Ambient Particulate Matter Size Distributions Drive Regional and Global Variability in Particle Deposition in the Respiratory Tract

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Abstract

Human exposure to airborne particulate matter (PM) increases the risk of negative health outcomes; however, substantial uncertainty remains in quantifying these exposure-response relationships. In particular, relating increased risk of mortality to exposure to PM with diameters smaller than 2.5 μm (PM$_{2.5}$) neglects variability in the underlying size distribution of PM$_{2.5}$ exposure and size-resolved deposition in human airways. In this study, we combine a size-resolved respiratory particle-deposition model with a global size-resolved aerosol model to estimate the variability in particle deposition along the respiratory tract due to variability in ambient PM size distributions. We find that the ratio of deposited PM mass in the tracheobronchial and alveolar regions per unit ambient PM$_{2.5}$ exposure (deposition ratio and DR$_{TB + AV}$) varies by 20–30% between populated regions due to variability in ambient PM size distributions. Furthermore, DR$_{TB + AV}$ can vary by as high as a factor of 4 between the fossil-fuel-dominated region of the Eastern United States and the desert-dust-dominated region of North Africa. When considering individual PM species, such as sulfate or organic matter, we still find variability in the DR$_{TB + AV}$ on the order of 30% due to regional variability in the size distribution. Finally, the spatial distribution of DR$_{TB + AV}$ based on number or surface area is substantially different than the DR$_{TB + AV}$ based on mass. These results suggest that regional variability in ambient aerosol size distributions drive variability in PM deposition in the body, which may lead to variability in the health response from exposure to PM$_{2.5}$.

1. Introduction

Exposure to particulate matter (PM) air pollution increases the risk of cardiovascular and respiratory disease. In particular, long-term exposure to the mass of PM with aerodynamic diameters less than 2.5 μm (PM$_{2.5}$) has been shown to have a causal association with an increased risk of mortality from cardiovascular and respiratory diseases (Dockery et al., 1993; Krewski et al., 2009; Pope et al., 2002; Pope & Dockery, 2006). Over 4 million deaths were estimated to be attributed to chronic exposure to ambient PM$_{2.5}$ pollution in 2015, making it one of the leading risk factors contributing to premature mortality (Cohen et al., 2017). In addition, a number of studies have shown correlations between short-term exposure to PM$_{2.5}$ and negative health consequences (Gan et al., 2017; Ostro et al., 2006; Pope & Dockery, 2006). Despite numerous associations between PM$_{2.5}$ exposure and increased risks of negative health effects, substantial uncertainty remains in quantifying the magnitude and shape of these exposure-response relationships.

As the exact mechanisms that relate exposure to PM$_{2.5}$ to increased risk of negative health outcomes (such as premature mortality) are unclear, health impact assessments (such as the widely cited Global Burden of Disease Study) rely on integrated exposure-response functions to estimate population-level health impacts for a given level of PM$_{2.5}$ exposure (e.g., Burnett et al., 2014). These integrated exposure-response functions are synthesized from available epidemiologic data. The 2.5-μm diameter cutoff is frequently used as a proxy for airborne PM that, upon inhalation, can penetrate to and deposit within the lower respiratory region of the lung. In addition, as ambient particle mass size distributions (when weighted according to PM mass) tend to feature a local minimum around 2.5 μm in diameter, this size cutoff separates fine particles (formed from combustion processes and secondary aerosol formation) and coarse particles (emitted mostly from mechanical emissions; e.g., dust and sea spray; Seinfeld & Pandis, 2012). Long-term ambient PM$_{2.5}$ measurement networks have existed in recent decades in the United States, Europe, and Canada. As a result, most of the epidemiology studies that relate exposure to ambient PM$_{2.5}$ are based in the North America and...
models to estimate the geographic variability in deposited particle mass. We use this to motivate the idea that particle deposition along the respiratory tract exhibits a strong size and shape dependence (Hussein et al., 2013; Löndahl et al., 2007, 2009), and thus, the extent of particle deposition may depend on the inhaled particle size distribution. Mechanisms of particle deposition in specific regions of the respiratory tract (e.g., head/nose, tracheobronchial, or alveolar) are complex. The fraction of particles deposited to different locations in the airways depends on particle size, shape, and density as well as airway geometry and breathing pattern (affecting flow rate and residence time). Briefly, Brownian diffusion, inertial impaction, interception, and gravitational settling are the dominant processes governing particle deposition in the respiratory tract (Hinds, 1999). Ultrafine particles (with diameters smaller than approximately 100 nm) deposit primarily through Brownian diffusion, lacking enough inertia to deposit through impaction or gravitational settling. Very few studies used in generating health-response functions have been done in low- and middle-income countries in Asia, sub-Saharan Africa, or South America, yet these regions bear the majority of the health burden due to air pollution exposure (Cohen et al., 2017).

The GBD 2015 study acknowledges that applying these exposure-response functions generated in limited regions to all regions of the world is a major source of uncertainty (Burnett et al., 2014; Cohen et al., 2017). Possible sources of variability in published exposure-response relationships include demographic factors (e.g., a population in a certain region is more or less susceptible to exposure) or differential toxicity of different chemical components present in PM$_{2.5}$ (such as the sulfate or organic aerosol [OA] mass fraction; e.g., Bates et al., 2015; Bell et al., 2007; Fang et al., 2017; Verma et al., 2015). Variability in the chemical composition of ambient PM$_{2.5}$ may reflect differing contributions from local emission sources, transport processes, or chemical and physical processes in the atmosphere. In addition to these sources of variability, this work suggests that variability in ambient PM size distributions may also lead to variability in health responses following PM$_{2.5}$ exposure.

