Supplemental Material

Global mortality from outdoor fine particle pollution generated by fossil fuel combustion: Results from GEOS-Chem

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Description of GEOS-Chem.

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**Table S2. Extended data.** Global regions, number of deaths, attributable fraction (%) for the population above 14 years old attributable to fine particulate matter (PM$_{2.5}$) exposure.

**References**

**Description of GEOS-Chem.**

GEOS-Chem is a three-dimensional chemical transport model that includes detailed oxidant-aerosol chemistry in the troposphere and is used by more than 80 groups worldwide (www.geos-chem.org). The model is widely cited in the peer-reviewed literature – e.g., more than 4000 times in the year 2017 alone (http://acmg.seas.harvard.edu/geos/geos_pub.html). The model has been frequently applied to interpret observed PM$_{2.5}$ in regions dominated by anthropogenic sources – e.g., China (Aunan et al., 2018), Korea (Lee et al., 2017), India (Venkataraman et al., 2018), and the US (Di et al., 2016; Silvern et al., 2017); and validation has been performed for specific source sectors – e.g., transportation (Travis et al., 2016), biogenic sources (Marais et al., 2017), and power plants (S. W. Wang et al., 2012). Here we use GEOS-Chem v10-01, driven by 2012 GEOS-5 meteorology (gmao.gsfc.nasa.gov/GEOS_systems/). The GEOS-5 data are produced at 0.5°×0.667° horizontal resolution and are re-gridded here to 2°×2.5° for the global simulation.

We also perform four regional simulations – for Europe, North America, Africa, and Asia – and for these simulations we keep the native grid resolution. Boundary conditions at 2°×2.5° from the global simulation are applied to these regional simulations. Most fine-scale, regional models, such as the Community Multiscale Air Quality Model, rely on chemical boundary conditions from global models with different chemical schemes, but our approach permits application of a consistent scheme across the globe. The 0.5°×0.667° horizontal resolution in GEOS-Chem over key regions is, however, relatively coarse compared to that in some other regional models. Y. Li et al. (2016) show that application of coarse resolution leads
to an underestimate of health impacts of 8%, implying that our mortality estimates are conservative. Our choice of 2012 as the simulation year is discussed below.

GEOS-Chem simulates the mass concentrations of key particle types including sulfate, nitrate, and ammonium (Park et al., 2004; L. Zhang et al., 2012), organic carbon (Heald et al., 2006; 2011) black carbon (Q. Q. Wang et al., 2014), dust (Fairlie et al., 2007), and sea salt (Jaegle et al., 2011). Particle chemistry is coupled to gas-phase chemistry as described by (Mao et al., 2013). Gas/particle partitioning of sulfate, nitrate and ammonium (SNA) particles is computed with the ISORROPIA II thermodynamic module (Fountoukis and Nenes, 2007; Pye et al., 2009). Wet and dry deposition of particles follow Liu et al. (2001) and L. M. Zhang et al. (2001), respectively.

Emissions in GEOS-Chem are computed by the Harvard-NASA Emission Component (HEMCO) (Keller et al., 2014), which combines and regrids ensembles of user-selected emission inventories. We apply global anthropogenic emissions but supersede these with regional emissions where the latter are more reliable (Table 1). Fossil fuel emissions in Africa include (1) industry and power plants from the global inventories and (2) diffuse and inefficient combustion sources (diesel and petrol generators, ad-hoc oil refining, gas flares, kerosene use, cars, and motorcycles) from the DICE-Africa inventory (Marais and Wiedinmyer, 2016). We scale all anthropogenic inventories to 2012, as described by van Donkelaar et al. (2008). Biogenic emissions are from MEGAN v2.1 for volatile organic compounds (Guenther et al., 2012) and from Hudman et al. (2012) for soil nitrogen oxides. Lightning emissions of nitrogen oxides are computed as a function of cloud top height as described by Murray et al. (2012). Dust entrainment and deposition follow the DEAD scheme of Zender et al. (2003) as implemented in GEOS-Chem by Fairlie et al. (2007). Biomass burning emissions are from the Global Fire Emissions Database version 4 (GFED4) (Giglio et al., 2013).

For this study, we first calculate the surface fine particle mass concentrations (PM$_{2.5}$), with all emissions sources turned on. For consistency with the PM$_{2.5}$ measurement protocol set by the U.S. Environmental
We assume 35% relative humidity everywhere (except for Europe) and standard ambient conditions, with temperature of 298.15 K and surface pressure of 1013.25 hPa. In Europe, we assume 50% relative humidity, as is the protocol there. We then perform the identical simulation with emissions arising from fossil fuel combustion turned off. The same meteorological fields are applied for both simulations – i.e., the simulation does not allow feedbacks from particles onto meteorology. In the no-fossil-fuel case, all fossil fuel sources are turned off in both the nested simulations and in the global simulation providing boundary conditions. The difference between the two simulations (with and without fossil fuel) represents the contribution of fossil-fuel combustion to surface PM$_{2.5}$. This approach assumes a linear response of surface PM$_{2.5}$ to changes in emissions.

