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Abstract

Global measurements of the elemental composition of fine particulate matter across several urban locations by the Surface Particulate Matter Network reveal an enhanced fraction of anthropogenic dust compared to natural dust sources, especially over Asia. We develop a global simulation of anthropogenic fugitive, combustion, and industrial dust which, to our knowledge, is partially missing or strongly underrepresented in global models. We estimate 2-16 $\mu g/m^3$ increase in fine particulate mass concentration across East and South Asia by including anthropogenic fugitive, combustion, and industrial dust emissions. A simulation including anthropogenic fugitive, combustion, and industrial dust emissions increases the correlation from 0.06 to 0.66 of simulated fine dust in comparison with Surface Particulate Matter Network measurements at 13 globally dispersed locations, and reduces the low bias by 10% in total fine particulate mass in comparison with global in situ observations. Global population-weighted $PM_{2.5}$ increases by 2.9 $\mu g/m^3$ (10%). Our assessment ascertains the urgent need of including this underrepresented fine anthropogenic dust source into global bottom-up emission inventories and global models.

1. Introduction

Outdoor $PM_{2.5}$ (fine particulate matter with aerodynamic diameter less than 2.5 micrometers) is the fifth largest risk factor for premature mortality worldwide (Forouzanfar et al 2016). Global atmospheric models are widely used for assessments of exposure to outdoor $PM_{2.5}$ (Anenberg et al 2010, Giannadaki et al 2014, Lee et al 2015, Lelieveld et al 2015, Brauer et al 2016, West et al 2016). Total $PM_{2.5}$ is mainly composed of a carbonaceous component, inorganic ions, and mineral dust. The latter includes three broad categories, mineral dust naturally windblown from arid desert regions (Prospero et al 2002), anthropogenic windblown dust from human disturbed soils due to changes in land use practices, deforestation and agriculture (Tegen et al 1996, 2004), and anthropogenic fugitive, combustion, and industrial dust (AFCID) from urban sources. Global models typically include natural mineral dust (Huneeus et al 2011, Astitha et al 2012) with recent developments to assess the relative contribution of anthropogenic windblown dust (Ginoux et al 2012, Huang et al 2015, Guan et al

2016). However, to our knowledge, AFCID is partially missing or strongly underrepresented from global models (Rind et al 2009) as evident from model descriptions published as part of several multi-model inter-comparison studies (Schulz et al 2006, Myhre et al 2013, Pan et al 2015, Silva et al 2013, Huneeus et al 2011).

Measurements of $PM_{2.5}$ and its chemical composition over several urban locations by the Surface Particulate Matter Network (SPARTAN) offer information about $PM_{2.5}$ sources (Snider et al 2015, 2016). Snider et al (2016) found an enhanced fraction of AFCID compared to natural sources over several Asian cities, evidenced by a high zinc (mainly anthropogenic as evidenced by Councell et al 2004 and Harrison et al 2012) to aluminum (mainly natural) ratio in $PM_{2.5}$ dust. Sources of anthropogenic fugitive, combustion, and industrial dust include elemental components from coal combustion (fly ash) and industrial processes (e.g. iron and steel production, cement production), resuspension from paved and unpaved roads, mining, quarrying, and agricultural operations, and road-residential-commercial construction (McElroy et al 1982, Watson and Chow 2000, Guttikunda et al 2014). Some evidence for the significance of these anthropogenic fugitive, combustion, and industrial sources to ambient $PM_{2.5}$ dust is emerging through measurements and source apportionment studies (Yang et al 2011, Yu et al 2013, Zhang R et al 2013, Zhang et al 2015, Viana et al 2008, Mooibroek et al 2011). Despite the majority of these emissions being in the coarse mode there is a tail that contributes to $PM_{2.5}$. AFCID includes several trace elements that are associated with adverse health effects, but not yet well understood (West et al 2016).