Ambient particle size distributions exhibit strong regional variability, reflecting local/regional differences in emission sources, transport, and ambient chemical/physical processes (e.g., Jaenicke, 1993). Different emission sources may emit particles at different sizes. For instance, combustion processes tend to emit particle mass in the 100- to 300-nm diameter range (Ban-Weiss et al., 2010; Janhäll et al., 2010; Sakamoto et al., 2015; Winijkul et al., 2015), while dust (fugitive and windblown) and sea-spray emissions tend emit aerosol mass in ranges above 1 μm (Jaeglé et al., 2011; Kok, 2011; Mahowald et al., 2014). This results in urban regions having a strong contribution of accumulation-mode mass to PM$_{2.5}$ concentration, and desert regions having a substantial contribution to coarse-mode mass. In addition to local emissions, observations show variability in particle size distributions and chemical composition due to transport of PM (e.g., Dunlea et al., 2009; Xu et al., 2017). Finally, observations and modeling studies show that microphysical processes such as new-particle formation events (e.g., Hodshire et al., 2016; Merikanto et al., 2009; Westervelt et al., 2014), condensation to and evaporation of particles (e.g., Bian et al., 2017; Bougiatioti et al., 2014; D’Andrea et al., 2013), in-cloud aqueous oxidation (e.g., Hoppel & Frick, 1990), coagulation (e.g., Sakamoto et al., 2016; Westphal & Toon, 1991), wet deposition (e.g., Croft et al., 2012; Dentener et al., 2006), and dry deposition (e.g., Sehmel, 1980; Zhang et al., 2001) substantially influence particle number and mass distributions (Croft et al., 2016; Kodros & Pierce, 2017).

Particle deposition along the respiratory tract exhibits a strong size and shape dependence (Hussein et al., 2013; Löndahl et al., 2007, 2009), and thus, the extent of particle deposition may depend on the inhaled particle size distribution. Mechanisms of particle deposition in specific regions of the respiratory tract (e.g., head/nose, tracheobronchial, or alveolar) are complex. The fraction of particles deposited to different locations in the airways depends on particle size, shape, and density as well as airway geometry and breathing pattern (affecting flow rate and residence time). Briefly, Brownian diffusion, inertial impaction, interception, and gravitational settling are the dominant processes governing particle deposition in the respiratory tract (Hinds, 1999). Ultrafine particles (with diameters smaller than approximately 100 nm) deposit primarily through Brownian diffusion, lacking enough inertia to deposit through impaction or gravitational settling at relevant time scales. Coarse-mode particles (greater than 2.5 μm) primarily deposit through inertial impaction or gravitational settling. There exists a minimum in deposition efficiency at diameters of approximately 200–700 nm, where particles neither diffuse nor impact/settle with sufficient magnitude to deposit efficiently. Thus, the mechanics governing particle deposition have many size-dependent features that are not accurately represented by a simple 2.5-μm cutoff measurement. Knowledge of the total particle size distribution is necessary to translate an exposure measurement into an estimate of PM mass deposited along the respiratory tract. A number of dosimetry models have been developed to estimate the fraction of particles that deposit in specific regions in the body. Such models include the empirical model of the International Commission on Radiological Protection (ICRP; ICRP, 1994) and the Multiple Path Particle Dosimetry (MPPD) model (Miller et al., 2016).

In this study, we combine simulated ambient aerosol size distributions with size-resolved particle deposition models to estimate the geographic variability in deposited particle mass. We use this to motivate the idea...
that variability in PM$_{2.5}$ exposure response may be partly explained by variability in ambient particle size distributions affecting deposition mass of PM in the body. In section 2, we discuss our methods of using particle deposition models and a chemical-transport model with online aerosol microphysics. In sections 3.1–3.2, we present simulations of ambient PM$_{2.5}$ mass concentration along with deposited PM mass in the body. In section 3.3, we explore geographic variability in species-specific PM size distribution and deposition. In section 3.4, we present geographic variability in particle number and surface-area deposition. We share our conclusions in section 4.

2. Methods

2.1. Modeling Size-Resolved Particle Deposition in the Body

To estimate the mass of PM that deposits at region-specific sites in the body, we use the ICRP particle deposition model (ICRP, 1994). We use equations fitted to measured deposition fractions as a function of particle diameter from the ICRP model averaged for various breathing conditions by Hinds (1999). Particle deposition fractions are shown in Figure 1 for all regions in the body, including the head/nose region, tracheobronchial region, alveolar region, and the sum of the tracheobronchial and alveolar regions. In later sections, we focus on the sum of deposition to the tracheobronchial and alveolar regions. Newer particle deposition models generally agree with the results of the ICRP model. To test the sensitivity to deposition model, we also consider regional deposition curves from the MPPD model (Miller et al., 2016). Deposition fractions from the MPPD model to the ICRP model are compared Figures S1–S3 in the supporting information.

