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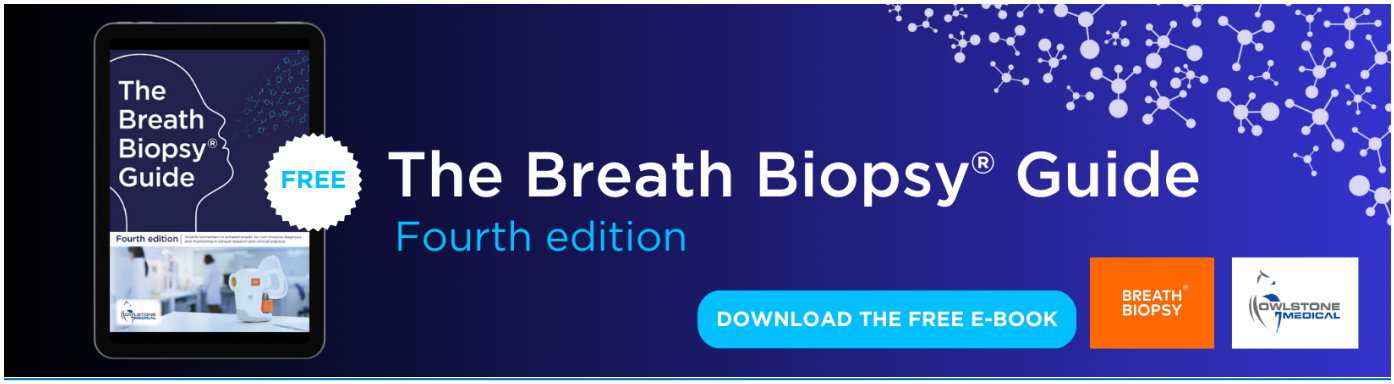

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LETTER

Anthropogenic fugitive, combustion and industrial dust is a significant, underrepresented fine particulate matter source in global atmospheric models

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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 *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* 2016). A few global simulations have included a portion of the AFCID inventory (Shindell *et al* 2012, Anenberg *et al* 2012, Myhre *et al* 2017). 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; www.iiasa.ac.at/web/home/research/researchPrograms/air/Global_emissions.html). Klimont *et al* (2016) 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, 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^{-1}.

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* 2016). 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* 2016).

We conduct a simulation of anthropogenic fugitive, combustion, and industrial dust with the GEOS-Chem global 3D 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 $2^{o} \times 2.5^{o}$. 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 *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 *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 $\sim$1 μg m^{-3} ($\sim$10%) over North America, $\sim$5 μg m^{-3} (5%–15%) over South and South East Asian cities (Kanpur, Dhaka, Hanoi) to $\sim$14 μg m^{-3} ($\sim$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 μg 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 *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

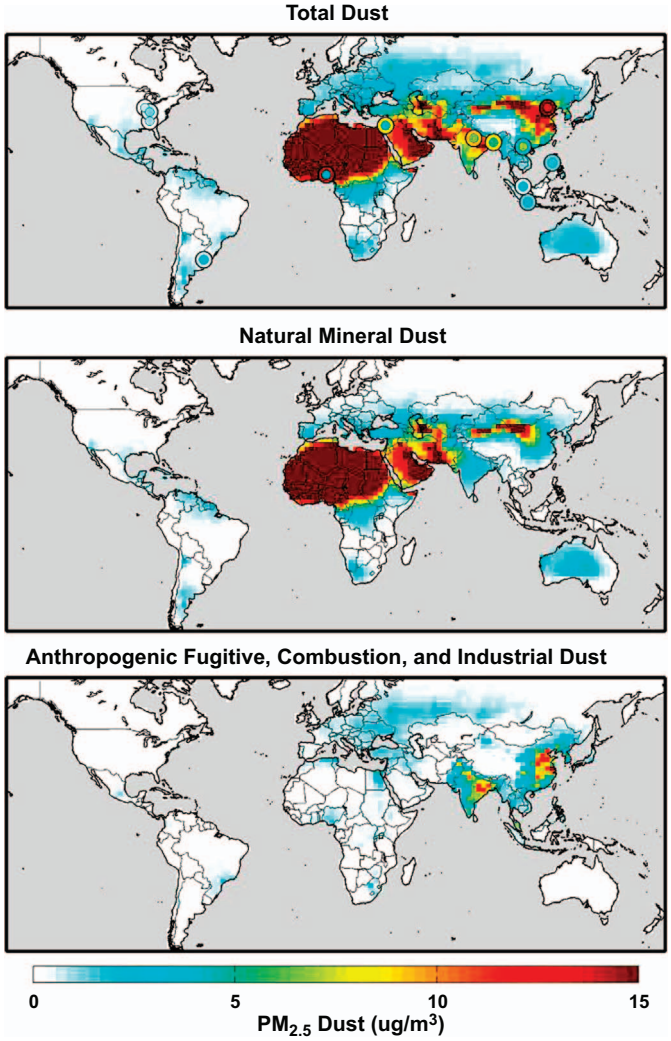

Figure 1. Annual mean (2014–2015) concentration of $PM_{2.5}$ total dust (top panel), natural mineral dust (middle panel), and anthropogenic fugitive, combustion, and industrial dust (bottom panel) simulated with the GEOS-Chem model. Colored concentric circles in the top panel denote SPARTAN-measured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

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.

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%

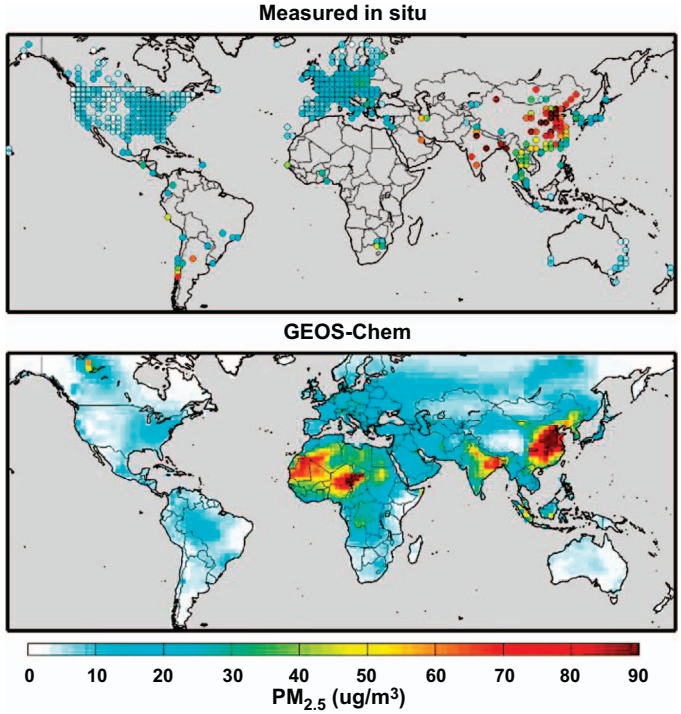

Figure 2. Long-term annual mean measured *in situ* $PM_{2.5}$ observations compiled by Brauer *et al* (2016) regridded to model horizontal resolution (top panel). Annual mean (2014–2015) $PM_{2.5}$ concentration simulated with the GEOS-Chem model incorporating anthropogenic fugitive, combustion, and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

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 μg 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 μg 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 μg 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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