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Global Spatial Variation in the $PM_{2.5}$ to AOD Relationship Strongly Influenced by Aerosol Composition

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Abstract Ambient fine particulate matter ($PM_{2.5}$) is the leading global environmental determinant of mortality. However, large gaps exist in ground-based $PM_{2.5}$ monitoring. Satellite remote sensing of aerosol optical depth (AOD) offers information to fill these gaps worldwide, when augmented with a modeled $PM_{2.5}$ to AOD relationship (η). This study aims to understand the spatial pattern and driving factors of η from both observations and modeling. A global observational estimate of η for the year 2019 is inferred from 6,118 ground-based $PM_{2.5}$ measurement sites and satellite retrieved AOD from the MAIAC algorithm. A global chemical transport model, GEOS-Chem, in its high performance configuration (GCHP), is used to interpret the observed spatial pattern of annual mean η. Measurements and the GCHP simulation consistently identify a global population-weighted mean η of 92 – 100 μg/m^3, with regional values ranging from 60.3 μg/m^3 for North America to more than 130 μg/m^3 in Africa. The highest η is found in arid regions where aerosols are less hygroscopic due to mineral dust, followed by regions strongly influenced by surface aerosol sources. Relatively low η is found over regions distant from strong aerosol sources. The spatial variation of η is strongly influenced by aerosol composition driven by its effects on aerosol hygroscopicity. Sensitivity tests with globally uniform parameters reveal that aerosol composition leads to the strongest η spatial variability, with a population-weighted normalized mean difference of 12.3 μg/m^3, higher than that from aerosol vertical profile (8.4 μg/m^3), reflecting the determinant composition effects on aerosol hygroscopicity and aerosol optical properties.

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1 Introduction

Exposure to ambient fine particulate matter ($PM_{2.5}$) has been recognized as the predominant environmental risk factor for the global burden of disease, leading to millions of deaths annually (Murray et al., 2020; Burnett et al., 2018). Even at low $PM_{2.5}$ concentrations, long-term exposure can increase circulatory and respiratory related mortality (Pinault et al., 2016; Christidis et al., 2019; Weichenthal et al., 2022). Despite the importance of $PM_{2.5}$, many of the world's countries do not provide publicly accessible $PM_{2.5}$ data (Martin et al., 2019). Satellite remote sensing of aerosol optical depth (AOD), an optical measure of aerosol abundance, offers information about the distribution of $PM_{2.5}$ (Kondragunta et al., 2022). A large community relies upon the spatial distribution of $PM_{2.5}$ concentrations inferred from satellite remote sensing for health impact assessment and epidemiological analyses of long-term exposure (Murray et al., 2020; Burnett et al., 2018; Hao et al., 2023; Cohen et al., 2017). Quantitative application of satellite AOD for long-term characterization of the spatial distribution of $PM_{2.5}$ would benefit from a better understanding of the factors affecting the $PM_{2.5}$-AOD relationship.

The relationship between satellite AOD and surface $PM_{2.5}$ can be established through a statistical method, a geophysical method, or their combination. A statistical method uses ground-based monitors for training and is well suited for regions with dense monitors (Xin et al., 2014; Hu et al., 2014; Di et al., 2016). A geophysical approach utilizes a chemical transport model to simulate the relationship (η) between $PM_{2.5}$ and AOD for application to satellite AOD (van Donkelaar et al., 2006, 2010; He et al., 2021), and thus depends on accurate model representation of η. van Donkelaar et al. (2015, 2016) combined the two methods by applying geographically weighted regression (GWR) on the geophysical $PM_{2.5}$, which further constrains geophysical $PM_{2.5}$ using ground measurements and other predictors. However, accuracy of geophysical $PM_{2.5}$ remains critical over vast areas with sparse monitoring, and knowledge about the factors affecting η spatial variability are needed to guide improvements of modeled η and geophysical $PM_{2.5}$.

Previous studies have identified several factors that affect η variability, including aerosol vertical distribution, aerosol hygroscopicity, aerosol optical properties, and ambient meteorological factors such as relative humidity (RH), planetary boundary layer height (PBLH), wind speed, temperature, and fire events (Wendt et al., 2023; Jin et al., 2019; Guo et al., 2017; Ford and Heald, 2015; Li et al., 2015; van Donkelaar et al., 2013). Most studies focused on the temporal variability of η and

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found association with meteorological variables such as PBLH (Yang et al., 2019; He et al., 2021; Chu et al., 2015; Gupta et al., 2006; Zhang et al., 2009; Damascena et al., 2021). A few studies have examined the regional-scale spatial variation of η with meteorological, land type variables, and aerosol vertical profile in North America (Jin et al., 2020; van Donkelaar et al., 2006; Li et al., 2015) and China (Yang et al., 2019). To our knowledge, none have examined the factors at the global scale affecting the spatial variation of η or the effects of chemical composition.

In this work, we examine this knowledge gap about the spatial variation in η at a global scale. We first collect data from more than 6,000 $PM_{2.5}$ monitoring sites provided by nine networks and satellite AOD to obtain an observationally based map of η. We further interpret the global η using the GEOS-Chem model of atmospheric composition with recent improvements in aerosol size representation, $PM_{2.5}$ diel variation, and vertical allocation. By decomposing the simulated η, we identify 2 strong drivers of η spatial variability: aerosol composition and aerosol vertical profile. We conduct sensitivity tests using GEOS-Chem to study how the two factors vary globally and how they contribute to the spatial variation in η.

2 Methods

2.1 Ground Measured $PM_{2.5}$

We collect ground-based measurements of $PM_{2.5}$ for the year 2019 from which to produce observational constraints on η. We obtain $PM_{2.5}$ measurements from 7 regional networks and 2 global networks, as shown in Figure A1. For the United States, we access data from the United States Environmental Protection Agency's Air Quality System (https://www.epa.gov/outdoor-air-quality-data/download-daily-data), including both Federal Reference Method and non-Federal Reference Methods $PM_{2.5}$ (e.g. IMPROVE network). $PM_{2.5}$ data for Canada are from the Environment Canada's National Air Pollution Surveillance (NAPS) program. $PM_{2.5}$ data for Europe are from the European Environment Agency Air Quality e-Reporting system (https://www.eea.europa.eu/data-and-maps/data/aqereporting). Over mainland China, $PM_{2.5}$ measurements are downloaded from http://beijingair.sinaapp.com/, which provides instantaneous air quality data records from the National and Provincial Environmental Protection Agencies. Over India, $PM_{2.5}$ data are originally from the Central Pollution Control Board Continuous Ambient Air Quality Monitoring network and the U.S. embassies. Over Australia, observations are downloaded

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for the Northern Territory (http://ntepa.webhop.net/NTEPA/), Queensland (https://www.data.qld.gov.au/dataset/), and New South Wales (https://www.dpie. nsw.gov.au/air-quality/air-quality-data-services/data-download-facility). We require at least 5 days of measurements for each month for a monitor to be included. Additionally, we obtain $PM_{2.5}$ measurements over other regions provided by the World Health Organization (WHO) Global Ambient Air Quality Database and by the Surface PARTiculate mAtter Network Network (SPARTAN), which is co-located with the Aerosol Robotic Network (AERONET). SPARTAN also provides filter based $PM_{2.5}$ chemical composition, which is initially described in Snider et al., (2016). Subsequent developments to the sampling and analysis procedure of SPARTAN include an upgrade to the AirPhoton SS5 sampling station to use a cyclone inlet, an automated weighing system (MTL AH500E) to improve precision and throughput, additional black carbon analysis by Hybrid Integrating Plate/Sphere (White et al., 2016), trace metal elements measured by X-ray Fluorescence (Liu et al., 2024) and a global mineral dust equation (Liu et al., 2022). We require 50 days of coincident $PM_{2.5}$ and AERONET AOD measurements for a SPARTAN site to be included in our analysis.