We account for swelling of airborne particles due to water uptake in the lung assuming that air in the lung and associated airways has a relative humidity of 99.5% and a temperature of 37 °C (Anselm et al., 1990). The supporting information contains a detailed description of our treatment of aerosol water uptake in the lung (Hussein et al., 2013; Löndahl et al., 2007, 2009; Petters & Kreidenweis, 2007). Briefly, we approximate growth to equilibrium sizes based on the total hygroscopicity of the particle in a given size range (assuming all particles in the size range are internally mixed). As deposition results are dependent on this assumption, we test the sensitivity of this treatment in Figure S4 by assuming no particle growth in the lung and growth based on lung relative humidities of 98% and 99.5%.

2.2. Simulating Ambient Aerosol Size Distributions

To simulate the transport of gas- and particle-phase species, we use the chemical-transport model, GEOS-Chem (Bey et al., 2001), version 10.01. GEOS-Chem is driven by reanalysis meteorology fields by GEOS-FP. We run simulations of GEOS-Chem with 47 vertical levels and horizontal resolutions of 4 × 5° and 2 × 2.5° for the full global domain, and we perform nested simulations at 0.5 × 0.666° over North America and eastern Asia. The standard setup of GEOS-Chem includes tracers for 52 gas-phase species. All simulations are for one full year (year 2010) with an additional 1-month spin-up not included in analysis to initiate concentrations of chemical species.

To simulate size-resolved aerosol mass and number concentrations, GEOS-Chem is coupled online to the Two Moment Aerosol Sectional (TOMAS) microphysical model (Adams & Seinfeld, 2002). The TOMAS model includes tracers for size-resolved particle number along with size-resolved mass for sulfate (SO$_4$), sea salt, OA, black carbon (BC), and dust. We use versions of TOMAS with 15- and 40-bin size sections both simulating dry diameters from approximately 1 nm to 10 µm (Lee & Adams, 2012; Lee et al., 2013). Due to computational limitations, we use the 40-bin TOMAS only with the GEOS-Chem 4 × 5° horizontal resolution and the 15-bin TOMAS with the GEOS-Chem 2 × 2.5° and 0.5 × 0.666° horizontal resolutions. We use the 40-bin version of TOMAS at the coarse spatial resolution to inform an interpolation of aerosol size distributions with the 15-bin version at finer spatial resolution. This is done to optimize computational expense. All results shown here use interpolated 40-bin aerosol size distributions at 2 × 2.5° or 0.5 × 0.666° resolution.

![Deposition curves based on the International Commission on Radiological Protection model with fitted equations from Hinds (1999) for total deposition in the body as well as regional deposition in the head/nose, tracheobronchial, alveolar, and sum of the tracheobronchial and alveolar region. In later sections, we focus on the sum of deposition in the tracheobronchial and alveolar region.](image-url)
Detailed descriptions of microphysical processes in TOMAS have been described elsewhere (Adams & Seinfeld, 2002; Lee & Adams, 2012 and Lee et al., 2013). TOMAS explicitly accounts for size-resolved aerosol emissions, coagulation, condensation, wet and dry deposition, aqueous oxidation, and new-particle formation. Emissions of primary SO₄ include two lognormal size modes: The first mode contains 15% of the mass with a mass median diameter (MMD) of 19 nm and geometric standard deviation (GSD) of 1.6 and the remainder in a second mode with a MMD of 296 nm and GSD of 2.0 (Adams & Seinfeld, 2003). BC and OA emissions are separated by emission source. Fossil fuel OA and BC are emitted into the model with an MMD of 126 nm and GSD of 2.0, while biofuel and biomass burning particles are emitted with an MMD of 422 nm and a GSD of 2.0 (Pierce et al., 2007). Dust emissions follow the DEAD scheme (Zender, 2003), while sea salt emissions are based on the scheme of Jaeglé et al. (2011). The TOMAS model includes a ternary nucleation scheme involving water, sulfuric acid, and ammonia following the parameterization of Napari et al. (2002), with nucleation rates scaled down by 5 orders of magnitude to better match observations (following Westervelt et al., 2013). When ammonia mixing ratios are less than 1 parts per thousand by volume, TOMAS defaults to a binary nucleation scheme (sulfuric acid and water; Vehkamäki, 2002). In addition to nucleation, TOMAS explicitly simulates condensation, coagulation, wet and dry deposition, and cloud processing.

Simulations using GEOS-Chem-TOMAS have been evaluated against observations of long-term, size-resolved number concentrations (Croft et al., 2016; D’Andrea et al., 2013; Kodros & Pierce, 2017; Westervelt et al., 2013) and aerosol optical depth (Kodros et al., 2016). While GEOS-Chem-TOMAS has shown skill at reproducing variability in size-resolved aerosol concentrations, we acknowledge that measurements of aerosol size distributions (and thus model evaluations) are limited. As such, we are unable to fully determine the accuracy of the simulated aerosol size distributions in GEOS-Chem-TOMAS; however, as we know particle size distributions exhibit regional variability (e.g., Jaenicke, 1993), TOMAS provides a reasonable option to explore how diversity in size distributions may affect respiratory PM deposition.

