it can identify where models do well or poorly not just for total mass, but also for different compositions and size, complimenting other data sources, such as remote sensing. A recent, independent and complementary effort collects all atmospheric composition data (not just aerosols) from many networks into one easy to use framework called GHOST (Globally harmonised dataset of surface atmospheric composition measurements; Bowdalo et al., 2024). The approach used in GHOST includes presenting the data in netcdf format, at the original resolution, with meta data about measurement type, etc. included, and is an important step forward (Bowdalo et al., 2024). At this point GHOST only includes a subset of the data available in this study: we hope that the GHOST effort can be expanded to include more spatial variability and be maintained into the future.

This study also highlights the importance of including all aerosol components into the models, and shows that in the CESM2 approximately 10% is missing. In many places, there is 50% of the particulate mass missing, due to lack of the nitrate particles (Fig. 10; Paulot et al., 2016; Adams et al., 1999; Thornhill et al., 2020). Because these particles are largely driven by agricultural sources and not fossil fuels, their concentrations will be hardly affected by the transition to renewable energy and may increase if agricultural production expands with population. Therefore, these nitrate aerosol particles represent important air quality and climate impacts that should be represented more accurately in future studies.

726 **Data availability:** The data compiled here is available as a csv table with citations as a supplemental data 1. This same file  
727 is available as well as gridded datasets with the compiled observations and modelled data in netcdf format at  
728 <https://zenodo.org/records/10459654>, **10.5281/zenodo.11391232** Mahowald et al., 2024. Additional underlying datasets  
729 available by request to [mahowald@cornell.edu](mailto:mahowald@cornell.edu).

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

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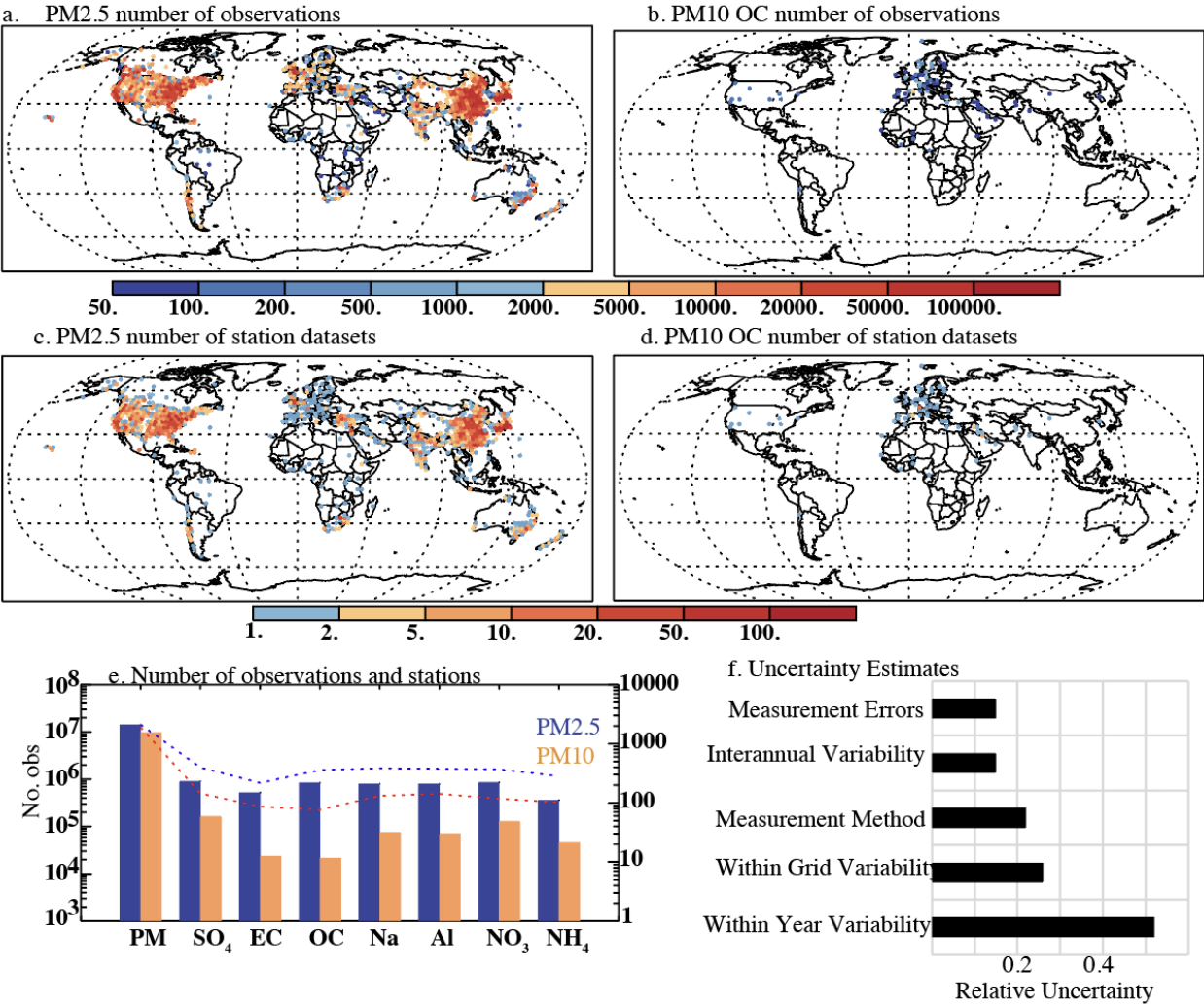
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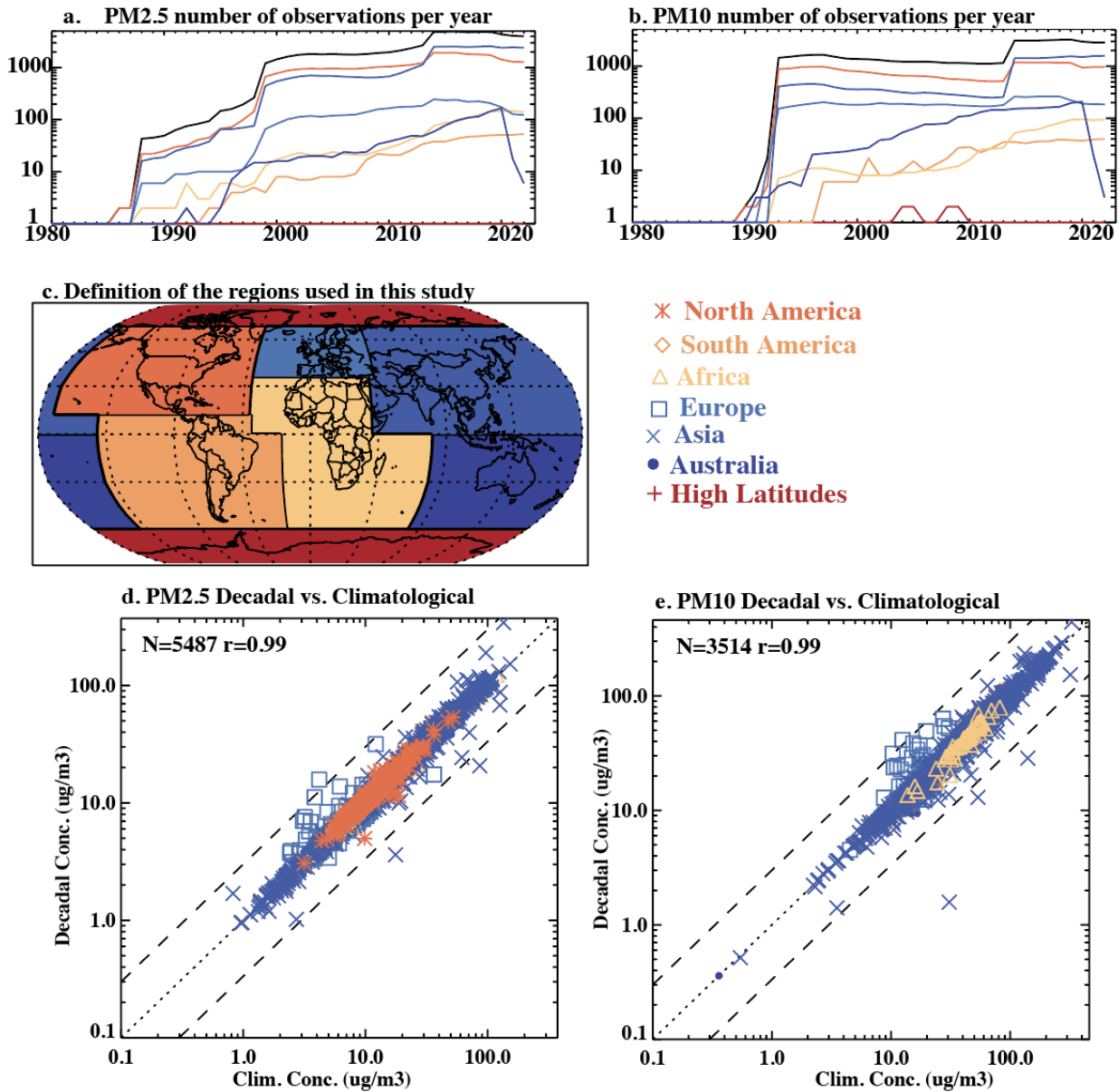
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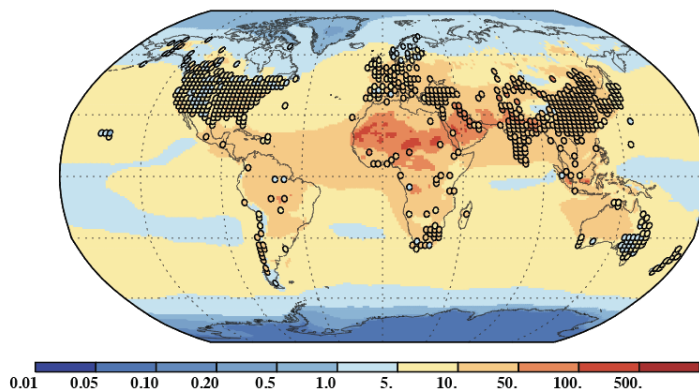
1582 **Figure 1:** Distribution of observations in the data base, showing the number of observations of PM<sub>2.5</sub> (a) and  
1583 PM<sub>10</sub> organic carbon (OC) (b) (with the colors indicating different numbers using the top color bar), as well as  
1584 the number of stations within each 2x2 grid locations for PM<sub>2.5</sub> (c) and PM<sub>10</sub> OC (d) (using the second color bar),  
1585 showing that there is much more PM<sub>2.5</sub> or PM<sub>10</sub> data, in contrast to speciated data. e) The number of observations  
1586 (bars) for total particulate matter (PM) or speciated data is summarized for the PM<sub>2.5</sub> (blue) and PM<sub>10</sub> (orange)  
1587 fraction using the left-hand side y-axis. The number of stations included in the study is shown as a dotted line (e)  
1588 and uses the right-hand size y-axis. f) Normalized (1 standard deviation over the mean) observational uncertainty  
1589 for PM<sub>2.5</sub> from measurement errors, interannual variability, measurement method, within grid variability and