The few global emission inventories that include anthropogenic primary emissions of total $PM_{2.5}$ have limited distinction between estimates of fugitive, combustion and industrial dust, and rather incomplete representation of fugitive sources (e.g., Janssens-Maenhout et al 2015; Klimont et al in review). A few global simulations have included a portion of the AFCID inventory (Shindell et al 2012, Anenberg et al 2012, Myhre et al in review). Some regional inventories explicitly provide some portion of $PM_{2.5}$ AFCID as a separate source category (e.g., Pouliot et al 2015) enabling inclusion in regional chemical transport models and air quality models (e.g., Park et al 2010, Guttikunda and Jawahar 2012, Appel et al 2013, Zhang et al 2015). However, the contribution of AFCID sources to $PM_{2.5}$ mass remains poorly quantified, especially at the global scale.

Several global and regional models tend to consistently underestimate aerosol loading (Moorthy et al 2013, Pan et al 2015, Lelieveld et al 2015, Brauer et al 2016). We hypothesize that inclusion of missing AFCID sources will reconcile some of the unexplained bias. Here, we develop a global simulation of anthropogenic fugitive, combustion, and industrial dust, and evaluate it with in situ measurements.

2. Materials and Methods

We interpret Surface Particulate Matter Network (www.spartan-network.org) measurements of $PM_{2.5}$ and trace metals collected from monitoring stations over geographically diverse global regions to evaluate our simulation of AFCID (Snider et al 2015, 2016). SPARTAN measurements include an AirPhoton SS4i automated air sampler to collect aerosol on PTFE filters for gravimetric assessment of $PM_{2.5}$ mass, and Inductively Coupled Plasma - Mass Spectrometry to quantify $PM_{2.5}$ trace metals used to determine crustal $PM_{2.5}$ (Snider et al 2016). Measurement sites are primarily in urban locations with site selection designed for spatial representativeness. SPARTAN measurements exhibit a high degree of consistency with independent measurements over Asia (Beijing, Bandung, Kanpur and Hanoi), the U.S. (Mammoth Cave and Atlanta) and elsewhere (Snider et al 2015, 2016).

We obtain global monthly mean anthropogenic emissions of primary particulate matter (including fugitive, combustion, and industrial dust) in 2015 from the ECLIPSE dataset (version V5a; http://www.iiasa.ac.at/web/home/research/researchPrograms/air/Global_emissions.html). Klimont et al (in review) developed this inventory with the GAINS (Greenhouse gas - Air pollution INteractions and Synergies) model (Amann et al 2011) for the European Union funded project ECLIPSE (Evaluating the Climate and Air Quality Impacts of Short-Lived Pollutants) (Stohl et al 2015, http://eclipse.nilu.no). AFCID is represented as the residual of anthropogenic primary emissions of $PM_{2.5}$, after excluding particulate organic mass and black carbon. We overwrite this global inventory with two regional monthly mean emission inventories, over India with the AFCID emission inventory from the Indian Institute of Technology - Bombay (IIT-B) for 2013, and over China with the Multi-resolution Emission Inventory for China (MEIC) inventory (Lei et al 2011, Zhang et al 2009, http://www.meicmodel.org) for 2012. We convert emission of organic carbon in MEIC inventory to

particulate organic mass following Philip et al (2014b). We treat primary emissions of sulfate as 3% of sulfur dioxide emissions (Chin et al 2000), and subtract it from the primary $PM_{2.5}$ emissions. The resultant global annual AFCID inventory is 13.1 Tg/yr.

The anthropogenic primary $PM_{2.5}$ emission inventories are derived using a dynamic technology-based approach employing high source-activity-sector resolution at a country or even subnational level. For each of the emission sources, the models applied to calculate these inventories define activity rate, unabated emission factors, penetration and removal efficiency of applicable emission control technologies (Lei et al 2011, Klimont et al in review). The data and assumptions used in the inventories draw on international and national statistics, on an array of measurement studies representative for typical sources and applied technologies considering local circumstances and studies, and on information about the air quality legislation and efficiency of its enforcement allowing defining of the penetration of control measures. These inventories include a harmonized calculation of mass-based size distribution ($PM_{2.5}$, PM_{10}) and primary carbonaceous aerosols. The characteristics of sources vary strongly with respect to the contribution of carbonaceous particles and the underlying models capture these features by defining mass-based consistent emission factors and removal efficiencies for total $PM_{2.5}$, black carbon, organic carbon and particulate organic mass. Compared to previous global work, ECLIPSE includes estimates for a number previously unaccounted or often underestimated PM sources, that is, gas flaring, kerosene lamps, diesel generators (Klimont et al in review).