2.2 Satellite AOD

We obtain AOD at 550 nm from the Multi-Angle Implementation of Atmospheric Correction (MAIAC) algorithm, which offers AOD at a high spatial resolution of 1 km worldwide over both land and coastal regions (Lyapustin et al., 2018). The radiances used in the retrieval are measured by the twin MODerate resolution Imaging Spectroradiometer (MODIS) instruments onboard the Terra and Aqua satellites. Terra follows a descending orbital path, crossing the equator at 10:30 local time, while Aqua is on an ascending orbit with 13:30 equatorial crossing local time. Both MODIS instruments offer a wide swath width of 2330 km, enabling nearly global daily coverage of the Earth (Sayer et al., 2014). $PM_{2.5}$ monitoring sites with annual mean satellite AOD less than 0.05 (background AOD level over land) are excluded to reduce the influence of retrieval uncertainties on our analysis.

2.3 AERONET AOD

AERONET is a worldwide sun photometer network that provides long-term measurement of AOD. We use the Version 3 Level 2 database, which includes an improved cloud screening algorithm

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(Giles et al., 2019). We sample AERONET AOD within ±15 min of the satellite overpass time and interpolate to 550 nm wavelength, based on the local Ångström exponent at 440 and 670 nm. For SPARTAN sites, we sample AERONET data coincidentally with SPARTAN aerosol composition to obtain the ground-based observation of η.

2.4 GEOS-Chem Simulation

We simulate η with the GEOS-Chem chemical transport model (www.geos-chem.org, last access: 26 October 2023), driven by offline meteorological data, MERRA-2, from the Goddard Earth Observing System (GEOS) of the NASA Global Modeling and Assimilation Office (Schubert et al., 1993). We use the high-performance configuration of GEOS-Chem (GCHP) (Eastham et al., 2018) version 13.4.0 (DOI: 10.5281/zenodo.7254268), which includes advances in performance and usability (Martin et al., 2022). The simulation is conducted for the year 2019, on a C90 cubed-sphere grid corresponding to a horizontal resolution of about 100 km, with a spin-up time of 1 month.

The GEOS-Chem aerosol simulation includes the sulfate-nitrate-ammonium (SNA) system (Fountoukis and Nenes, 2007), primary and secondary carbonaceous aerosols (Park et al., 2003; Wang et al., 2014; Pai et al., 2020), sea salt (Jaeglé et al., 2011), and natural (Fairlie et al., 2007; Meng et al., 2021) and anthropogenic (Philip et al., 2017) dust. The primary emission data are from the Community Emissions Data System ($CEDS_{GBD\text{-}MAPS}$; McDuffie et al., 2020). Emissions from stacks are distributed vertically (Bieser et al., 2011). Diel variation of anthropogenic emissions is included (Li et al., 2023). Resolution-dependent soil NO_x, sea salt, biogenic VOC, and natural dust emissions are calculated offline at native meteorological resolution to produce consistent emissions across resolutions (Weng et al., 2020; Meng et al., 2021). Biomass burning emissions use the Global Fire Emissions Database, version 4 (GFED4) at daily resolution (van der Werf et al., 2017). We estimate organic matter (OM) from primary organic carbon using an OM/OC parameterization (Philip et al., 2014; Canagaratna et al., 2015). For secondary aerosol components, the concentration at 2 m above the surface is used to calculate $PM_{2.5}$, following Li et al. (2023). A 50% reduction of the surface nitrate concentration is applied to account for the long-persisting bias in surface nitrate simulated by GEOS-Chem (Heald et al., 2012; Zhang et al., 2012; Zhai et al., 2021; Miao et al., 2020; Travis et al., 2022) and other models (Zakoura and Pandis,

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2018; Shimadera et al., 2014). Dry and wet deposition follows Amos et al. (2012), with a standard resistance-in-series dry deposition scheme (Wang et al., 1998). Wet deposition includes scavenging processes from convection and large-scale precipitation (Liu et al., 2001).

Global RH-dependent aerosol optical properties are based on the Global Aerosol Data Set (GADS) (Kopke P., 1997), as originally implemented by Martin et al. (2003), with updates for SNA and OM dry size (Zhu et al., 2023), hygroscopicity (Latimer and Martin, 2019), mineral dust size distribution (Zhang et al., 2013), and absorbing brown carbon (Hammer et al., 2016). We artificially increase simulated AOD by 0.04 globally to address a poorly understood systematic bias. $PM_{2.5}$ is calculated as the sum of each component at 35% RH to align with common measurement protocols.

2.5 Population

Global population information is obtained from the Gridded Population of the World provided by the NASA Socioeconomic Data and Applications Center (Center for International Earth Science Information Network - CIESIN - Columbia University, 2018).

2.6 Sensitivity Tests with Globally Uniform Parameters

We conduct sensitivity tests of factors affecting the spatial variation of η, with a focus on aerosol composition and aerosol vertical profile. To understand the relative importance of these factors, we impose a constant for each factor and simulate the corresponding η. The difference between the test scenario and the base scenario reflects the change due to variation of the factor. We use the global population-weighted mean (PWM) and population-weighted mean difference (PWMD) to summarize changes with a focus on relevance to population exposure:

$$X_{PWM} = \frac{\sum_j \sum_i P_{i,j} X_{i,j}}{\sum_j \sum_i P_{i,j}}$$

$$PWMD = \frac{\sum_j \sum_i P_{i,j} |X_{i,j} - Y_{i,j}|}{\sum_j \sum_i P_{i,j}}$$

where i and j are grid box identifiers. X and Y could be any variable of interest. $|X_{i,j} - Y_{i,j}|$ is the absolute value of their difference.

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The first test imposes globally uniform aerosol chemical composition calculated as the global PWM aerosol component fraction ($F_{s,k,PWM}$):

$$F_{s,k,PWM} = \frac{\sum_j \sum_i P_{i,j} F_{i,j,s,k}}{\sum_j \sum_i P_{i,j}}$$

where i, j, and k are grid box identifiers along latitude, longitude, and vertical layer. P represents population density in each grid box. F_S is the fraction of aerosol component S in total aerosol mass. This test keeps the total columnar aerosol mass and aerosol vertical profile unchanged.

The second test imposes a globally uniform aerosol vertical profile calculated as the PWM column relative vertical profile ($R_{s,k,PWM}$):

$$R_{s,k,PWM} = \frac{\sum_j \sum_i P_{i,j} R_{i,j,k,s}}{\sum_j \sum_i P_{i,j}}$$

where $R_{i,j,k,s}$ is the relative dry mass ratio compared to the surface. The total mass loading and relative chemical composition are unchanged.

We analyze global and regional variations of η, as well as that for the driving factors. The definition of each region used in this study is summarized in Figure A2.