1590 within year variability at the same station. Interannual variability and within grid uncertainty are defined as the  
1591 normalized standard deviation in the variability for stations that have more than 10 years of data. Within grid  
1592 variability is the normalized standard deviation of 2x2 grid cells that have more than 10 stations. Measurement  
1593 errors are the normalized standard deviation of the reported measurement errors for PM<sub>2.5</sub>. Measurement method  
1594 error derives from differences between different measurement methods (e.g., Prank et al., 2016; Burgos et al.,  
1595 2020; Hand et al., 2017). The stations included derive from the following sources (see supplemental dataset for  
1596 more details): Alastuey et al., 2016; Almeida et al., 2005; Amato et al., 2016; Andreae et al., 2002; Arimoto et al.,  
1597 2003; Artaxo et al., 2002; Barkley et al., 2019; Barraza et al., 2017; Bergametti et al., 1989; Bouet et al., 2019;  
1598 Bozlaker et al., 2013; Chen et al., 2006; Chuang et al., 2005; Cipoli et al., 2023; Cohen et al., 2004; da Silva et  
1599 al., 2008; Dongarrà et al., 2007, 2010; Engelbrecht et al., 2009; Formenti et al., 2003; Fuzzi et al., 2007; Hand et  
1600 al., 2017; Heimbürger et al., 2012; Herut and Krom, 1996; Herut et al., 2001; Hsu et al., 2016; Hueglin et al.,  
1601 2005; Furu et al., 2022, 2015; Gianini et al., 2012a, b; Kalivitis et al., 2007; Kaly et al., 2015; Kubilay et al.,  
1602 2000; Kyllönen et al., 2020; Laing et al., 2014b, a; Lucarelli et al., 2014, 2019; Mackey et al., 2013; Maenhaut et  
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1605 Mkoma et al., 2009; Morera-Gómez et al., 2018, 2019; Nava et al., 2015, 2020; Nyanganyura et al., 2007;  
1606 Oliveira, 2009; Oliveira et al., 2010; Pérez et al., 2008; Pio et al., 2022; Prospero et al., 1989, 2012, 2020;  
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1608 1993; Silva et al., 2010; Smichowski et al., 2004; Swap et al., 1992; Tørseth et al., 2012; Uematsu et al., 1983;  
1609 Vanderzalm et al., 2003; Virkkula et al., 1999; Xiao et al., 2014; Zihan and Losno, 2016. Data from several  
1610 online networks are also included ( e.g., <https://www.airnow.gov/international/us-embassies-and-consulates/>,  
1611 <https://quotsoft.net/air/>, <https://app.cpcbcr.com/ccr/#/caaqm-dashboard-all/caaqm-landing/data>,  
1612 <https://sinca.mma.gob.cl/index.php/>. <https://tenbou.nies.go.jp/download/>). See the supplemental data set for more  
1613 details and the doi links for the datasets.