We conduct a simulation of anthropogenic fugitive, combustion, and industrial dust with the GEOS-Chem global 3-D chemical transport model (Bey et al 2001) version 11-01b (http://geos-chem.org) driven with assimilated meteorological fields from the Goddard Earth Observing System (GEOS-FP) at the NASA Global Modeling Assimilation Office, with a horizontal resolution of 2^o x 2.5^o. GEOS-Chem includes a detailed simulation of oxidant-aerosol chemistry (Bey et al 2001, Park et al 2004) with secondary inorganic aerosols (Park et al 2004), black carbon and organic carbon (Park et al 2003), secondary organic aerosol (SOA) (Pye et al 2010), and sea salt (Jaegle et al 2011). The mineral dust simulation in GEOS-Chem follows the Dust Entrainment and Deposition (DEAD) mobilization scheme (Zender et al 2003) with a topographic source function (Ginoux et al 2001, Chin

et al 2004) implemented by Fairlie et al (2007), and an optimized dust particle size distribution implemented by Zhang L et al (2013). For computational convenience, we treat AFCID as part of the finest GEOS-Chem dust bin (with diameter less than 2 μm). GEOS-Chem simulations have been extensively applied to natural mineral dust (Fairlie et al 2007, 2010, Ridley et al 2012, Johnson et al 2012, Wang et al 2012, Zhang L et al 2013), $PM_{2.5,}$ (van Donkelaar et al 2010, Tai et al 2012, Xu et al 2015, Ford and Heald 2016, Koplitz et al 2016), and chemical components of $PM_{2.5}$ (Park et al 2004, Philip et al 2014a, Kim et al 2015).

We use the HEMCO module (Keller et al 2014) to implement the AFCID emission inventory into GEOS-Chem. We conduct simulations from January 1, 2014 to December 31, 2015 following a one month spin-up. We use operator durations of 10 min for transport and 20 min for chemistry for optimized computational speed and accuracy (Philip et al 2016). We calculate ground-level $PM_{2.5}$ at 35% relative humidity to follow common measurement protocols. We convert organic carbon to particulate organic mass following Philip et al (2014b). We evaluate simulated $PM_{2.5}$ with annual mean direct $PM_{2.5}$ in situ measurements collected for the GBD-2013 study (van Donkelaar et al 2015, Brauer et al 2016), and SPARTAN measurements of campaign-mean (2013-2015) $PM_{2.5}$ composition (Snider et al 2016). We use population for the year 2015 from the National Aeronautics and Space Administration Socioeconomic Data and Applications Center (CIESIN 2016) to estimate population-weighted $PM_{2.5}$.

3. Results and Discussion

The top panel of Figure 1 shows filled concentric circles of campaign-mean $PM_{2.5}$ dust (inner circles) measured by the SPARTAN network over 13 globally dispersed locations, for the years 2013-2015 (Snider et al 2016). SPARTAN dust mass (and % of total $PM_{2.5}$) varies from ~1 $\mu g/m^3$ (~10%) over North America, ~5 $\mu g/m^3$ (5-15%) over South and South East Asian cities (Kanpur, Dhaka, Hanoi) to ~14 $\mu g/m^3$ (~25%) over Beijing (Snider et al 2015, 2016). Enhanced Zn:Al ratios measured over these sites provide evidence of an anthropogenic source (Snider et al 2016). The middle panel of Figure 1 shows the GEOS-Chem simulated natural mineral dust. Natural mineral dust concentrations

are enhanced over regions with accumulated alluvial sediments, predominantly over arid and semi-arid regions of North Africa, the Middle East and Central Asia (Zender et al 2003, Fairlie et al 2007, Huneeus et al 2011). It is evident that the pronounced dust concentrations measured over East and South Asia cannot be explained by natural mineral dust alone (Lei et al 2011, Zhang et al 2015).