3. Results

3.1. Simulated Annual PM₂.₅ Concentration

In this study, we define PM₂.₅ as the dry particle mass concentration contained in particles with wet aerodynamic diameters smaller than 2.5 μm. We use this definition so that our simulated PM₂.₅ is directly comparable to PM₂.₅ networks that use an inertial cyclone to size-select particles with a wet aerodynamic diameter smaller than 2.5 μm, where these particles are then dried at ~35% RH, which removes most of the aerosol water. The annual-average PM₂.₅ mass simulated with GEOS-Chem-TOMAS is plotted in Figure 2a. There is substantial geographic variability in PM₂.₅ concentrations, ranging from 10–20 μg m⁻³ in the Eastern United States to 80–100 μg m⁻³ in Eastern China. Regions with the highest concentration of PM₂.₅ include the Sahara, Eastern China, and Northern India. In addition to the evaluations of simulated GEOS-Chem-TOMAS size distributions mentioned in the previous section, evaluation of simulated PM₂.₅ by GEOS-Chem has been done in several previous studies (Park et al., 2006; van Donkelaar et al., 2010), and so we do not include this here.

In addition to geographic variability in total PM₂.₅ mass concentration, there is also substantial variability in the underlying mass distributions and chemical composition by size. In Figures 2b–2e, we plot dry mass distributions at wet aerodynamic diameters showing particle composition for a grid cell in the Eastern United States, Algeria, Indonesia, and Northern India. These four representative regions were chosen to demonstrate the variability in size distribution and composition. We refer to these regions throughout this section. The mass distributions in the Eastern United States, Northern India, and Indonesia have a substantial contribution of accumulation-mode particles to PM₂.₅ mass. The accumulation-mode particles are primarily composed of OA (primary and secondary) along with SO₄ and trace amounts of BC. The Eastern United States and India also have a contribution from coarse-mode particles to PM₂.₅ mass concentration; however, much of the coarse-mode mass is larger than a wet ambient diameter of 2.5 μm and hence would not be captured by a PM₂.₅ monitor. Conversely, the PM₂.₅ mass concentration in Algeria is composed almost entirely of dust even though the PM₂.₅ dust is only a small fraction of the total dust in this location.

Differences in regional emissions sources explain much of the differences in particle size and composition, while chemical and physical processing (coagulation, condensation, and cloud processing) in the
atmosphere (discussed later) explains the rest. PM emissions in the Eastern United States are largely derived from fossil-fuel combustion (e.g., road transportation, industry, and power generation), resulting in PM composed mainly of SO4, OA, and BC. In GEOS-Chem-TOMAS, OA and BC from fossil fuel are emitted with smaller MMDs (126 nm) than OA and BC from biomass burning emissions (MMD of 422 nm). The large contribution from fossil-fuel emissions in the Eastern United States (such as from road transport) results in an accumulation-mode MMD between 250 and 300 nm, indicating particle growth from emission of MMD of approximately 130 nm likely through coagulation and condensation of sulfuric acid and organics. India and Indonesia have substantial PM emissions from biomass combustion (including residential, agricultural, and wildfire). These biomass emissions result in a relatively larger OA contribution to accumulation-mode PM mass (MMD between 400 and 500 nm) than in the Eastern United States. Finally, local dust emissions in North Africa are predominantly emitted into the coarse mode resulting in an MMD of 6 μm in Algeria.

3.2. Respiratory Deposited PM Mass and Deposition Ratio

To estimate geographic variability in the respiratory deposition of PM mass, we multiply the size-dependent ICRP deposition fraction to the simulated aerosol mass distributions from GEOS-Chem-TOMAS. This product results in deposited PM mass per volume of inhaled air (units of μg PM deposited in lung, written here as μg_{dep} per m^3 of inhaled air, written here as m_{inh}^{-3}). In Figure 3a, we show the geographic distribution of total deposited PM mass in the body. Similar to the spatial distribution of PM_{2.5} mass concentrations, the desert regions (the Sahara, Middle East, and Gobi deserts) have the highest deposited PM mass concentration (larger than 50–100 μg_{dep} m_{inh}^{-3}). India and Eastern China also have high concentrations of deposited PM mass (35–50 μg_{dep} m_{inh}^{-3}). In addition to geographic variability, there is also variability in magnitude of deposition mass in each region of the body. We show maps of deposition in the (b) head/nose region, (c) tracheobronchial region, and (d) alveolar region in Figure 3. Most of the inhaled PM mass deposits in the head/nose region; however, concentrations ranging from 0.5 to 30 μg_{dep} m_{inh}^{-3} deposit in the tracheobronchial and alveolar regions.