3 Results and Discussion

3.1 Global Spatial Pattern of η

The top panel of Figure 1 shows the observationally based annual mean η, inferred from the ratio of ground-measured $PM_{2.5}$ to MAIAC AOD. Measurements are most dense in North America, Europe, and East Asia. The annual mean η varies substantially, from 7.8 μg/m^3 in Hawaii to 321 μg/m^3 in Central Asia, with a PWM of 95.7 μg/m^3 and standard deviation (σ) of 36.6 μg/m^3. Higher PWM η of 132 μg/m^3 to 154 μg/m^3 exist over desert regions such as the North Africa and West Asia, followed by PWM η of 97 μg/m^3 to 121 μg/m^3 by regions strongly influenced by anthropogenic aerosols, such as East Asia, South Asia (Figure A3 and Table A1). Over North America, η is around 60 μg/m^3 in the east and in California, which is more than double that in the Rockies, driven by the spatial pattern of surface $PM_{2.5}$ (Figure A3). The PWM η in North America of 60.3 μg/m^3 is about 30% lower than the global PWM. The η pattern found here is similar to that reported by Jin et al. (2020) for the U.S. In Europe, η also varies noticeably between the east and

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the west, driven by the spatial pattern of surface $PM_{2.5}$, as $PM_{2.5}$ increases by 60% from west to east while AOD increases by only 8%. The PWM η in Europe is 94.0 $\mu g/m^3$, slightly lower than the global PWM. In Asia, measured η is concentrated in China and India. In China, the η spatial pattern shows a clear distinction between the northern and southern regions, driven by the higher AOD in the south, where relative humidity is high. A similar η spatial pattern and a negative correlation between η and RH are reported by Yang et al. (2019). In India, η is highest in the northwest, with a PWM η of 128 $\mu g/m^3$, and decreases to about 80 $\mu g/m^3$ toward the east and the south. Both $PM_{2.5}$ and AOD follow the same spatial pattern, while $PM_{2.5}$ exhibits a stronger decreasing tendency. PWM η in Asia is 100 $\mu g/m^3$, the highest among the populous regions and 4.5% higher than the global PWM. Globally, from west to east, η increases by about 62%, despite that both $PM_{2.5}$ and AOD increased more than threefold (Figure A5). The coefficient of variation (standard deviation divided by mean) in η is higher in Europe ($\mu = 0.31$) and Asia ($\mu = 0.34$), than North America ($\mu = 0.23$, Figure A5).

The bottom panel in Figure 1 shows the GCHP simulated η, the ratio between simulated 24-hour mean surface $PM_{2.5}$ and simulated total column AOD at satellite overpass time. The simulation generally reproduces the global observations of η with a tendency for high values in arid regions influenced by dust and low values in regions distant from strong surface sources. The global simulated PWM η is 4% higher than the observations (99.5 $\mu g/m^3$ vs. 95.7 $\mu g/m^3$), mostly driven by an overestimation in Asia (107 $\mu g/m^3$ vs. 100 $\mu g/m^3$), that reflects an overestimation of PWM $PM_{2.5}$ (47.0 $\mu g/m^3$ vs. 43.6 $\mu g/m^3$). The simulation generally reproduces the regional spatial pattern in North America and Asia but underestimates the η variability in Europe as it overestimates η in central Europe and underestimates η in Eastern Europe, due to positive bias in simulated $PM_{2.5}$ in central Europe and positive bias in simulated AOD in Eastern Europe. Nonetheless, the PWM η in Europe (84.1 $\mu g/m^3$) is within 11% of observations. Globally, there is overall consistency between the simulated η and observed η, with a correlation of 0.64, resulting in a high degree of consistency between geophysical $PM_{2.5}$ and measured $PM_{2.5}$ ($r = 0.90$, Figure A5).

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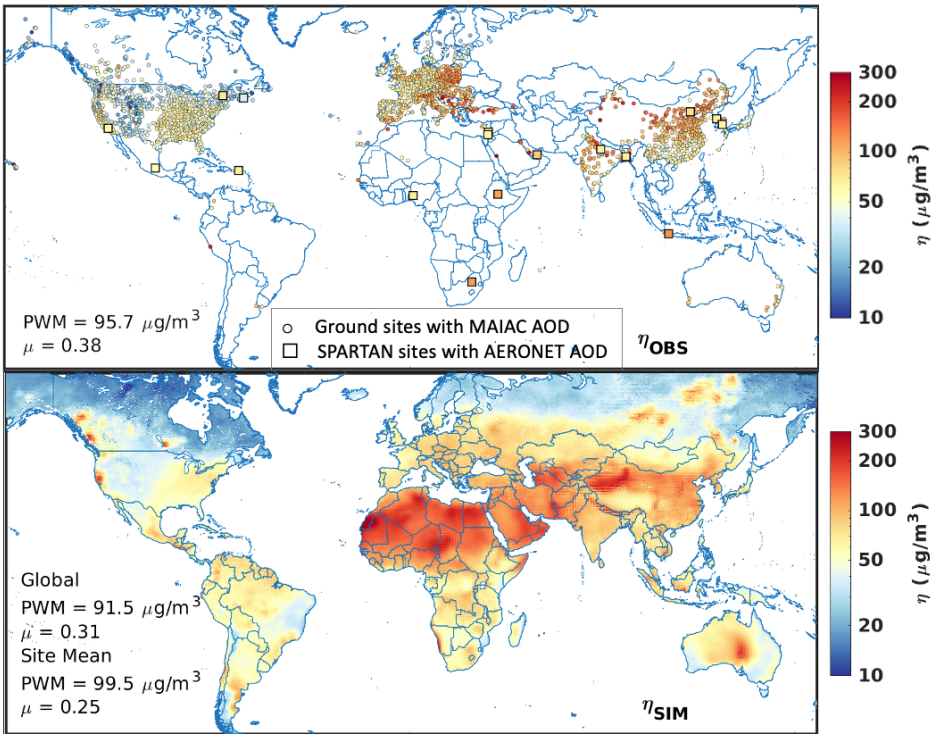

Figure 1. Observed (top) and simulated (bottom) annual mean η for 2019. Circles represent ground measurement sites from regional networks or the World Health Organization. Squares represent co-located ground measured $PM_{2.5}$ from SPARTAN and AOD from AERONET. PWM = population-weighted mean, μ = coefficient of variation (standard deviation divided by mean).

We explore the dominant driving factors for η spatial variation by calculating the spatial correlation between each candidate factor and the observation-based η. Candidate factors examined include meteorological fields (MERRA-2), aerosol vertical profile, and aerosol composition as collected from the GCHP simulation or SPARTAN. Meteorological fields include those commonly considered to represent the temporal variation in η, such as PBLH, RH at 700 hPa, wind speed at 10 m, and temperature at 2 m (Yang et al., 2019; He et al., 2021; Chu et al., 2015; Damascena et al., 2021). The aerosol vertical profile is represented as the AOD fraction below 1 km (AOD % below 1 km). Aerosol composition includes SNA, OM, dust, black carbon, and sea salt, all represented as the fractional contributions (%) to surface $PM_{2.5}$. Figure 2 shows the spatial correlation of annual mean factors versus observation-based η. Aerosol components,

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particularly those with strong primary sources (dust, OM, and black carbon), exhibit the strongest correlations (>0.3) with observationally based η. Significant positive correlations are found for mineral dust and black carbon, both of which are non- or weakly-hygroscopic. Significant negative correlations are found for organic matter and sea salt, reflecting a weak connection between surface concentrations and AOD aloft. Processes are further discussed in sections 3.2 and 3.4. The aerosol vertical profile exhibits a moderate correlation with η (0.18), which is notably higher than any meteorological factors (<0.12). Ground-based data from SPARTAN and AERONET corroborate the correlation between aerosol composition and η (Figure A6).