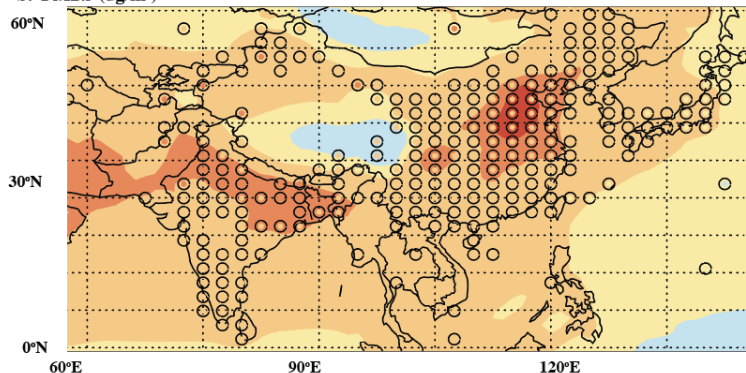


**Figure 2:** The temporal change in the number of observations of PM<sub>2.5</sub> (a) and PM<sub>10</sub> (b) available in this study (black) and by region: Dark blue: Australia, Blue: Asia, Light Blue: Europe, Yellow: Africa, Orange: South America, Red/orange: North America and Red: High latitudes; the regions are shown in (c), and are used throughout this study. Scatterplots comparing the climatological mean versus the decadal (2010-2019) mean surface concentration for PM<sub>2.5</sub> (d) and PM<sub>10</sub> (e), using symbols which indicate the region of the dataset point plotted.

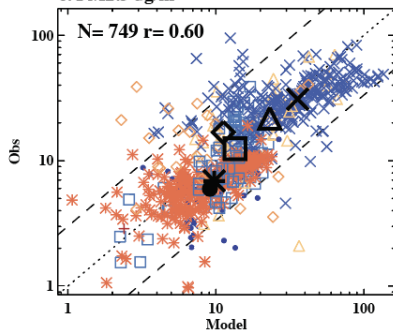
a. PM<sub>2.5</sub> (ug/m<sup>3</sup>)



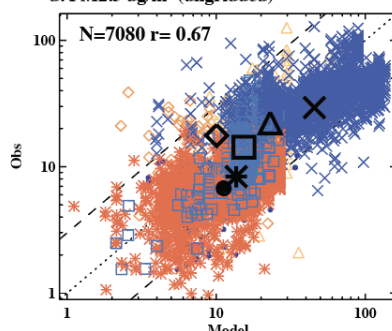
b. PM<sub>2.5</sub> (ug/m<sup>3</sup>)



c. PM<sub>2.5</sub> ug/m<sup>3</sup>



d. PM<sub>2.5</sub> ug/m<sup>3</sup> (ungridded)

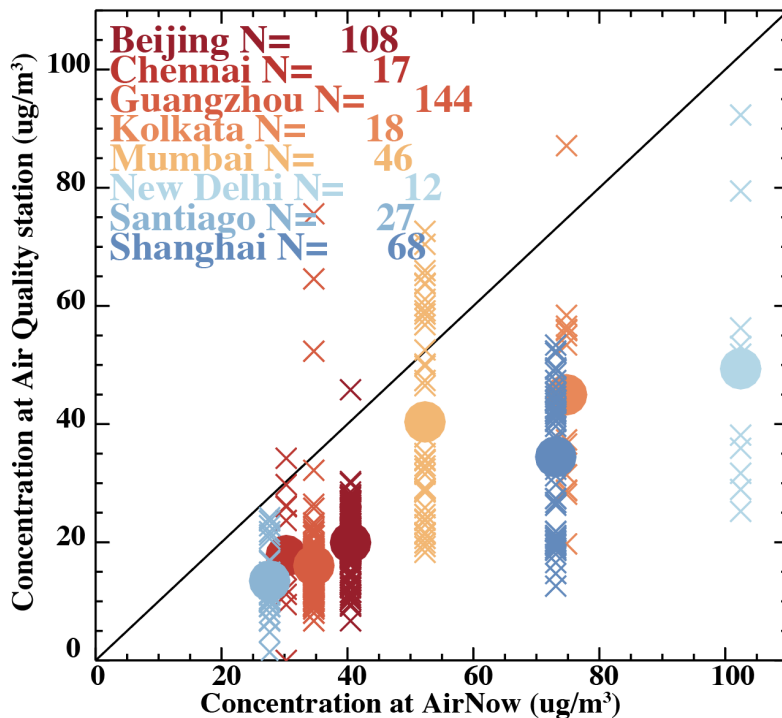


△ Africa    ● Australia    ✱ North America    + High Latitudes  
× Asia    □ Europe    ◇ South America

**Figure 3:** Model results and gridded observations for PM<sub>2.5</sub> in  $\mu\text{g}/\text{m}^3$  spatially mapped globally (a) and focused on just Asia (b) where the model is plotted as the background and the observations are circles with the colors indicating the amount of PM<sub>2.5</sub> 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 color and symbols indicate the regions, the bold black symbols are the average across each region (indicated by the symbol), the

1627     dotted line is the 1:1 line and the dashed lines are the factor of 3 uncertainty estimates. More statistics are shown  
1628     in Table S7, and maps focused on different regions are available in Figure S1.

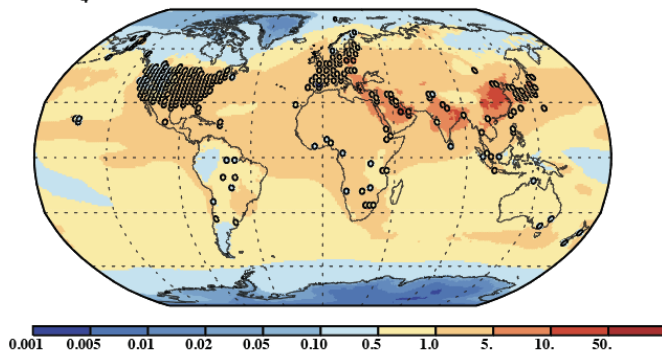
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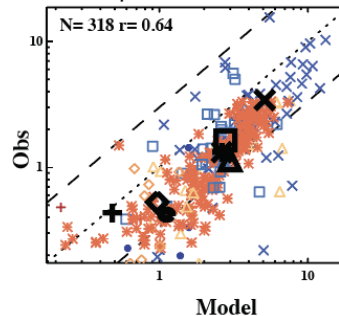
**Figure 4:** Comparison of PM<sub>2.5</sub> observations from the US Embassy’s AirNow network (<https://www.airnow.gov/international/us-embassies-and-consulates/>) versus observations from the Chinese air quality network (downloaded from <https://quotsoft.net/air/>) (Beijing 39.9N 116.4E, Guangzhou 23N 113E, Shanghai 31N 121E) and the Indian (Chennai 13N 80E, Kolkata 23N 88E, New Delhi 27N 77E) network (<https://app.cpcbcr.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.