Much of the geographic variability in deposited PM mass in Figure 3 arises from the magnitude of ambient PM mass. To remove this influence and understand the role of ambient particle size distributions in determining PM deposition in the tracheobronchial and alveolar regions (TB + AV), we normalize TB + AV deposited PM mass concentration (PM_{TB + AV}) by the ambient PM_{2.5} concentration. We define the “deposition ratio” in the tracheobronchial and alveolar regions (DR_{TB + AV}) as the ratio of PM_{TB + AV} per unit ambient PM_{2.5}.
mass concentration. \( \text{DRTB + AV} \) is thus the ratio of the total deposited mass concentration across all size bins to the total ambient PM\(_{2.5}\) mass concentration. This ratio is meant to demonstrate the variability of PM mass that deposits compared to what would be measured as ambient PM\(_{2.5}\). Here we focus on the lower-respiratory regions (the tracheobronchial and alveolar regions), as cardiovascular and respiratory diseases may be most sensitive to particle deposition in these respiratory regions; however, this is a simplifying assumption and the exact mechanisms relating particle deposition to effective dose are unclear. \( \text{DRTB + AV} \) is plotted in Figure 4a (central map). Across different highly populated regions, the \( \text{DRTB + AV} \) tends to vary by 20–30% (such as between Eastern United States and India); however, globally, this variability can be as high as a factor of 4 when considering desert or oceanic regions where aerosol size distributions can vary considerably (relative to urban regions). Regions with the highest \( \text{DRTB + AV} \) are desert regions (e.g., the Sahara, Middle East, Gobi, and Southwest US), while continental regions with the lowest \( \text{DRTB + AV} \) include fossil-fuel-dominated regions of the Eastern United States and Western Europe (and even lower in the Southern Ocean).

The cause of the variability in the \( \text{DRTB + AV} \) comes from the underlying PM mass size distribution. In Figures 4b–4e, we show the mass distribution at ambient conditions (light blue line), the mass distribution accounting for hygroscopic growth due to the RH of the lung (black solid line), the deposited mass concentration for each size (black dashed line), and the ICRP fractional deposition curve (red line, left axis) for comparison. In the Eastern United States (Figure 4b), the ambient mass distribution has a large contribution from accumulation-mode particles with MMD between 250 and 300 nm. These particles are, to some degree, hygroscopic and grow in the high RH of the lung to a median diameter of 1 \( \mu m \). This growth increases PM deposition as it moves more particles out of the deposition minimum (centered at 250 nm). The Eastern United States has some coarse-mode mass at sizes larger than 2.5 \( \mu m \), which contributes to PM\(_{10}\) but not the PM\(_{2.5}\) concentration. The net result is a \( \text{DRTB + AV} \) of 0.14. Conversely, in Algeria, most of the mass at ambient conditions is coarse-mode dust (see Figure 2). Dust particles are modeled here as hydrophobic (a kappa value of 0.01; see Supporting Information), resulting in less growth in the lung. Even though most coarse-mode mass is deposited in the head/nose region and not counted here (Figures 1 and 3), the ICRP model still estimates deposition fractions in tracheobronchial and alveolar region of 10% at 2.5 \( \mu m \) decreasing to near 0% at 10 \( \mu m \). Much of this (TB + AV)-deposited mass is not included in the PM\(_{2.5}\) cutoff. The net result is a \( \text{DRTB + AV} \) of 0.55. This higher value of \( \text{DRTB + AV} \) is largely a result of the contribution of coarse-mode

![Figure 3. Deposited PM mass (integrating from 0.001 to 10 \( \mu m \)) in (a) all regions, (b) head/nose region, (c) tracheobronchial region, and (d) alveolar region.](image-url)
particle mass to PM_{TB + AV} (the numerator of DR_{TB + AV}) that is not included in the ambient PM_{2.5} concentration (the denominator of DR_{TB + AV}). The ambient mass distribution in Indonesia is representative of a region with substantial emissions contribution from biomass burning. The ambient mass concentration is dominated by accumulation-mode particles with a MMD between 400 and 500 nm. Similar to the Eastern United States, these particles are partly hygroscopic and grow in the lung to a MMD between 1 and 1.5 μm.

The ambient mass distribution in Northern India contains both an accumulation mode (likely from residential solid fuel use and fossil-fuel combustion) as well as a coarse-mode mass from dust. The accumulation-mode particles are partly hygroscopic and grow to larger sizes, while the coarse mode is hydrophobic with slower condensational growth time scales. The larger contribution of coarse-mode particles to deposition mass results in a higher DR_{TB + AV} in India (0.18) than Indonesia (0.15).

To further investigate the geographic variability of TB + AV particle deposition at continental scales, we include nested simulations at finer horizontal resolutions of 0.5 × 0.666° over a domain centered on North America and a domain centered on Asia. In Figure 5, we show predicted DR_{TB + AV} for the North American and Asian domains. In the finer-resolution North America domain the variability DR_{TB + AV} increases to 50–60%, ranging from a maximum in the desert Southwest United States to a minimum in areas more influenced by fossil-fuel combustion and secondary aerosol formation. The higher-resolution simulation for North America shows increased spatial variability compared to the global simulation with deposition minima in the populated areas of California, Pacific Northwest, Salt Lake City, Denver, Calgary, and Mexico City. The primary reason for the lower DR_{TB + AV} in more populated areas relative rural areas is the higher hydrophobic particle fraction (mainly hydrophobic OA) emitted in urban areas. Downwind of urban areas, hydrophobic OA (and externally BC) undergoes atmospheric processing and converts to hydrophilic OA (and internally mixed BC). Meanwhile, sulfur dioxide is oxidized and may form sulfate aerosol on a time scale of ~2 days. The net result is increased SO_{4} and hydrophilic OA outside of urban areas (similar to results in Jimenez et al., 2009; Philip et al., 2014). The more hydrophilic particles are able to grow to larger diameters in the high RH of the lung, thus moving further out of the minimum in particle deposition (centered at 250 nm, see Figure 1). In the Asian domain, there exists a strong gradient in DR_{TB + AV} from the Gobi desert region to the more urban areas of Eastern China and Northern India. Conversely to the North American domain, the spatial gradient of DR_{TB + AV} in Asia seems less influenced by urban areas. This is likely a result of high aerosol and precursor concentrations in this region leading larger accumulation mode diameters through condensation and coagulation. These larger-sized particles have higher deposition efficiencies in the
TB + AV (Figure 1). While these nested simulations have a finer spatial resolution than the global simulations, they are still too coarse to resolve near-source gradients in particle concentrations and size distributions. Thus, the variability in aerosol size distributions and TB + AV particle deposition may be greater than what is shown here.