The bottom panel of Figure 1 shows the simulation of anthropogenic fugitive, combustion, and industrial dust. AFCID increases $PM_{2.5}$ dust concentrations by 2-16 $\mu g/m^3$ over much of East and South Asia. The concentration of simulated AFCID is comparable to that of natural mineral dust over parts of Europe and Eastern North America. Other regional studies (Appel et al 2013, Park et al 2010) offer additional evidence of AFCID sources.

The top panel of Figure 1 shows that GEOS-Chem simulated AFCID in addition to default natural mineral dust reduces the bias in total dust mass measured at SPARTAN sites over Asia. A high AFCID over Beijing reveals the significance of regional fugitive sources (Yu et al 2013, Zhang R et al 2013, Zhang et al 2015). Zhang et al (2015) use the adjoint of GEOS-Chem together with the MEIC inventory to attribute 27% of wintertime $PM_{2.5}$ over Beijing from emissions of AFCID from North China.

Table 1 contains statistics describing the comparison of GEOS-Chem simulated concentrations versus in situ observations. The inclusion of AFCID increases the correlation versus $PM_{2.5}$ dust mass concentration from 0.06 to 0.66 over all SPARTAN sites compared to campaign-mean data. A test case study that excludes two arid sites (Ilorin, Nigeria and Rehovot, Israel) dominated by large simulated natural mineral dust loading also reveals an improved consistency from slope = 0.29 (r = 0.77) to slope = 1.29 (r = 0.91) further demonstrating the importance of AFCID at the global scale.

Figure 2 shows the in situ and simulated concentration of total $PM_{2.5}$. The top panel shows enhanced $PM_{2.5}$ concentrations in the in situ measurements over rapidly developing Asia. The bottom panel shows that the simulation with AFCID largely reproduces these enhancements. We find that simulated AFCID comprises 5-15% of total $PM_{2.5}$ across large parts of East and South Asia.

Table 1 quantifies the comparison of GEOS-Chem simulated $PM_{2.5}$ concentrations versus long-term annual mean in situ measurements compiled by Brauer et al (2016) for the Global Burden

of Disease Study. Site locations span a diversity of environments including routine monitoring networks in both densely populated and remote areas. The additional $PM_{2.5}$ source from AFCID increases the slope of the best fit line from 0.83 to 0.93. This analysis reveals that neglect of AFCID in $PM_{2.5}$ can underestimate ambient $PM_{2.5}$ concentrations by 5-10% globally, and by up to 15% in East and South Asia. Global population-weighted $PM_{2.5}$ concentrations increase by 2.9 $\mu g/m^3$ (10%) with implications for future assessments of $PM_{2.5}$ health effects.

4. Conclusions

$PM_{2.5}$ health impact assessments require a complete description of $PM_{2.5}$ sources. We interpret global crustal $PM_{2.5}$ observations from the SPARTAN network and find evidence for anthropogenic fugitive, combustion, and industrial dust. A collection of emission inventories (ECLIPSE, IIT-B and MEIC) was used to estimate AFCID emissions for inclusion into a GEOS-Chem simulation. Inclusion of AFCID increased total $PM_{2.5}$ mass by 2-16 $\mu g/m^3$ over anthropogenic polluted regions across East and South Asia, reducing the observed bias from 17% to 7% in comparison with the global $PM_{2.5}$ in situ observations, and increasing the correlation from 0.06 to 0.66 of $PM_{2.5}$ dust concentration compared to SPARTAN in situ observations. Global population-weighted $PM_{2.5}$ concentrations increase by 2.9 $\mu g/m^3$ (10%). The noteworthy contribution of this underrepresented AFCID source to $PM_{2.5}$ mass as evaluated with observations, motivate further development and incorporation of AFCID emission into global models. To our knowledge, this is the first global assessment of the importance of anthropogenic fugitive, combustion, and industrial dust. Nonetheless some portion of this anthropogenic dust source might not be captured well in our inventories, with potential uncertainty in our estimates. Future work should assess the implications of coarse mode AFCID that may be associated with the $PM_{2.5}$ examined here. Although we focus on the ground-level $PM_{2.5}$ owing to its importance in human health impact studies, estimating AFCID and understanding its optical and transport properties could benefit studies of climate forcing (Rind et al 2009) and visibility.