The indicators of spatial variation in η found here differ from that for temporal variation of η in prior work (e.g. He et al. 2021), reflecting the different processes involved. Meteorological parameters drive short-term variability in the aerosol vertical profile, such as day-to-day variation in mixed layer depth or in advection from a point source. In contrast, the spatial variation in annual mean η reflects the spatial variation in processes affecting the long-term relation of surface $PM_{2.5}$ at controlled RH of 35% with AOD at ambient RH. Aerosol composition and the aerosol vertical profile reflect spatial variation in aerosol hygroscopicity, mass extinction efficiency, and sources. The following sections explore how aerosol composition and aerosol vertical profile vary globally and examine how they affect the spatial pattern of η by conducting two sensitivity tests. In each sensitivity test, we replace the spatial variability of a factor with a globally uniform value. The variability of aerosol composition and aerosol vertical profile are discussed in sections 3.2 and 3.3, respectively. The sensitivity test results are discussed in section 3.4.

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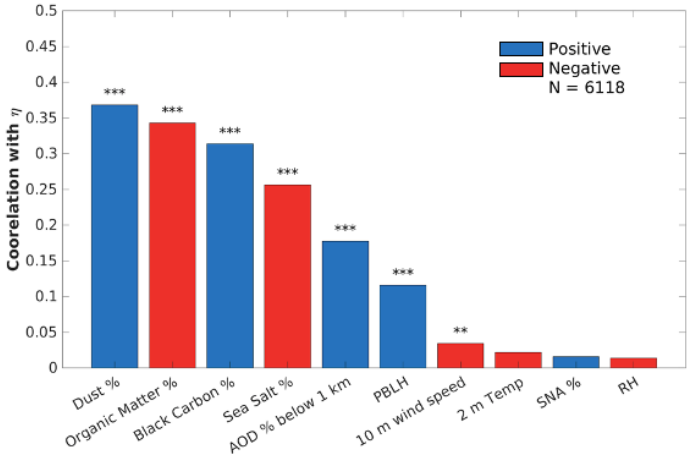

Figure 2. Spatial correlation between annual mean modeled parameters and observationally-based η. Blue bars indicate positive correlations. Red bars indicate negative correlations. Stars above each bar indicate the p-value associated with each correlation. '***' indicates the p-value is lower than 0.001 and '**' indicates lower than 0.01.

3.2 Spatial Variability in Aerosol Composition

Figure 3 shows the simulated PWM aerosol composition globally and regionally, as well as the global area-weighted mean (AWM). The top panel shows the compositional contribution to $PM_{2.5}$. Globally, dust is the leading PWM $PM_{2.5}$ component (34.7%), followed by OM (31.9%) and SNA (29.3%). The bottom panel shows the compositional contribution to AOD. PWM AOD composition is more evenly distributed, with more contribution from SNA (49.9%), followed by OM (27.2%) and dust (16.1%). Overall, more hygroscopic aerosols such as SNA tend to contribute a larger fraction of AOD which is at ambient RH, while less hygroscopic aerosols, such as mineral dust tend to contribute a larger fraction of $PM_{2.5}$ which is at controlled RH of 35%. The AWM $PM_{2.5}$ and AOD composition exhibit weaker contributions from SNA, primarily reflecting a larger contribution from dust in remote regions than in more densely populated areas. Over populous regions such as North America, Europe, and Southeast Asia, there are greater SNA and OM fractions than the global mean (Figure 3). Arid regions, such as West Asia, the Middle East, North Africa, and Sub-Saharan Africa, have large fractions of non-hygroscopic mineral dust that (1) reduce aerosol mass extinction efficiency, yielding less AOD per unit mass, and (2) are unaffected

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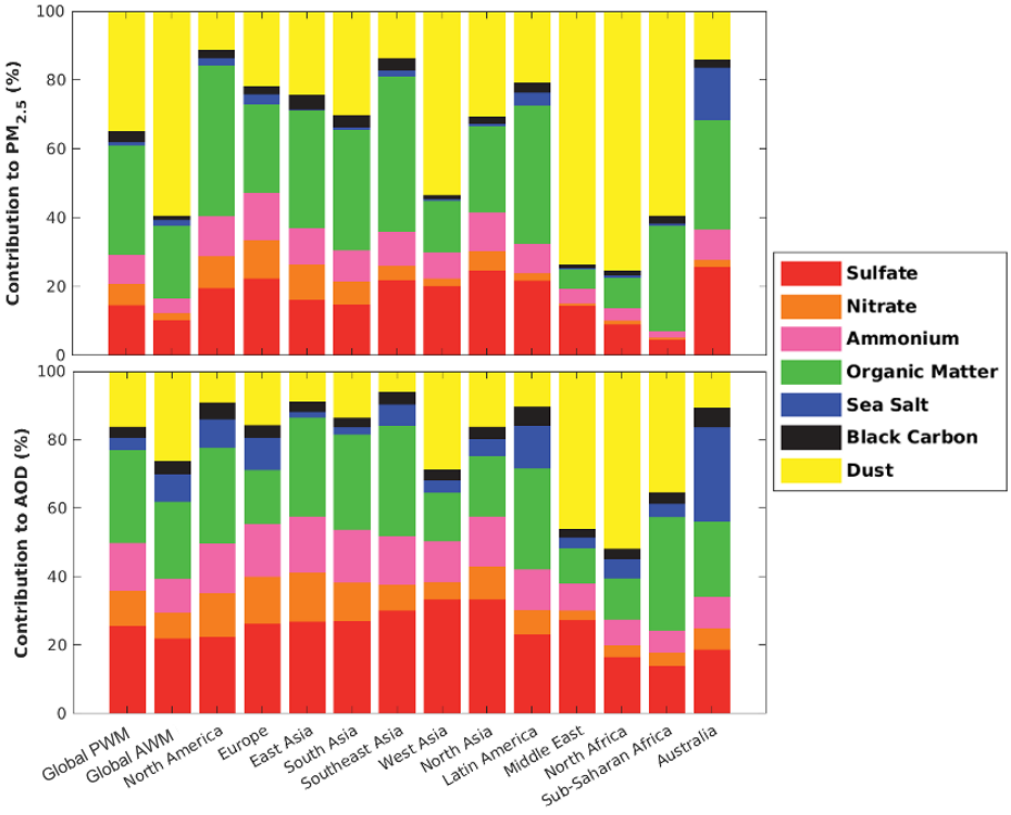
274 by the controlled RH of $PM_{2.5}$. Both of these factors increase η in dusty regions compared with
275 regions dominated by hygroscopic SNA aerosols.



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277 Figure 3. Global and regional PWM contributions of aerosol composition to surface $PM_{2.5}$ (top) and AOD
278 (bottom). The global area-weighted mean (AWM) over land is also included as the second column.