The spatial distribution of DRTB + AV is sensitive to our assumptions regarding particle growth in the lung. In Figure S4, we show DRTB + AV with no particle growth, particle growth based on an RH of 98%, and particle growth based on an RH of 99.5%. The spatial variability between urban regions and desert regions of DRTB + AV decreases with increasing particle growth in the lung. This is due to accumulation-mode particles growing out of the minimum in the deposition fraction of the ICRP model (Figure 1), thus depositing more efficiently, while coarse-mode particles grow to sizes that deposit less efficiently. Assuming particle growth based on a lung RH of 99.5% may thus provide a lower limit on the spatial variability of respiratory particle deposition.

3.3. Speciated Size-Dependent Deposition

A current focus of much recent research is the possibility that regional variability in the health response to PM2.5 exposure may be partly influenced by differential toxicity of different PM species. To some degree, differences in size distributions reflect changes in composition of PM. Figure 6 shows speciated distributions of PM_{TB + AV} (units of μg_{dep} m^{-3}). These plots are analogous to the ambient mass distributions in Figure 2; however, they now show the TB + AV deposited particle mass concentrations at each size interval. Across the four representative regions shown here, there is variability not just in PM_{TB + AV}, but in the composition of particles that deposit. In Eastern United States and India, much of the PM_{TB + AV} is OA, SO4, and dust with trace contributions from BC. The relative contribution of dust to PM_{TB + AV} increases relative the contribution of dust to ambient PM mass. Conversely, the PM_{TB + AV} in Algeria is almost entirely dust, reflecting the dominant particle emission source in this region. Due to the large contribution of biomass burning to the local PM burden in Indonesia, accumulation-mode OA makes up much of the PM_{TB + AV} in this region. The variability in composition of PM_{TB + AV} further motivates understanding the differential toxicity of different PM species.

To some degree, variability in size distributions and variability in particle composition are connected. Much of the accumulation-mode mass is made up of OA, SO4, and BC, while the coarse-mode mass is made up of dust and sea salt. To test if variability in the respiratory deposition of PM mass could be resolved by focusing only on variability in chemical species, Figure 7 shows the DR for specific chemical species of PM. In Figures 7a and 7c, we show deposited PM mass of SO4 per unit ambient PM_{2.5} mass of SO4 in all body regions (DR_{total}) and DRTB + AV, respectively. In Figures 7b and 7d, we show the same ratios for OA. When focusing on a specific PM species, there still exists potentially significant geographic variability in DR_{total}. For deposition in any region of
the body, the DRtotal of SO4 deposited mass to ambient SO4 PM2.5 mass varies by 40–50%, ranging from minima in South America, sub-Saharan Africa, and Western Europe to maxima in the desert Sahara and Middle East. The maxima in desert regions are likely caused by mixing of SO4 mass into the coarse mode via coagulation with and condensation onto dust particles. The DRtotal for OA has a similar range of variability.

Figure 6. Speciated distribution of PMT + AV at each size interval for the region representing the (a) Eastern United States, (b) Algeria, (c) Indonesia, and (d) India.

Figure 7. Geographic variability of (a and c) sulfate DRtotal and DRT + AV and (b and d) organic aerosol DRtotal and DRT + AV per unit ambient sulfate and organic aerosol PM2.5 mass, respectively.
but with a different geographic distribution. The OA $\text{DR}_{\text{total}}$ has a maximum in South America and all of Africa and the Middle East with minima in North America and Europe. The maxima in South America and central Africa are likely caused by the larger emission MMD of biomass burning particles, as well as condensation of secondary OA onto a relatively low number concentration (relative the Eastern United States) allowing for particle growth to larger diameters with higher TB + AV deposition efficiency. Similar to SO$_4$, the maxima in the desert regions of North Africa and the Middle East are likely caused by coagulation with coarse-mode dust particles. The minima in $\text{DR}_{\text{total}}$ in North America and Europe are due to primary OA emitted at diameters similar to the minimum in the ICRP deposition fraction curve (Figure 1). The $\text{DR}_{\text{TB + AV}}$ for SO$_4$ and OA varies by up to 25%, similar to when considering $\text{DR}_{\text{TB + AV}}$ from all PM species. The $\text{DR}_{\text{TB + AV}}$ for SO$_4$ and OA differ in South America and sub-Saharan Africa, showing minima for SO$_4$ but maxima for OA. Conversely, both show lower $\text{DR}_{\text{TB + AV}}$ for the United States and Europe relative India and China. These results suggest that even if the health effects of PM can be attributed to specific PM species, we may still need to consider variability in the size distributions of these species.