Our choice of 2012 as the simulation year requires explanation. Air quality is influenced not just by emissions but also by meteorological variables such as surface temperature and wind speed, which can vary greatly on inter-annual timescales. Ideally, our analysis would involve multi-year simulations on both the coarse- and fine-scale grids, but such effort would be computationally expensive. We choose instead to do a one-year simulation for a year not influenced by El Niño conditions, which can worsen or ameliorate air pollution, depending on the region (e.g., Chang et al. (2016), Shen and Mickley (2017)). To gauge the error implied by our choice to simulate just one year rather than a span of years, we examine the inter-annual variation in total PM$_{2.5}$ concentrations at the surface estimated from the Dalhousie University archive (van Donkelaar et al., 2016). The PM$_{2.5}$ values in the Dalhousie archive are calculated by first combining satellite observations with GEOS-Chem estimates, and then calibrating the resulting concentrations with available ground-based observations (mostly Europe, the US, India and China). We find that the global mean average of the relative standard deviation of total PM$_{2.5}$ in the Dalhousie archive over 2008 to 2016 is just 7%.

Averaged over large regions on the continental scale, the relative standard deviation ranges from 4% over Australia to 11% over the Asia nested grid domain (Figure S1). Inter-annual variability in this metric is greatest (> 60%) for smaller regions influenced by wildfires or biomass burning – e.g., Indonesia and remote areas at high northern latitudes where few people live. To test our choice of 2012 as a representative year,
we calculate the 2012 anomaly in the Dalhousie PM$_{2.5}$ time series (Figure S2). Again on a continental scale, we find that 2012 concentrations range from 0.7 µg m$^{-3}$ less to 0.4 µg m$^{-3}$ greater than the 2008-2016 average (Figure S2). Given the relatively small inter-annual variability in surface PM$_{2.5}$ in the Dalhousie archive over most populated regions, as well as the small anomalies in PM$_{2.5}$ in 2012 relative to the long-term mean, we conclude that the 2012 GEOS-Chem simulation provides a representative snapshot of global air quality.

To validate the 2012 PM$_{2.5}$ results from GEOS-Chem, we rely on archived PM$_{2.5}$ concentrations from the World Health Organization database (WHO). We find that GEOS-Chem captures the observed annual mean PM$_{2.5}$ concentrations with a correlation of 0.70, mean absolute error of 3.4 µg m$^{-3}$, and normalized mean bias of 27% (Figure S3). Our high bias in the US (where most North American WHO data are located) is opposite to the low bias estimated by Ford and Heald (2016) in urban (-25%) and rural (-6%) areas; such biases may be due to differences in US emission inventories for both gas-phase aerosol precursors and primary particles (Xing et al., 2015). A caveat in our comparison is that most observations (95%) in the WHO database with at least 75% temporal coverage in 2012 are in North America and Europe. We add to Figure S3 the 2012 observations from the US embassy in Shanghai (those for Beijing are already in the WHO dataset), and national monitoring sites embassies in Delhi (Cusworth et al., 2018), and the Highveld region in South Africa (South African Air Quality Information System; data obtained by request from the South African Weather Service in July 2018). Over the European domain in Figure S1, we find that GEOS-Chem yields a correlation of 0.60, mean absolute error of 5.2 µg m$^{-3}$ and a normalized mean bias of 33% in surface PM$_{2.5}$; over the North American domain in Figure S1, these values are 0.52, 1.8 µg m$^{-3}$ and 20% (Figure S4). Taken together, these validation statistics are similar to those reported by other studies examining surface PM$_{2.5}$ in global models (e.g., Shindell et al. (2018)) and regional models (e.g., Xing et al. (2015)).
Table S1. GEOS-Chem anthropogenic emissions. All emissions are scaled to 2012 conditions.