279 **3.3 Spatial Variability in Aerosol Vertical Profile**

280 Figure 4 shows the AOD fraction below 1 km in the GEOS-Chem simulation. Globally, 35.3% of
281 the PWM AOD is below 1 km. The PWM value is greater than the AWM value since populated
282 areas tend to have more surface emissions of particles and precursors. Over North America, Europe,
283 and East Asia, the PWM surface AOD fractions are much higher than the medians and AWM,
284 indicating high spatial heterogeneity between urban and remote areas. Europe exhibits the highest
285 variation and the largest discrepancy between PWM and AWM, reflecting the largest spatial
286 heterogeneity in aerosol vertical profile, driven by influences from regional pollution, marine

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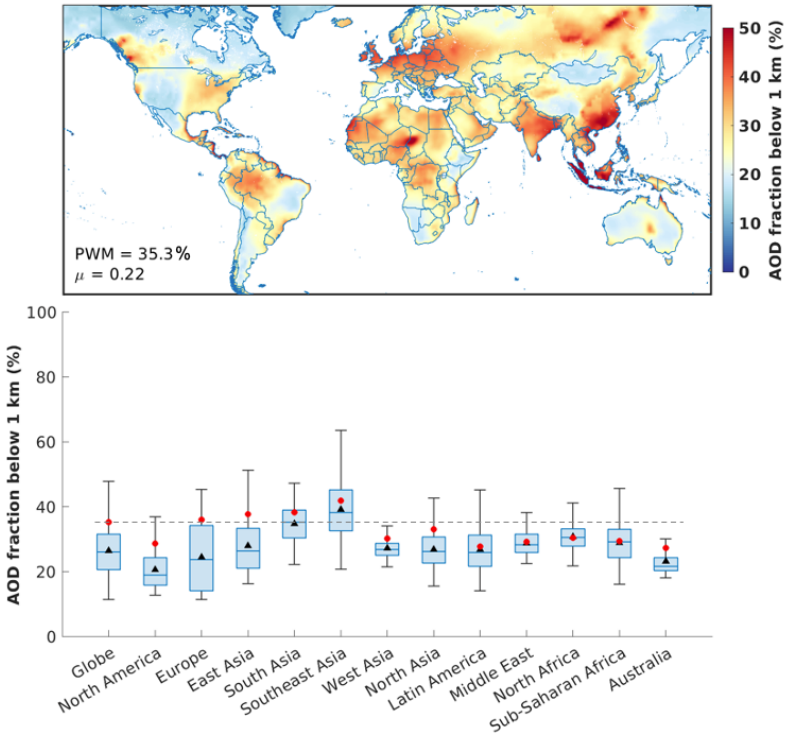
287 aerosols, and transported dust (Zhao et al., 2018). Southeast Asia has the highest surface AOD
288 fraction and a large variation. Local sources, long-range transported dust, and the influence of
289 trade winds all contribute to the unique spatial variation in aerosol vertical profile in this region
290 (Nguyen et al., 2019; Banerjee et al., 2021). Globally, PWM values exhibit less variation than
291 AWM, indicating moderate variation in aerosol profile across populous areas.



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293 Figure 4. (Top) Map of AOD fraction below 1 km. (Bottom) Global and regional statistics for AOD
294 fraction below 1 km. Black triangles show the area-weighted mean. Red circles show the PWM. The line
295 inside each box is the sample median. Each box's top and bottom edges are the 75 and 25 quartiles,
296 respectively. Vertical bars are the maximum and minimum values within 1.5 times the interquartile range.
297 The dashed line indicates global PWM.

298 **3.4 Sensitivity Tests with Globally Uniform Parameters**

299 Figure 5 shows the global changes in the spatial variation in η due to variations in aerosol chemical
300 composition (top) and aerosol vertical profile (bottom). Globally, neglect of spatial variation in

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aerosol composition induces a 12.3 μg/m^3 PWMD in η spatial variation. Both $PM_{2.5}$ and AOD are strongly affected by aerosol composition, following a similar spatial pattern (Figure A7). Over mid- and low-latitude areas, the change in AOD is stronger than in $PM_{2.5}$, which gives the opposite pattern in the η. Neglect of spatial variation in chemical composition reduces η over North Africa and the Middle East, desert regions where aerosols contain more weakly hygroscopic components such as mineral dust, compared to populous areas, which contain more secondary inorganic aerosol (Figure 3). For smaller deserts in the Southwest U.S., Argentina, and Southwest Africa, the dust fractions of surface aerosols are higher than the global mean (36%, 76%, and 49%, respectively), but the dust fraction for AOD is similar to the global mean (15%, 25%, and 14%, respectively). Therefore, neglect of the spatial variation of chemical composition increases η over these small deserts by increasing the fraction of hygroscopic components in $PM_{2.5}$ and leaving AOD almost unchanged (Figure A7). It also reduces η over the boreal forests and the Amazon, where surface aerosols contain little dust and are more hygroscopic compared to populous areas (Figure 3). Neglect of spatial variation in chemical composition increases η over the eastern U.S. and eastern China, where $PM_{2.5}$ contains more hygroscopic SNA and less dust than the global mean. It also increases η in coastal regions where aerosol contains more hygroscopic sea salt than the global mean.

Neglect of spatial variation in the aerosol vertical profile induces an 8.4 μg/m^3 PWMD in η spatial variation (Figure 5), following the spatial pattern of the change in surface $PM_{2.5}$ (Figure A8). The most apparent feature is an increase in η throughout the remote northern hemisphere, driven by an increased aerosol fraction near the surface where the fraction is normally small (Figure 4). The uniform aerosol vertical profile decreases η over northern Africa and biomass burning regions of the boreal forests, the Amazon, and Indonesia, driven by a decreased aerosol fraction near the surface in regions where that fraction is normally high.

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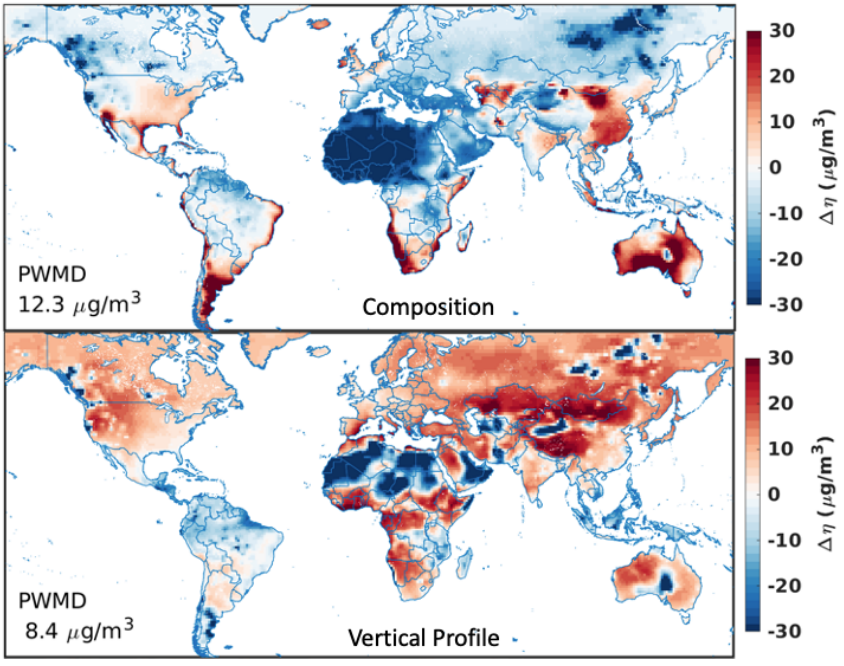

Figure 5. Changes in η (test -base) for each sensitivity test. In the first test, a global PWM aerosol composition replaces the actual composition (top). In the second test, a global PWM aerosol profile replaces the actual profiles (bottom). Numbers inset indicate population-weighted mean difference (PWMD).