We show the $\text{DR}_{\text{TB + AV}}$ for SO$_4$ and OA for the finer-resolution simulations in the North American and Asian domains in Figure 8. In North America, the spatial variability in $\text{DR}_{\text{TB + AV}}$ for both SO$_4$ and OA follows a strong urban versus rural split. The minima in $\text{DR}_{\text{TB + AV}}$ are limited to urban areas such as Minneapolis, Denver, Houston, coastal California, Calgary, and Mexico City. Similar to $\text{DR}_{\text{TB + AV}}$ based on all-species PM respiratory deposition, this occurs due to a larger hydrophobic particle fraction in urban areas (more hydrophobic OA and less SO$_4$) leading to less particle growth in the airways. While the deposited mass in Figures 7 and 8

![Figure 8. The $\text{DR}_{\text{TB + AV}}$ based on (a) SO$_4$ in the higher-resolution North American domain, (b) SO$_4$ in the Asian domain, (c) organic aerosol (OA) in the North American domain, and (d) OA in the Asian domain.](image-url)
are only from SO$_4$ and OA, the water uptake in the lung is based on the total particle hygroscopicity in a given size range (i.e., we assume internally mixed particles). Particle sizes in urban areas tend to be closer to the minimum in deposition fraction (Figure 1) than rural regions, where increased atmospheric processing increases hydrophilic OA and SO$_4$ concentrations, thus increasing overall hygroscopicity and particle growth to sizes with more efficient deposition in the TB + AV. Thus, contributions from non-SO$_4$ and OA species still influence the particle size distributions. Conversely, this urban/rural split is less apparent in India and China due to the larger ambient accumulation-mode median diameters in these regions. Biomass burning leads to a lower DRTB + AV than fossil-fuel-emission regions in both the North American domain (wildfires in Northern and Eastern Canada) and the Asian domain (in Indonesia and the Philippines) due to larger emitted particle diameters and higher hygroscopic particle fractions than fossil-fuel emissions.

3.4. Deposition Ratios Based on Number and Surface Area

In addition to considering the health response due to different PM species, it has also been suggested that particle number and/or surface area may be key parameters influencing health response due to PM$_{2.5}$ exposure (e.g., Breitner et al., 2011; Delfino et al., 2005; Peters et al., 1997, 2006; Valavanidis et al., 2008). To test the sensitivity to these other PM quantities, Figures 9a–9c show the DR$_{total}$ based on (a) particle number concentration (total respiratory deposited number per unit ambient PM$_{2.5}$ mass), (b) particle surface area concentration (total respiratory deposited dry surface area per unit ambient PM$_{2.5}$ mass), and (c) particle mass concentration (total respiratory deposited mass per unit ambient PM$_{2.5}$ mass). As seen in Figures 9a and 9b, DR$_{total}$ based on number and surface area have substantially different geographic variability than the analogous DR$_{total}$ based on mass (Figure 9c). The DR$_{total}$ based on number has maxima in regions where new particle formation events are common but low overall PM$_{2.5}$ mass concentration. This results in high concentrations of sub-50-nm particles, where the ICRP deposition curves have maximum values (Figure 1).
Such regions include the high-altitude regions of the Rocky Mountains and Himalayas, along with South Africa and South America. Maxima in DR\textsubscript{total} by surface area and mass occur in desert regions where there is substantial coarse-mode mass at particle size with efficient deposition fractions in the head/nose region (Figure 1).

Similarly, in Figures 9d–9f, we show the DR\textsubscript{TB + AV} based on (d) number, (e) surface area, and (f) mass. Figure 9f is identical to Figure 6a except with a different color bar. Comparing DR\textsubscript{total} to DR\textsubscript{TB + AV}, much of the variability in respiratory deposition based on number is driven by deposition in the TB + AV. Conversely, much of the variability in deposition by surface area is driven by deposition in all respiratory regions (especially the head/nose regions). The spatial variability in DR\textsubscript{TB + AV} based on surface area seems to share features with DR\textsubscript{TB + AV} from both number (such as in the Rocky Mountains in the western United States) and mass (such as in the Sahara). Deposition based on mass shows substantial variability for DR\textsubscript{total} and DR\textsubscript{TB + AV}, suggesting a high degree of variability due to ambient particle size distribution for each respiratory deposition site considered here. The spatial distributions present in Figure 9 demonstrate the potential variability in health response to PM\textsubscript{2.5} exposure if the health response to exposure is more strongly correlated to deposition by number, surface area, or mass. Given the DR\textsubscript{TB + AV} based on mass, surface area, and number show different spatial distributions, it is essential to understand what PM parameters control the health response to particle exposure.

4. Discussion and Conclusions

While studies consistently find a robust relationship with PM\textsubscript{2.5} exposure and negative health effects, variability in the health response to PM\textsubscript{2.5} exposure remains. The aim of this work is to motivate consideration of the role that particle size may play in determining the variability of the health response to PM\textsubscript{2.5} exposure. To explore this, we combine simulated aerosol size distributions from a chemical-transport model with a size-resolved particle dosimetry model to explore the geographic variability of deposited particle mass in the body. We find that on global scales, the ratio of deposited PM mass per unit ambient PM\textsubscript{2.5} mass in the tracheobronchial and alveolar regions varies by 20–30% between different highly populated regions (e.g., Eastern United States versus India) due to variability in ambient particle size distributions. This variability is also seen within countries using higher-resolution nested simulations, though these higher-resolution simulations are likely still too coarse to capture near-source gradients in aerosol size distributions. This variability is a reflection of local emissions and microphysical processes that affect ambient aerosol size distributions. In some regions, coarse-mode mass contributes a substantial portion of deposited PM mass in the tracheobronchial and alveolar regions. This mass is not included in ambient PM\textsubscript{2.5} exposure measurements, resulting in variability up to a factor of 4 between regions dominated by fossil-fuel emissions (e.g., the Eastern United States and western Europe) and regions dominated by dust (e.g., North Africa).