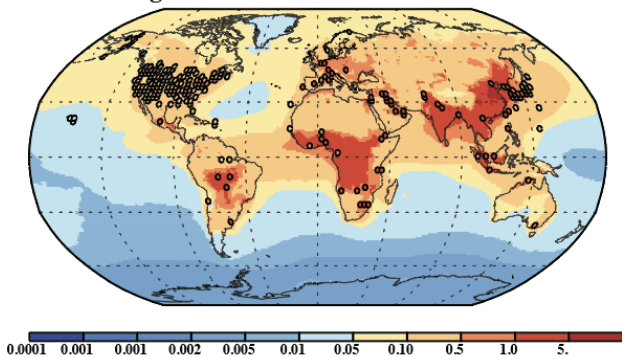
a.  $\text{SO}_4$  PM<sub>2.5</sub>  $\mu\text{g}/\text{m}^3$



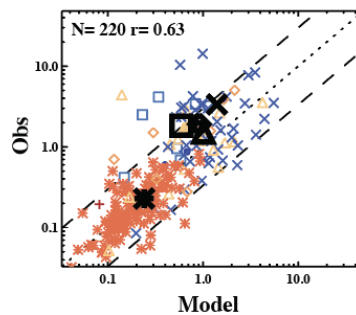
b.  $\text{SO}_4$  PM<sub>2.5</sub>  $\mu\text{g}/\text{m}^3$



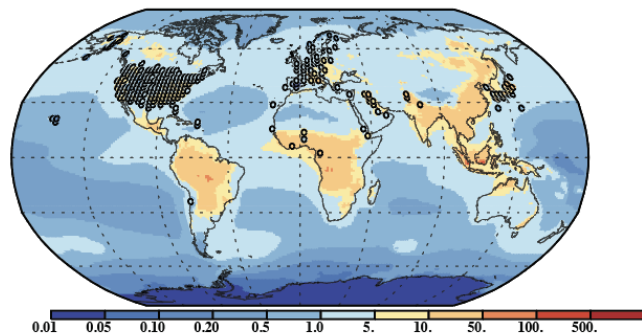
c. BC PM<sub>2.5</sub>  $\mu\text{g}/\text{m}^3$



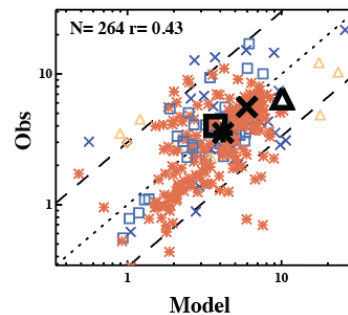
d. BC PM<sub>2.5</sub>  $\mu\text{g}/\text{m}^3$



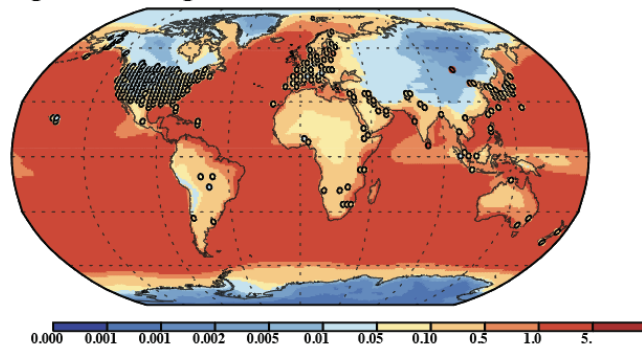
e. OM PM<sub>2.5</sub>  $\mu\text{g}/\text{m}^3$



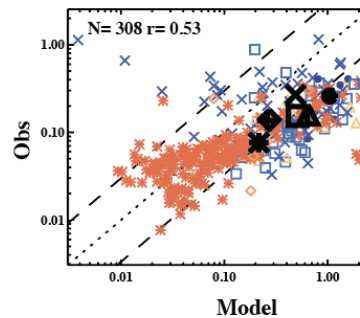
f. OM PM<sub>2.5</sub>  $\mu\text{g}/\text{m}^3$



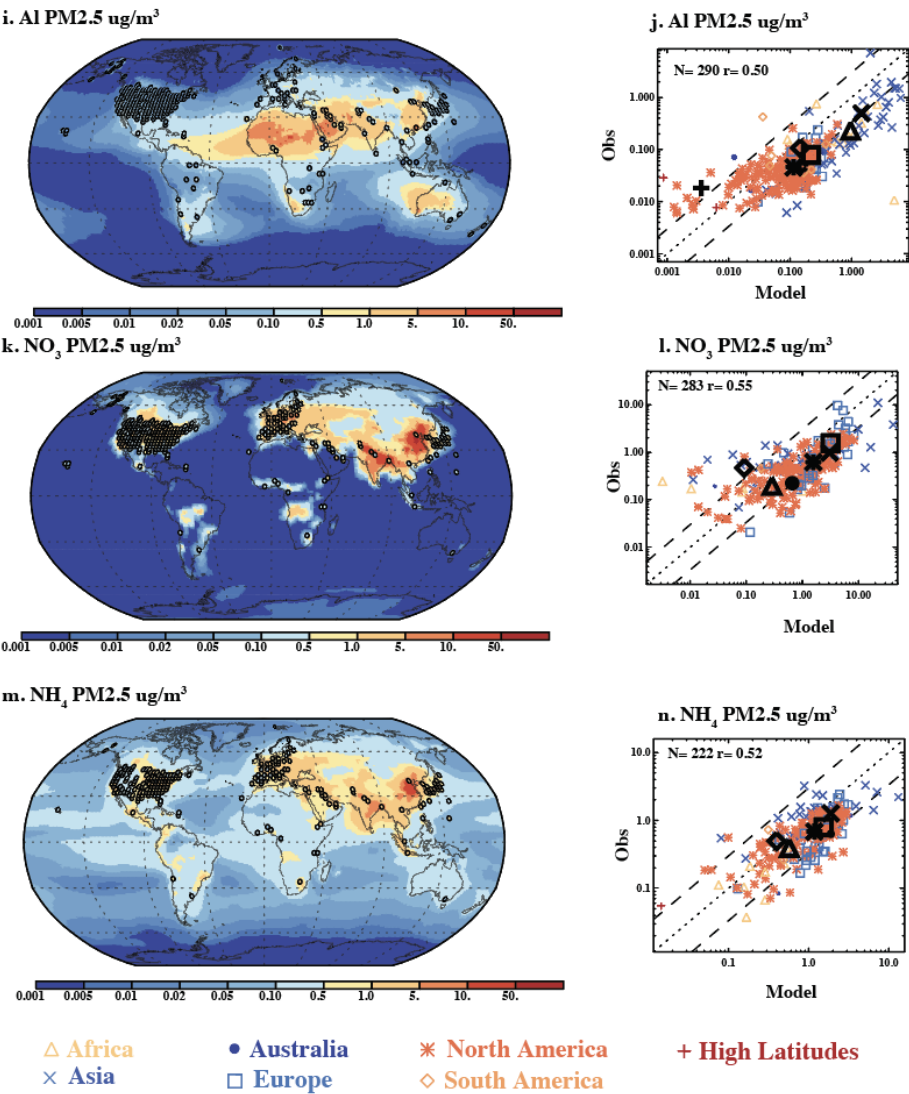
g. Na PM<sub>2.5</sub>  $\mu\text{g}/\text{m}^3$