Conclusion

Understanding the global variation of the $PM_{2.5}$ and AOD relationship (η) offers insight into the geophysical inference of $PM_{2.5}$ from satellite AOD observations. We collected ground-based $PM_{2.5}$ measurements from 6188 sites and MODIS MAIAC satellite AOD throughout the year 2019 to obtain, for the first time, a global scale observationally based η map. Observed annual mean η ranges from 7.8 μg/m^3 in Hawaii to 321 μg/m^3 in Central Asia. We observed enhanced η of 132 μg/m^3 to 154 μg/m^3 over arid regions such as North Africa and West Asia, due to their low aerosol extinction efficiency. Moderate η of 97 μg/m^3 to 121 μg/m^3 was found in industrial areas such as East Asia and South Asia, where anthropogenetic emissions increase the near-surface $PM_{2.5}$ concentrations. Over remote areas, low η (< 50 μg/m^3) was usually observed.

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We simulated the global annual mean η with the GEOS-Chem chemical transport model in its high performance configuration (GCHP). The simulation generally represented observed η with PWM within 4% (99.5 μg/m^3 vs 95.7 μg/m^3), a correlation of 0.64 over the 6,118 measurement sites, and a slope of 0.81. We examined the correlation between simulation and measurements to identify the two most impactful drivers for η spatial variation - aerosol composition and aerosol vertical profile, both of which strongly affect the annual mean relation of columnar AOD at ambient RH with surface $PM_{2.5}$ at controlled RH of 35%. We conducted sensitivity tests by eliminating the spatial variation of each driver and quantified the impact on η spatial variability. Imposing a globally uniform aerosol composition led to more pronounced changes (PWMD = 12.3 μg/m^3) reflecting how changes in aerosol composition affect both AOD and surface $PM_{2.5}$, due to the effects of aerosol hygroscopicity on both quantities. Imposing a globally uniform aerosol vertical profile had a moderate effect (PWMD = 8.4 μg/m^3), reflecting changes in the fraction of aerosol near the surface.

These findings motivate additional efforts to develop the simulation of aerosol composition and aerosol vertical profile. Promising avenues include: (1) enhancing global long-term measurements of $PM_{2.5}$ chemical composition to evaluate and improve simulations, (2) exploiting new and emerging information about aerosol type from satellite remote sensing (e.g. PACE, MAIA), (3) advancing simulations at finer spatial resolution to better represent processes affecting aerosol composition and vertical profile, (4) leveraging aircraft, lidar, and collected AOD-to-$PM_{2.5}$ measurements for constraints on the vertical profile, and (5) exploiting nascent capabilities in applying satellite remote sensing (e.g. TROPOMI, TEMPO, GEMS) for top-down constraints on emissions that affect aerosol composition.

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Appendix

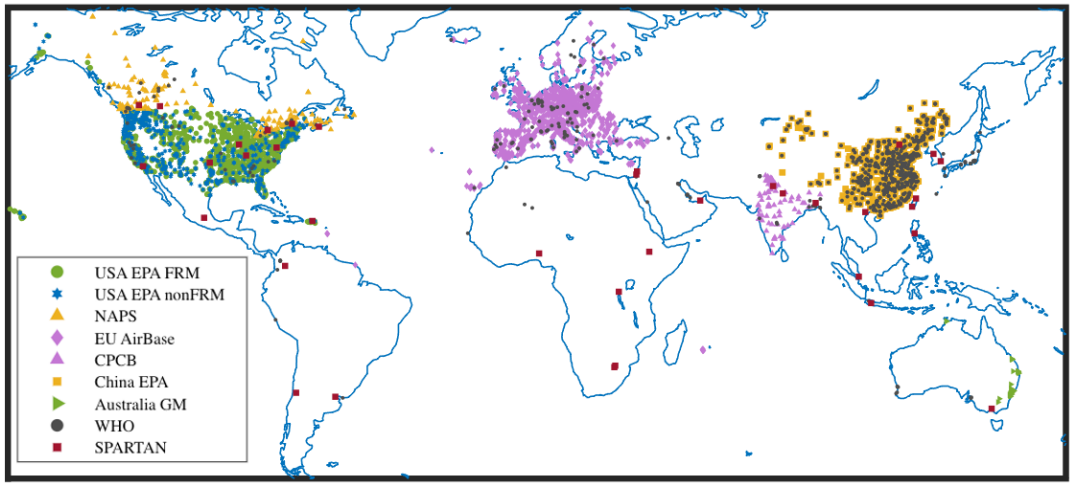

Figure A1. $PM_{2.5}$ measurement sites from publicly available networks.

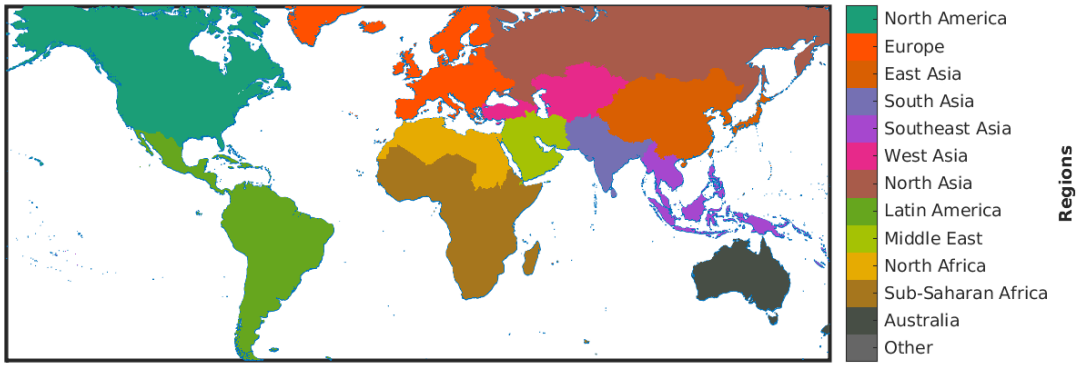

Figure A2. Region definition.

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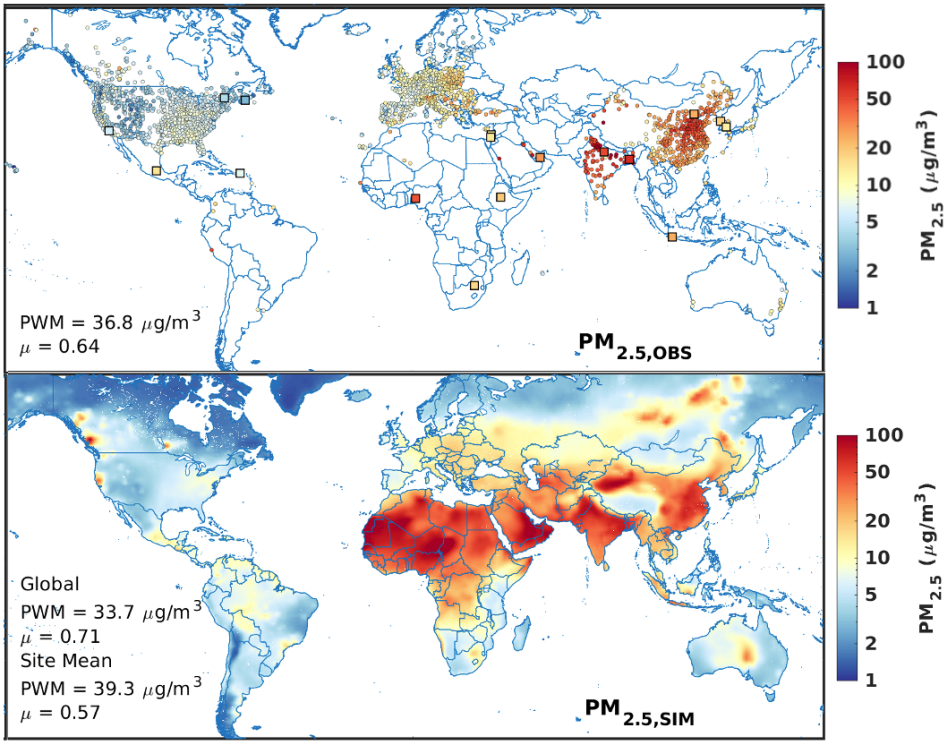

Figure A3. Observed (top) and simulated (bottom) annual mean $PM_{2.5}$ for 2019. Circles represent measurement sites from regional networks or reported by the WHO. Squares represent measured $PM_{2.5}$ from SPARTAN. PWM = population-weighted mean, μ = coefficient of variation.