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Table 1. Comparison of GEOS-Chem simulated concentrations (2014-2015) versus measured in situ observations of long-term annual mean $PM_{2.5}$ mass compiled by Brauer et al (2016), and of campaign-mean (2013-2015) crustal $PM_{2.5}$ by the SPARTAN network (Snider et al 2016). AFCID denotes anthropogenic fugitive, combustion, and industrial dust. Reduced major axis regression is used to calculate correlation statistics.

	$PM_{2.5}$ Data from Brauer et al (2016)				$PM_{2.5}$ Dust SPARTAN (All sites)				$PM_{2.5}$ Dust SPARTAN (Except arid sites) [a]			
	r	Slope	Offset ($\mu g/m^3$)	N	r	Slope	Offset ($\mu g/m^3$)	N	r	Slope	Offset ($\mu g/m^3$)	N
GEOS-Chem Default	0.82	0.83	-1.17	441	0.06	1.06	-1.75	13	0.77	0.29	-0.30	11
GEOS-Chem with AFCID	0.83	0.93	-2.01	441	0.66	1.55	-1.00	13	0.91	1.29	-1.53	11

[a] Excluding sites in North Africa (Ilorine, Nigeria) and Middle East (Rehovot, Israel) where natural mineral dust dominates.

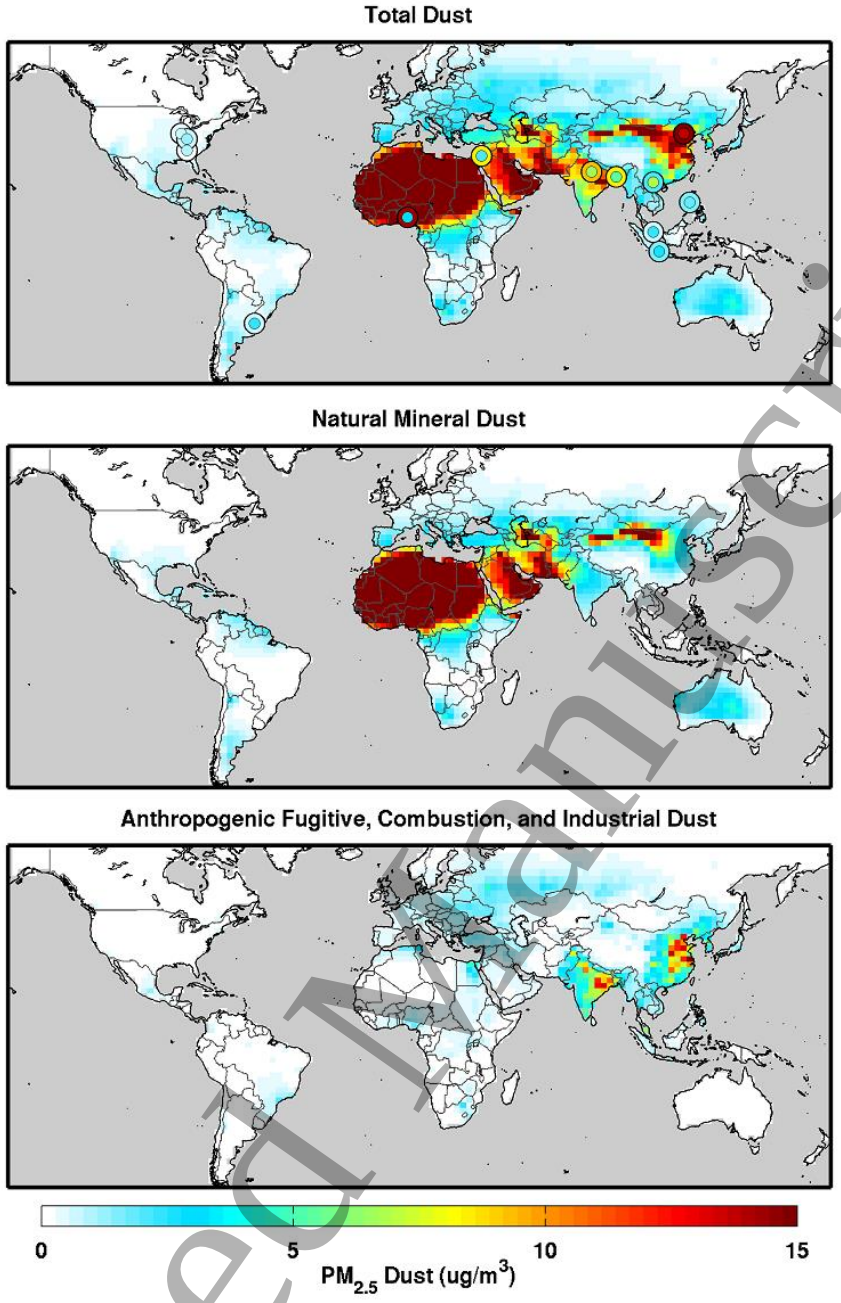

Figure 1. Annual mean (2014-2015) concentration of $PM_{2.5}$ total dust (top panel), natural mineral dust (middle panel), and anthropogenic fugitive, combustion, and industrial dust (bottom panel) simulated with the GEOS-Chem model. Colored concentric circles in the top panel denote SPARTAN-measured campaign-mean (2013-2015) $PM_{2.5}$ dust concentration (inner circle) and the coincident simulated value (outer circle).

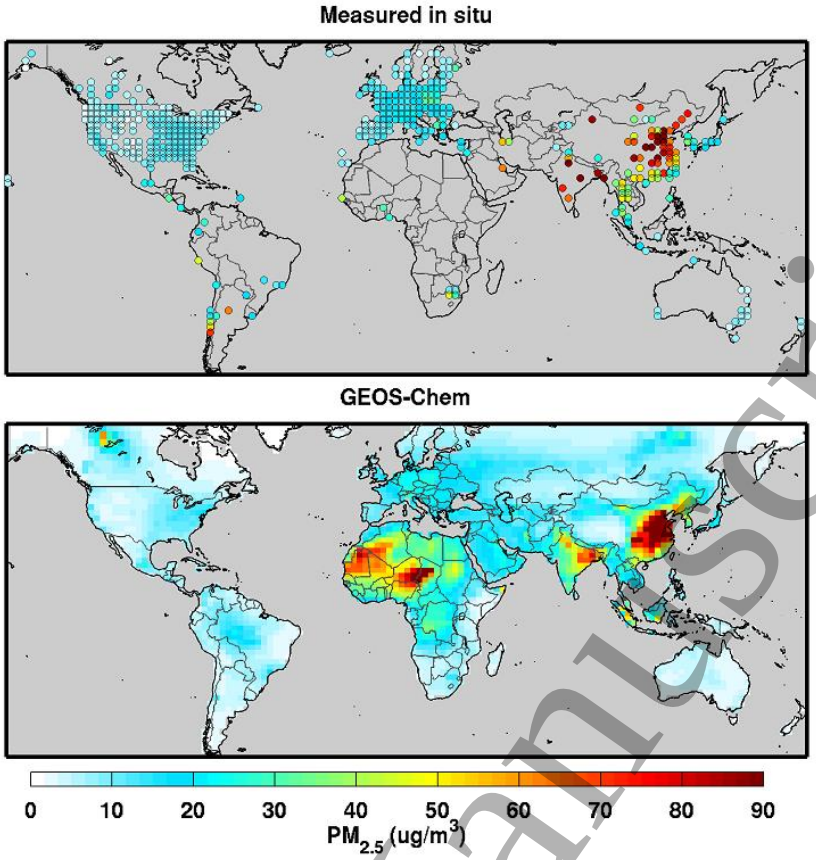

Figure 2. Long-term annual mean measured in situ $PM_{2.5}$ observations compiled by Brauer et al (2016) regridded to model horizontal resolution (top panel). Annual mean (2014-2015) $PM_{2.5}$ concentration simulated with the GEOS-Chem model incorporating anthropogenic fugitive, combustion, and industrial dust (bottom panel).