h. Na PM<sub>2.5</sub>  $\mu\text{g}/\text{m}^3$



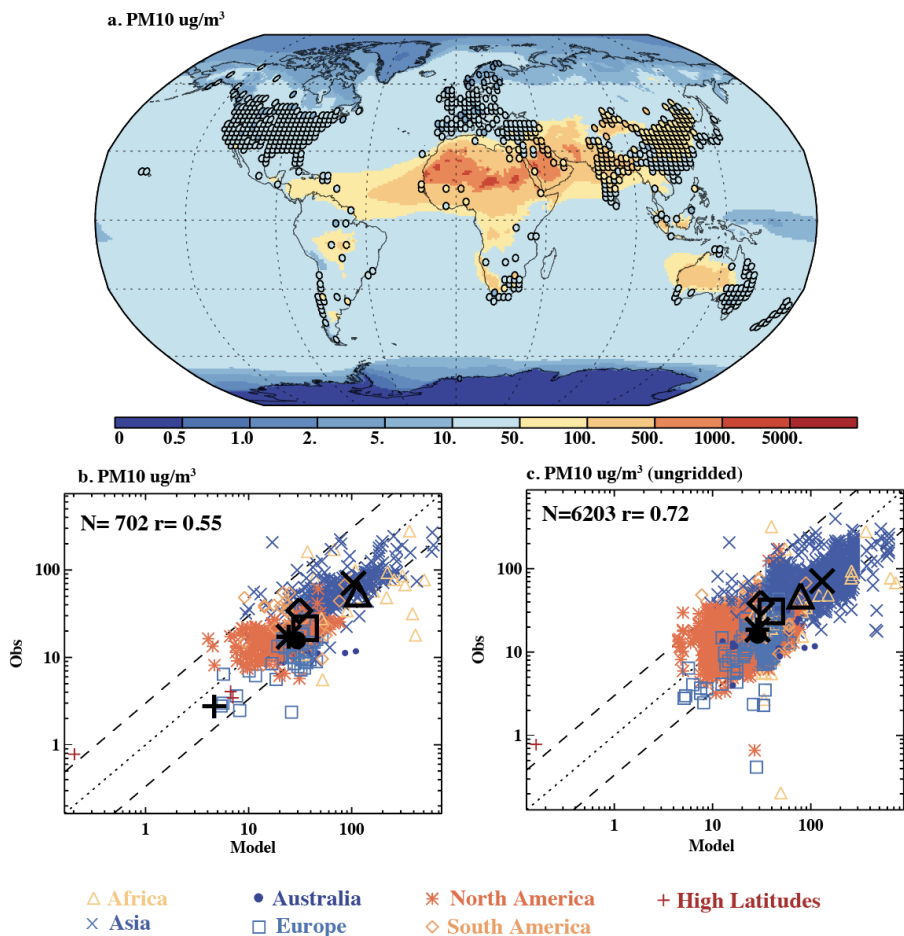




1645 **Figure 5:** Model results and gridded observations for different types of PM<sub>2.5</sub> in  $\mu\text{g}/\text{m}^3$  spatially mapped globally  
1646 where the model is plotted as the background and the observations are circles with the colors indicating the  
1647 amount PM<sub>2.5</sub> using the same scale for (a) SO<sub>4</sub><sup>2-</sup>, (c) BC (black carbon), (e) OM (organic material=1.8 times  
1648 organic carbon (OC)), (g) Na, (i) Al, (k) NO<sub>3</sub><sup>-</sup>, (m) NH<sub>4</sub><sup>+</sup>. A scatter plot comparison of the model (x-axis) to the

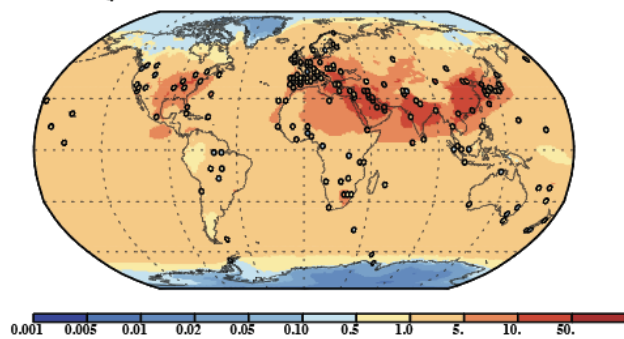
1649 observations (y-axis) is shown for the gridded observational data for for (b)  $\text{SO}_4^{2-}$ , (d) BC (f) OM, (h) Na, (j) Al,  
1650 (l)  $\text{NO}_3^-$ , (n)  $\text{NH}_4^+$ . In the scatter plots, the colors and symbols indicate the regions, the bold black symbols are  
1651 the average across each region (indicated by the symbol), the dotted line is the 1:1 line and the dashed lines are  
1652 the factor of 3 uncertainty estimates. More statistics are shown in Table S5, and the maps focused on specific  
1653 regions are available in Figure S3-S9 for  $\text{SO}_4^{2-}$ , BC, OM, Na, Al,  $\text{NO}_3^-$ , and  $\text{NH}_4^+$ , respectively.