To some degree, the variability in this deposition ratio is influenced by particle composition. While the ICRP model predicts deposition fractions of around 10% for particle diameters greater than 2.5 \(\mu\text{m}\), the large mass concentration of coarse-mode dust in desert regions leads to high deposition ratios. This result further motivates the question of the toxicity of dust relative anthropogenic emitted species of sulfate, OA, and BC. We still find geographic variability on the order of 30% for sulfate and OA in the tracheobronchial and alveolar regions. As this spatial pattern is different than total PM mass, this further motivates understanding potential differential toxicity of PM chemical species. In addition to chemical composition, we also compared deposition ratios based on number and surface area and find that the spatial variability is substantially different when based on number, surface area, or mass. These results show that regardless of the specific health impacts of individual PM species or parameters (number, surface area, and mass), we may likely need to consider variability in the aerosol size distribution to fully connect exposure to dose and ultimately the health impacts.

A limitation of this work is the lack of observed aerosol size distributions. Long-term measurements of aerosol size distributions ranging from diameters of 1–10 nm to 5–10 \(\mu\text{m}\) are limited. Size distribution measurements in polluted regions sub-Saharan Africa, India, and China are especially limited. In this work, we use a sectional aerosol microphysical model (TOMAS) that has been shown to have skill in capturing the variability in long-term size-resolved aerosol measurements within size ranges commonly measured at sites in Europe and North America. Despite this skill, the coarse spatial and temporal resolution of the model reduces gradients...
in aerosol size distributions, particularly in urban areas. This suggests that variability in aerosol size distributions at regional and continental scales may be greater than what is considered here. We also note here that there is likely substantial uncertainty in size-specific particle deposition fractions in various regions of the body. Despite these uncertainties, we know that particle deposition has a strong size dependence across several orders of magnitude in particle diameter, and geographic variability in aerosol size distributions has been commonly observed. The results of this work are a first step in exploring the geographic variability of respiratory particle deposition rates.

References

Adams, P. J., & Seinfeld, J. H. (2002). Predicting global aerosol size distributions in general circulation models. *Journal of Geophysical Research*, 107(D19), 4370. https://doi.org/10.1029/2001JD001010

Adams, P. J., & Seinfeld, J. H. (2003). Disproportionate impact of particulate emissions on global cloud condensation nuclei concentrations. *Geophysical Research Letters*, 30(5), 1239. https://doi.org/10.1029/2002GL016303

Anselm, A., Heibel, T., Gebhart, J., & Ferron, G. (1990). “In vivo”-studies of growth factors of sodium chloride particles in the human respiratory tract. *Journal of Aerosol Science*, 21, 5427–5430. https://doi.org/10.1016/0021-8502(90)90272-Y

Ban-Weiss, G. A., Lunden, M. M., Kirchstetter, T. W., & Harley, R. A. (2010). Size-resolved particle number and volume emission factors for on-road gasoline and diesel motor vehicles. *Journal of Aerosol Science*, 41(1), 5–12. https://doi.org/10.1016/j.jaerosc.2009.08.001

Bates, J. T., Weber, R. J., Abrams, J., Verma, F., Fang, T., Klein, M., et al. (2015). Reactive oxygen species generation linked to sources of atmospheric particulate matter and cardiorespiratory effects. *Environmental Science & Technology*, 49(22), 13605–13612. https://doi.org/10.1021/acs.est.5b02967

Beelen, R., Hoek, G., van den Brandt, P. A., Goldbohm, R. A., Fischer, P., Schouten, L. J., et al. (2008). Long-term effects of traffic-related Air pollution on mortality in a Dutch cohort study (NLCS-AIR study). *Environmental Health Perspectives*, 116(2), 196–202. https://doi.org/10.1289/ehp.10767

Bell, M. L., Dominici, F., Ebisu, K., Zeger, S. L., & Samet, J. M. (2007). Spatial and temporal variation in PM(2.5) chemical composition in the United States for health effects studies. *Environmental Health Perspectives*, 115(7), 989–995. https://doi.org/10.1289/ehp.9621

Bey, I., Jacob, D. J., Yantosca, R. M., Logan, J. A., Field, B. D., Fiore, A. M., et al. (2001). Global modeling of tropospheric chemistry with a fully interactive aerosol module: TOMAS model is freely available to model-mppd-v-304. The GEOS-Chem particle deposition model is available: https://www.ara.com/products/multiple-path-particle-dosimetry-model-mppd-v-304. The GEOS-Chem-TOMAS model is freely available in ICRP Publication 66 (ICRP, 1994). The MPFD particle deposition model is available: https://www.arca.com/products/gcst/gcst-geochem. Documentation for GEOS-Chem is available here: http://acmg.seas.harvard.edu/geos/doc/man/index.html.

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