<table>
<thead>
<tr>
<th>Region</th>
<th>Inventory name</th>
<th>Species</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Global</td>
<td>EDGAR v4.2(^a),(^b),(^c)</td>
<td>NO, CO, SO(_2), sulfate, ammonia</td>
<td>Olivier and Berdowski (2001)</td>
</tr>
<tr>
<td>Global</td>
<td>RETRO(^a),(^c)</td>
<td>Non-methane VOCs</td>
<td>Schultz et al. (2007)</td>
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<tr>
<td>Global</td>
<td>---</td>
<td>Ethane</td>
<td>Xiao et al. (2008)</td>
</tr>
<tr>
<td>Global</td>
<td>GEIA</td>
<td>Biofuel ammonia</td>
<td><a href="http://www.geiacenter.org">www.geiacenter.org</a></td>
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<tr>
<td>Global</td>
<td>BOND(^a),(^c)</td>
<td>Carbonaceous particles</td>
<td>Bond et al. (2007)</td>
</tr>
<tr>
<td>Global</td>
<td>AEIC aircraft v2.0</td>
<td>NO, CO, etc.</td>
<td>Stettler et al. (2011)</td>
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<tr>
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<td>SO(_2)</td>
<td>Eyring et al. (2005)</td>
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<td>Global</td>
<td>ICOADS ship</td>
<td>CO</td>
<td>C. Wang et al. (2008)</td>
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<tr>
<td>Global</td>
<td>PARANOX ship</td>
<td>NO</td>
<td>Vinken et al. (2011)</td>
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<td>United States</td>
<td>NEI 2011(^a),(^b),(^c)</td>
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<td>US EPA, www3.epa.gov/airtrends</td>
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<tr>
<td>Europe</td>
<td>EMEP(^b),(^c)</td>
<td>Many species</td>
<td><a href="http://www.emep.int">www.emep.int</a></td>
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<tr>
<td>Asia</td>
<td>MIX(^c)</td>
<td>Many species</td>
<td>M. Li et al. (2017), Venkataraman et al. (2018), X. Li et al. (2018)</td>
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<td>Africa</td>
<td>DICE(^c),(^d)</td>
<td>Many species</td>
<td>Marais and Wiedinmyer (2016)</td>
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<tr>
<td>Africa</td>
<td>---</td>
<td>Open waste burning species</td>
<td>Wiedinmyer et al. (2014)</td>
</tr>
</tbody>
</table>

\(^a\) Includes biofuel sources

\(^b\) Includes ship emissions

\(^c\) Includes land-based transport emissions

\(^d\) Includes only diffuse and inefficient sources of anthropogenic emissions – residential fuelwood, diesel and petrol generators, ad-hoc oil refining, gas flares, kerosene use, charcoal production and use, road transport (including motorcycles). For emissions from formal industry and powerplants, we use the global inventories.
Figure S1. Uncertainty in 2012 PM$_{2.5}$ due to interannual variability. Interannual variability is estimated as the relative standard deviation of the Dalhousie satellite-derived PM$_{2.5}$ product (van Donkelaar et al., 2016) for 2008-2016 at 0.1°×0.1°. Values inset are the domain mean relative standard deviations for North America, South America, Western Europe (including portions of North Africa and the Middle East), Africa (including a portion of the Middle East), Southeast Asia, and Australia.
Figure S2. Representativeness of PM$_{2.5}$ in 2012, calculated as the absolute difference in 2012 and 2008-2016 mean PM$_{2.5}$ from Dalhousie (van Donkelaar et al., 2016) at 0.1°×0.1°. Values inset are domain mean anomalies for North America, South America, Western Europe (including portions of North Africa and the Middle East), Africa (including a portion of the Middle East), Southeast Asia, and Australia.
**Figure S3.** Evaluation of GEOS-Chem PM$_{2.5}$. Points are annual mean PM$_{2.5}$ for coincident $0.5^\circ \times 0.667^\circ$ grid squares with at least 75% temporal coverage in the observations. GEOS-Chem PM$_{2.5}$ is estimated at 50% relative humidity (RH) in Europe and 35% RH everywhere else, following standard protocols in measurements of PM$_{2.5}$. Reduced major axis (RMA) regression line (solid black line) and statistics, and the Pearson’s correlation coefficient for all coincident grid squares are given inset. Points in red are in Europe and in blue are in North America. Only 7 out of 957 points exceed the range shown.
Figure S4. Comparison of the spatial distribution of observed and modeled PM$_{2.5}$ in Europe and North America. Data are on a uniform $0.5^\circ\times0.667^\circ$ grid. Only observations with at least 75% temporal coverage are used. PM$_{2.5}$ are obtained at 50% RH in Europe and 35% RH in North America. Data for the two domains are plotted on different scales. Mean PM$_{2.5}$ for coincident grid squares is given inset.
**PM$_{2.5}$ mortality concentration –response model**

We estimated the number of premature deaths attributable to fossil-fuel related PM$_{2.5}$ using a health impact function. To estimate the excess number of deaths associated with PM$_{2.5}$ exposure one requires estimates of exposure, the size of the population exposed, the mortality rate for that population, and the fraction of total deaths attributable to that exposure (AF%).