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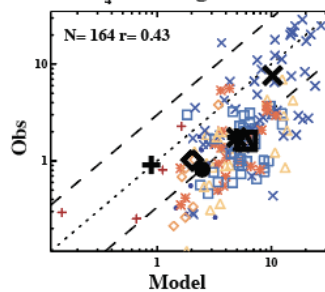


**Figure 6:** Model results and gridded observations for PM<sub>10</sub> in  $\mu\text{g}/\text{m}^3$  spatially mapped globally (a). A comparison of the model (x-axis) to the observations (y-axis) is shown for the gridded data (b) and including all stations (c). In the scatter plots, the colors and symbols indicate the regions, the bold black symbols are the average across each region (indicated by the symbol), the dotted line is the 1:1 line and dashed lines are the factor of 3 uncertainty estimates. More statistics are shown in Table S7, and maps focused on different regions are shown in Fig. S10

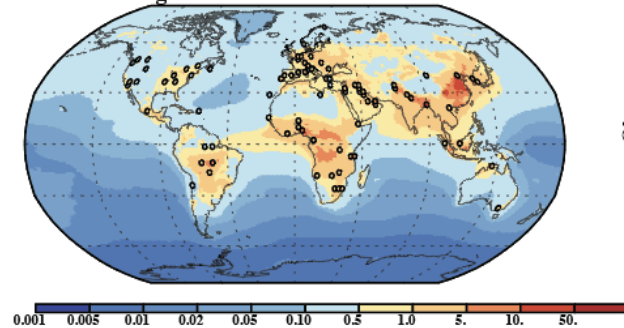
a.  $\text{SO}_4$  PM10  $\mu\text{g}/\text{m}^3$



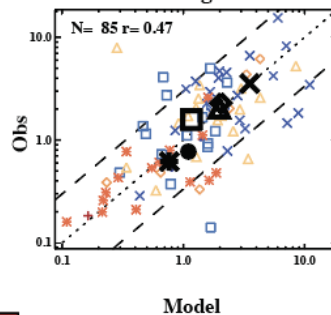
b.  $\text{SO}_4$  PM10  $\mu\text{g}/\text{m}^3$