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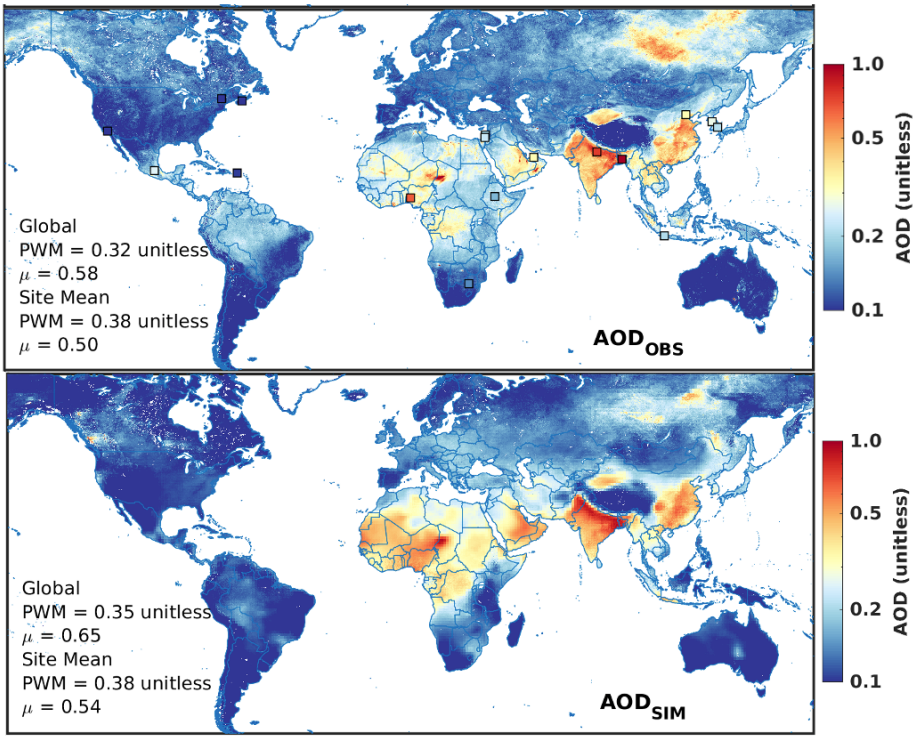

Figure A4. Satellite retrieved (top) and GCHP simulated (bottom) annual mean AOD for 2019. Squares represent ground-measured AOD from AERONET. PWM = population-weighted mean, μ = coefficient of variation.

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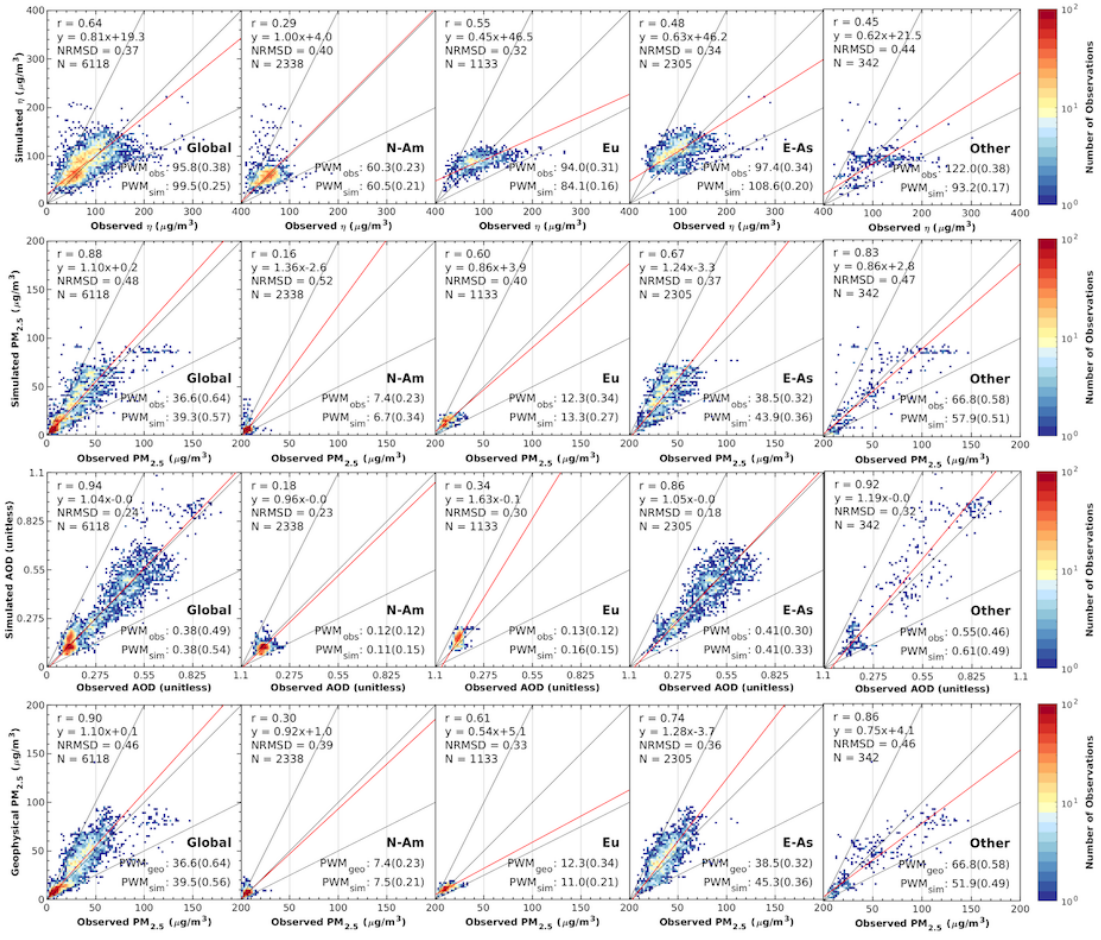

Figure A5. Scatter plots of simulated and observed η (top row), simulated and ground measured $PM_{2.5}$ (second row), simulated and MAIAC AOD (third row), and geophysical and observed $PM_{2.5}$ (bottom row). The red line shows the line of best fit using Reduced Major Axis Linear Regression. Insets on the top left show the coefficient of determination (R^2), line of best fit, normalized root mean square deviation (NRMSD), and total number of data points (N). The bottom right insets show the population-weighted mean of observed, simulated, or geophysical estimation of each dataset, coefficients of variation are bracketed. Detailed regional mean and coefficients of variation for other regions can be found in Table A1.

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Table A1. Regional population-weighted mean η, $PM_{2.5}$, and AOD from both observation and simulations. Geophysical $PM_{2.5}$ is also included. Coefficients of variation are bracketed. Regional mean and coefficients of variation for North America, Europe, and East Asia can be found in Figure A5.