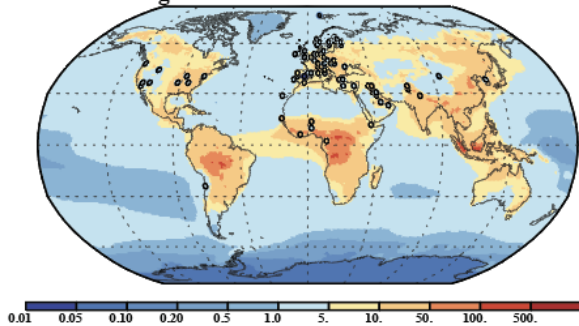
c. BC PM10  $\mu\text{g}/\text{m}^3$



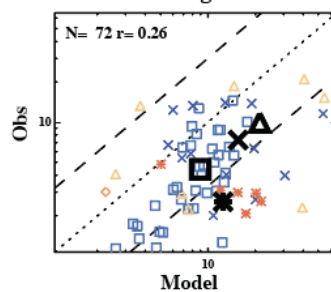
d. BC PM10  $\mu\text{g}/\text{m}^3$



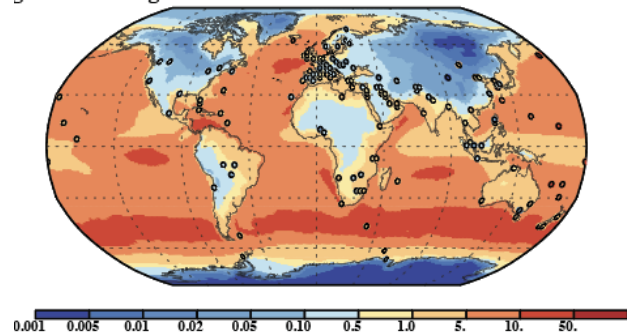
e. OM PM10  $\mu\text{g}/\text{m}^3$



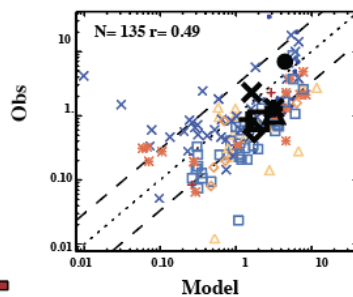
f. OM PM10  $\mu\text{g}/\text{m}^3$

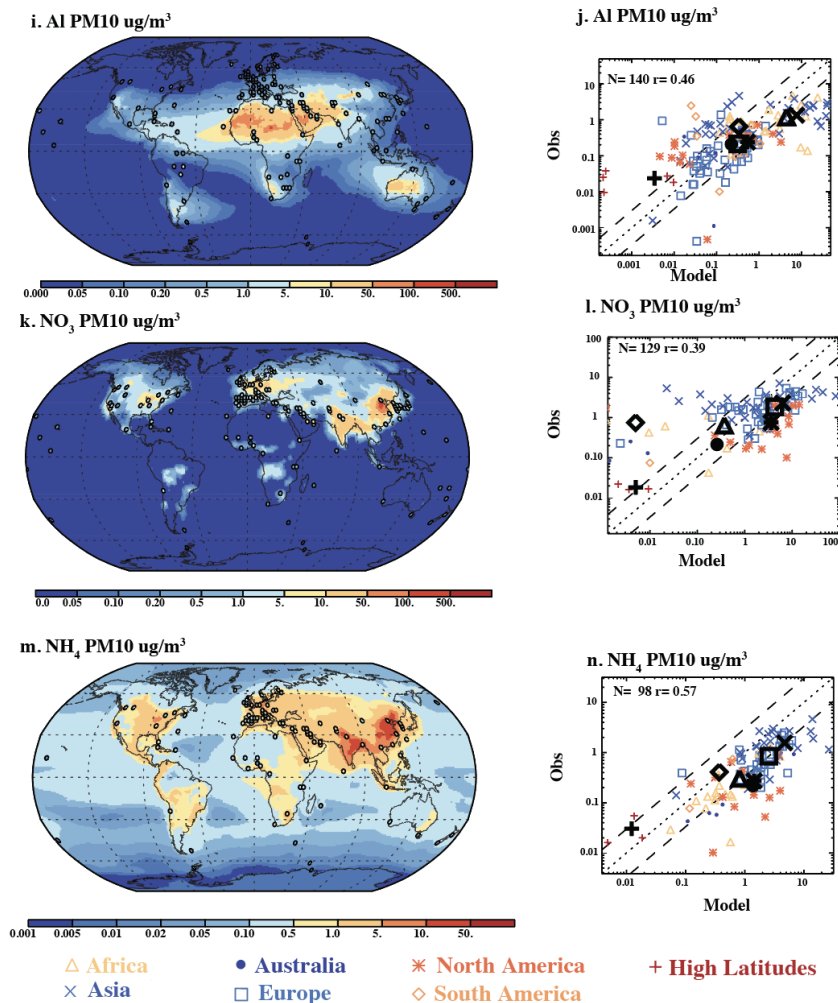


g. Na PM10  $\mu\text{g}/\text{m}^3$



h. Na PM10  $\mu\text{g}/\text{m}^3$

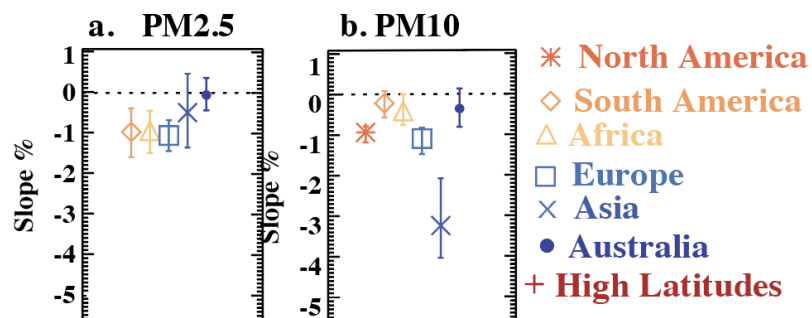




**Figure 7:** Model results and gridded observations for different types of  $\text{PM}_{10}$  in  $\mu\text{g}/\text{m}^3$  spatially mapped globally where the model is plotted as the background and the observations are circles with the colors indicating the amount  $\text{PM}_{10}$  using the same scale for (a)  $\text{SO}_4^{+2}$ , (c) BC (black carbon), (e) OM (organic material=1.8 times organic carbon (OC)), (g) Na, (i) Al, (k)  $\text{NO}_3^-$ , (m)  $\text{NH}_4^+$ . A scatter plot comparison of the model (x-axis) to the observations (y-axis) is shown for the gridded observational data for (b)  $\text{SO}_4^{+2}$ , (d) BC (f) OM, (h) Na, (j) Al, (l)  $\text{NO}_3^-$ , (n)  $\text{NH}_4^+$ . In the scatter plots, the colors and symbols indicate the regions, the bold black symbols are the average across each region (indicated by the symbol), the dotted line is the 1:1 line and the dashed lines are the

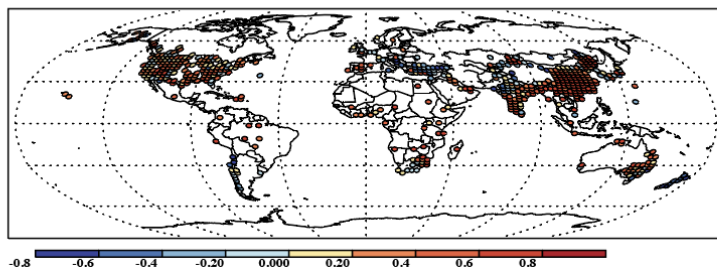
1671 factor of 3 uncertainty estimates. More statistics are shown in Table S7, and the maps focused on specific  
1672 regions are available in Figure S11-S17 for  $\text{SO}_4^{2-}$ , BC, OM, Na, Al,  $\text{NO}_3^-$ , and  $\text{NH}_4^+$ , respectively.

1673

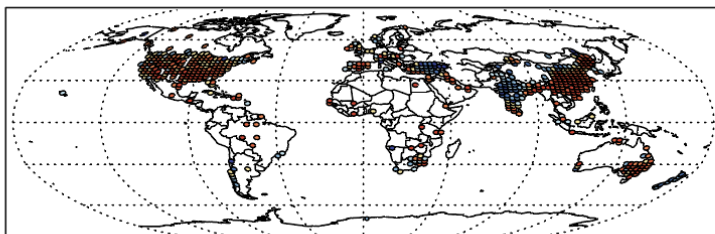


**Figure 8:** Trends in the observations of aerosols in different regions during the 1980-2000 and 2000-2024 time periods for PM<sub>2.5</sub> (a) and PM<sub>10</sub> (b). Error bars indicate the 1-sigma uncertainty using a Thiel regression approach.

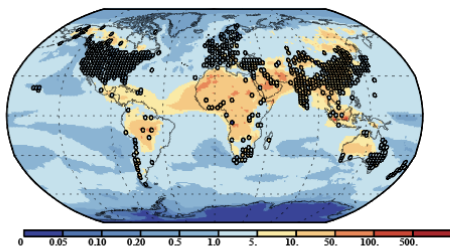
a. Seasonal Correlation PM2.5



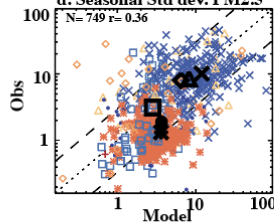
b. Seasonal Correlation PM10



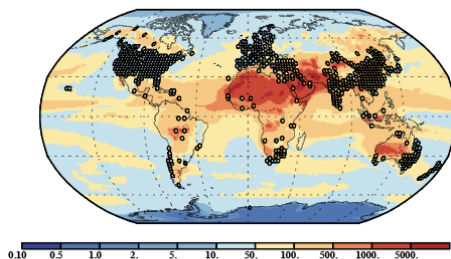
c. Seasonal Std. Dev. PM2.5



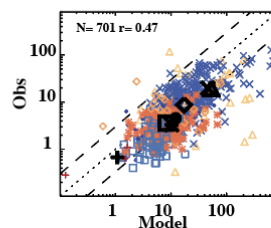
d. Seasonal Std dev. PM2.5



e. Seasonal Std. Dev. PM10



f. Seasonal Std. Dev. PM10

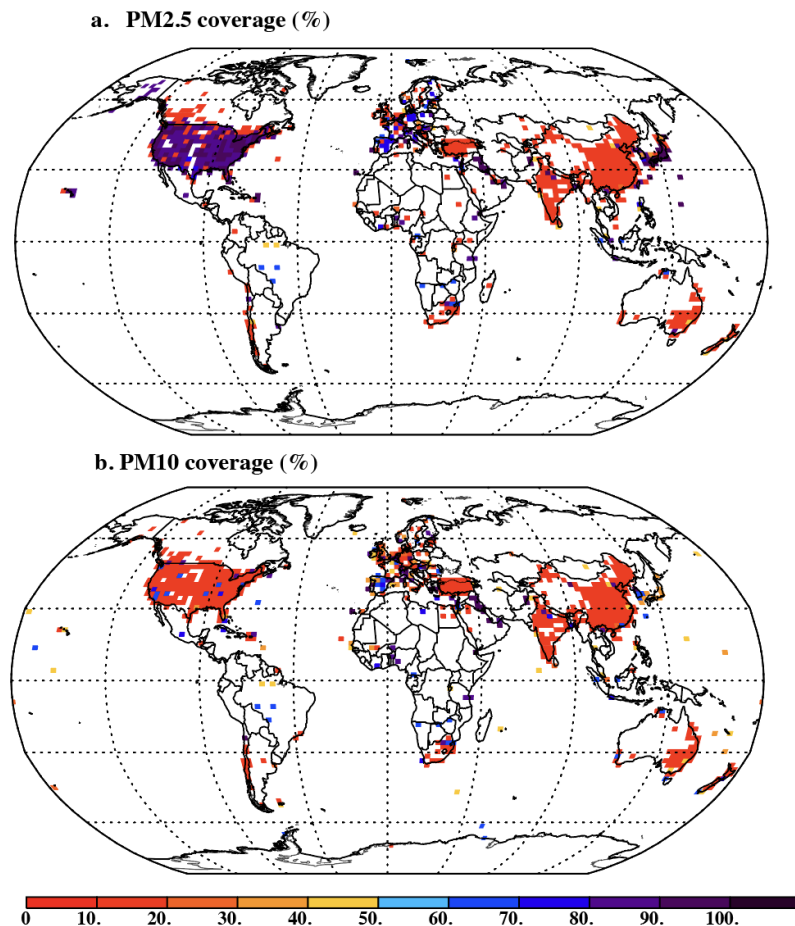


△ Africa    ● Australia    ✱ North America    + High Latitudes  
× Asia    □ Europe    ◇ South America

**Figure 9:** Model data comparison for the seasonal cycle. The correlation coefficient between the 12 climatological monthly means in the observations and the model for those station datasets with a larger seasonal cycle than within monthly variability (see Section 2.5 for more details), averaged to  $2^\circ \times 2^\circ$  grid for plotting for PM<sub>2.5</sub> (a) and PM<sub>10</sub> (b). A comparison of the magnitude seasonal cycle in the observations versus the model (defined as the standard deviation of the 12 climatological monthly means) spatially for (c) PM<sub>2.5</sub> and (e) PM<sub>10</sub> and a scatterplot for PM<sub>2.5</sub>



1685 (d) and PM<sub>10</sub> (f). The correlation coefficient is only calculated in locations where the standard deviation from the  
1686 seasonal cycle is stronger than the within month variability (see Section 2.5 for details).

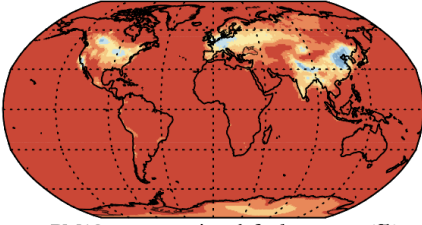


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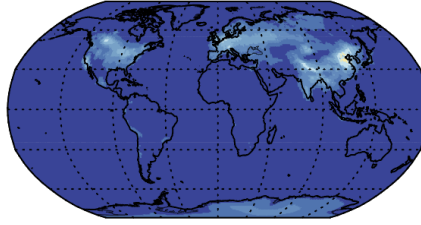
1688 **Figure 10:** Observational coverage (%) for gridded observations, showing within each grid box (2°x2°) the % of  
1689 the constituents that are measured assuming that PM, SO<sub>4</sub><sup>2-</sup>, BC, OM, Na, Al, NO<sub>3</sub><sup>-</sup>, and NH<sub>4</sub><sup>+</sup> are required to  
1690 constrain the PM distribution for (a) PM<sub>2.5</sub> and (b) PM<sub>10</sub>.

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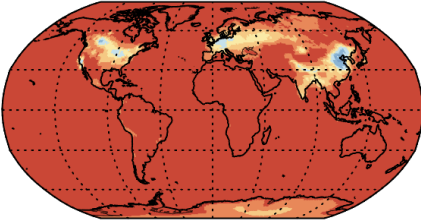
a. PM<sub>2.5</sub> concentration default sources (%)



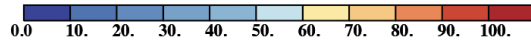
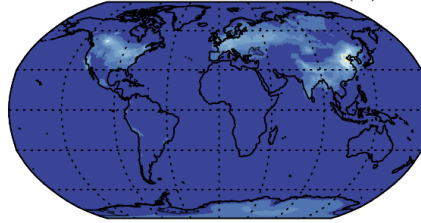
b. PM<sub>2.5</sub> concentration new sources (%)



c. PM<sub>10</sub> concentration default sources (%)



d. PM<sub>10</sub> concentration new sources (%)



**Figure 11:** Modelled estimates of what percent of the surface concentration of PM<sub>2.5</sub> is considered in the default CAM6 climate model (a) or is new in this study (b). Similarly PM<sub>10</sub> is shown for the default model (c) and new sources in this study (d). The new sources added in this study are the nitrogen oxides as described in Section 2.3.

| Composition     | Measurement Method                                                                                   | Variables        |  | Example Networks      | Example Citations                         |
|-----------------|------------------------------------------------------------------------------------------------------|------------------|--|-----------------------|-------------------------------------------|
| Fine and Coarse | Stacked Filter Unit (SFU)                                                                            | Fine, Coarse     |  | U. Gent               | Maenhaut et al., 2002a                    |
| PM2.5 and PM10  | Reference Method/Federal Equivalent Method (FRM/FEM),                                                | PM2.5, PM10      |  | IMPROVE, CASNET, EMEP | Hand et al, 2019, Putaud et al., 2004     |
| PM2.5 and PM10  | Hi Vol Sampler                                                                                       |                  |  | EMEP, SINCA           | Putaud et al., 2004                       |
| Elemental       | Particle Induced x-ray emission Spectrometry (PIXE), Instrumental nuclear activation analysis (INAA) | Al, S, Na        |  | U. Gent, EMEP         | Maenhaut et al., 2002a                    |
| Elemental       | Inductively Coupled Plasma-Mass Spectrometry (ICP-MS)                                                | Al, S, Na        |  | EMEP, SPARTAN         | Putaud et al., 2004; Phillip et al., 2017 |
| Elemental       | XRF                                                                                                  | Al, S, Na        |  | IMPROVE, CASNET       | Hand et al, 2019                          |
| Chemistry       | Ion Chromatography                                                                                   | SO4--, NO3-, NHr |  | IMPROVE, CASNET, EMEP | Hand et al, 2019, Putaud et al., 2004     |
| Carbonaceous    | Thermal Optical Reflectance                                                                          | EC, OC           |  | IMPROVE, CASNET       | Hand et al, 2019                          |
|                 | Evolved Gas Analysis Non-dispersive Infrared (EGA+NDIR)                                              | OC, EC           |  | EMEP                  | Putaud et al., 2004                       |
|                 |                                                                                                      |                  |  |                       |                                           |

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1702 Table 2: Global Aerosol Modelling Budgets

1703 Global modelled deposition (Tg/year), percentage of aerosol that is PM<sub>2.5</sub>, and globally and annually averaged surface  
1704 concentration (µg/m<sup>3</sup>) and aerosol optical depth for each of the sources used in the model. An asterisk indicates that there  
1705 are additions to the model from the default CAM6.

|  | PM <sub>10</sub>        | PM <sub>2.5</sub> |                              |                   |
|--|-------------------------|-------------------|------------------------------|-------------------|
|  | Deposition<br>(Tg/year) | %                 | Conc<br>(µg/m <sup>3</sup> ) | AOD<br>(unitless) |

|                                   |      |     |      |       |
|-----------------------------------|------|-----|------|-------|
| Sulfate                           | 121  | 100 | 2.1  | 0.018 |
| Black carbon                      | 10   | 100 | 0.5  | 0.009 |
| Primary<br>organic<br>aerosol     | 34   | 100 | 1.6  | 0.008 |
| Secondary<br>organic<br>aerosol   | 37   | 100 | 1.0  | 0.007 |
| Sea salts                         | 2520 | 3   | 13.0 | 0.045 |
| Dust                              | 2870 | 1   | 19.4 | 0.030 |
| NH <sub>4</sub> NO <sub>3</sub> * | 20   | 100 | 0.4  | 0.013 |

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