Region		South Asia	Southeast Asia	West Asia	Latin America	Middle East	North Africa	Sub-Sahara Africa	Australia
Number of sites		162	3	43	2	46	29	3	5
η [μg/m^3]	Observed	121.6 (0.37)	128.6 (0.12)	154.0 (0.23)	72.0 (0.29)	94.1 (0.56)	132.3 (0.35)	196.0 (0.01)	133.9 (0.34)
	Simulated	93.4 (0.10)	82.4 (0.09)	93.4 (0.03)	74.1 (0.04)	83.6 (0.21)	126.6 (0.17)	105.9 (0.01)	187.3 (0.26)
$PM_{2.5}$ [μg/m^3]	Observed	81.0 (0.41)	35.7 (0.44)	22.0 (0.21)	12.0 (0.23)	21.7 (0.51)	28.7 (0.61)	24.0 (0.00)	35.5 (0.29)
	Simulated	70.2 (0.30)	31.8 (0.20)	20.8 (0.08)	20.9 (0.06)	10.2 (0.25)	38.3 (0.53)	16.7 (0.03)	90.0 (0.31)
	Geo-physical	62.7 (0.30)	22.7 (0.29)	13.9 (0.08)	12.4 (0.08)	20.4 (0.37)	27.3 (0.49)	12.9 (0.03)	58.1 (0.74)
AOD [unitless]	Observed	0.67 (0.25)	0.27 (0.35)	0.14 (0.08)	0.17 (0.03)	0.24 (0.21)	0.21 (0.28)	0.12 (0.01)	0.30 (0.66)
	Simulated	0.73 (0.28)	0.38 (0.18)	0.22 (0.09)	0.28 (0.02)	0.12 (0.14)	0.29 (0.32)	0.16 (0.01)	0.51 (0.26)

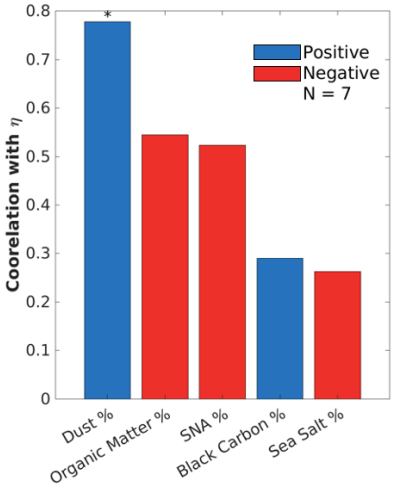

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Figure A6. Correlation with η of ground-measured aerosol fractional composition from SPARTAN. Organic matter is inferred through residual (Snider et al., 2016). Blue bars indicate positive correlations. Red bars indicate negative correlations. Stars above each bar indicate the p-value associated with each correlation. '***' means the p-value is lower than 0.001, '**' means lower than 0.01, and '*' means lower than 0.5.

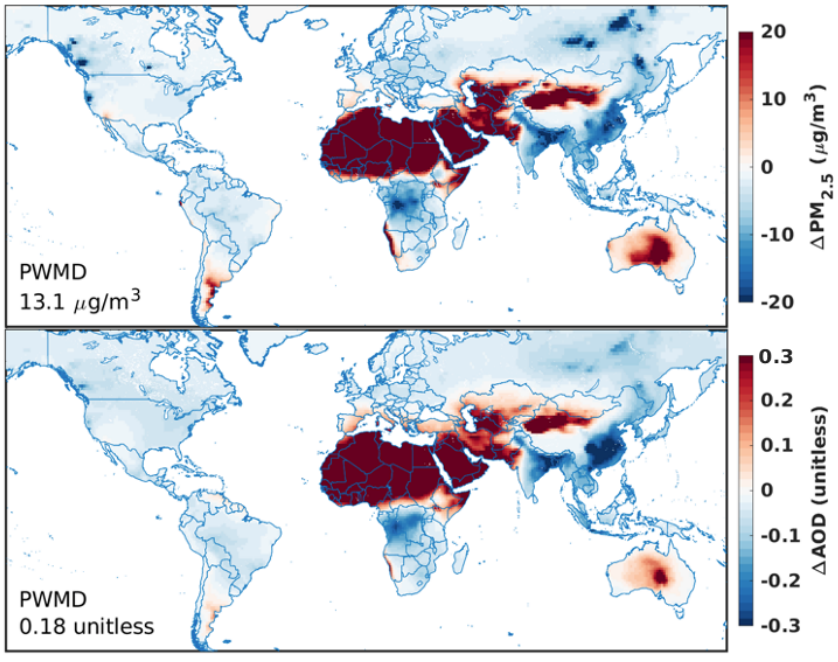

Figure A7. Changes in $PM_{2.5}$ (top) and AOD (bottom) (test - base) when imposing a global PWM aerosol composition.

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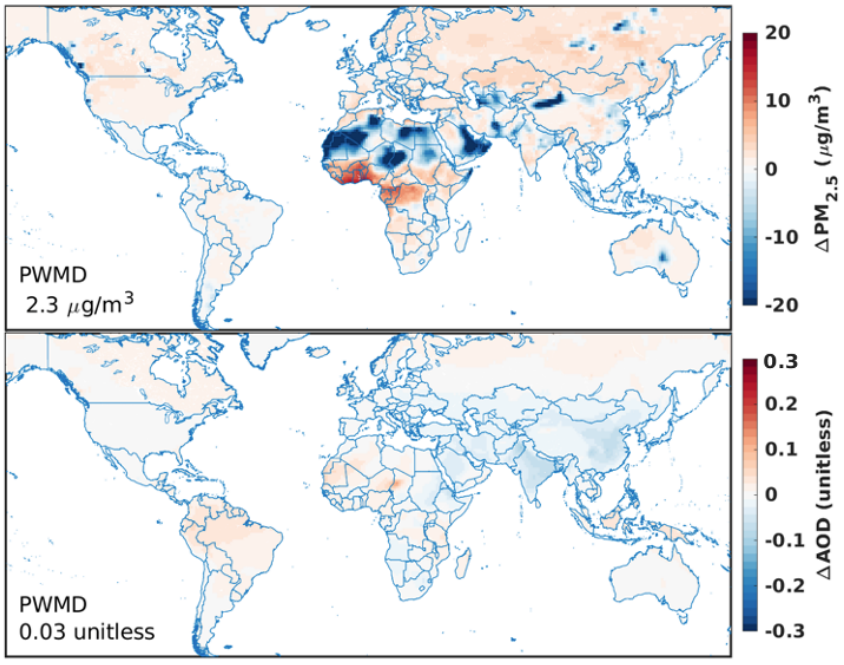

Figure A8. Changes in $PM_{2.5}$ (top) and AOD (bottom) (test - base) when imposing a global PWM aerosol profile.

Data availability. GEOS-Chem in its high-performance configuration version 13.4.0 can be downloaded at https://zenodo.org/records/6564711.

Author contributions. HZ and RVM designed the study. HZ performed the data analysis and model simulation with contributions from AvD, CL, YL, DZ, JM, MH & IS. AvD contributed to the compiled the MAIAC AOD dataset and ground-based observation datasets for $PM_{2.5}$. AL contributed to the original MAIAC AOD dataset. CRO and XL contributed to the SPARTAN data utilization and analysis. The manuscript was written by HZ and RVM with contributions from all authors.

Competing interests. The authors declare no competing financial interest.

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