Recent meta-analysis of the association between long-term PM$_{2.5}$ and mortality (Vodonos et al., 2018) applied a multivariate linear random effects meta-analysis and meta-regression models that polled 135 hazard ratio estimates derived from 53 studies examined long-term PM$_{2.5}$ and mortality. This meta-analysis provided an evidence of a nonlinear association where the exposure-mortality slopes decreased at higher concentrations (**Figure S5**). For example, each 1 µg m$^{-3}$ increase in PM$_{2.5}$ was associated with a 1.29% increase in all-age all-cause mortality (95%CI 1.09-1.50) at a mean exposure of 10 µg m$^{-3}$, which decreased to 0.94% (95%CI 0.76-1.12) at a mean exposure of 20 µg m$^{-3}$, to 0.81% (95%CI 0.52-1.12) at 30 µg m$^{-3}$ and to 0.79% (95%CI 0.40-1.13) at 40 µg/m$^3$.

Hence, for examining a reduction of PM$_{2.5}$ levels from 15 to 10 µg/m$^3$, we calculated the mean slope as area under the curve between 0.014 and 0.011= 0.0125. A reduction of PM$_{2.5}$ levels from 30 to 20 µg/m$^3$, the mean slope was calculated as area under the curve between 0.009 and 0.008 = 0.00814.

Mean value of estimates of mortality ($\bar{\beta}$) for each grid cell was calculated as area under the curve for the concentration-specific $\beta$ in each grid cell from the low PM$_{2.5}$ scenario (without fossil fuel emissions) to the high PM$_{2.5}$ scenario (with all emissions, including fossil fuel) following the form shown in Equation

$$\bar{\beta}(PM_{2.5}) = \int_{PM_{2.5} \text{ no fossil fuel}}^{PM_{2.5} \text{ all emissions}} \beta(PM_{2.5})$$
**Figure S5.** Estimates for long-term PM\textsubscript{2.5} mortality dose-response, drawn from the meta-analysis of long-term association between PM\textsubscript{2.5} and mortality (Vodonos et al., 2018).
Table S2. Extended data. Global regions, number of deaths, attributable fraction (%) for the population above 14 years old attributable to fine particulate matter (PM$_{2.5}$) exposure in 2012

<table>
<thead>
<tr>
<th>Country name</th>
<th>Total Deaths &gt;14 years old</th>
<th>Mean population weighted annual PM$_{2.5}$ (μg m$^{-3}$)</th>
<th>Attributable deaths$^a$</th>
<th>Mean attributable fraction (%)$^b$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>With all emission sources</td>
<td>Without fossil fuel</td>
<td>Estimated fossil fuel PM$_{2.5}$</td>
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<td>North America</td>
<td></td>
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<td>0.3</td>
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<td>3.3</td>
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<td>4.7</td>
<td>4.4</td>
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<td>Country name</td>
<td>Total Deaths &gt;14 years old</td>
<td>Mean population weighted annual PM$_{2.5}$ (µg m$^{-3}$)</td>
<td>Attributable deaths$^a$</td>
<td>Mean attributable fraction (%)$^b$</td>
</tr>
<tr>
<td>------------------------------------</td>
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<td>--------------------------------------------------------</td>
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<tr>
<td></td>
<td></td>
<td>With all emission sources</td>
<td>Without fossil fuel PM$_{2.5}$</td>
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<td>Venezuela</td>
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</table>

$^a$ Estimated deaths attributable to PM$_{2.5}$ exposure.

$^b$ Mean attributable fraction of all deaths attributable to PM$_{2.5}$.
<table>
<thead>
<tr>
<th>Country name</th>
<th>Total Deaths &gt;14 years old</th>
<th>Mean population weighted annual PM$_{2.5}$ (µg m$^{-3}$)</th>
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<td></td>
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<td>With all emission sources</td>
<td>Without fossil fuel PM$_{2.5}$</td>
<td>Estimated fossil fuel PM$_{2.5}$</td>
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<td><strong>Europe</strong></td>
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<tr>
<td>Albania</td>
<td>20,072</td>
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<td>Mean attributable fraction (%)$^b$</td>
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<td>1.2                                                      0.8</td>
<td>0.4</td>
<td>1</td>
</tr>
<tr>
<td>Ontario</td>
<td>90,996</td>
<td>15                                                       1.6</td>
<td>13.4</td>
<td>15,728</td>
</tr>
<tr>
<td>Prince Edward Island</td>
<td>1,269</td>
<td>4.3                                                      1.4</td>
<td>2.9</td>
<td>61</td>
</tr>
<tr>
<td>Quebec</td>
<td>66,494</td>
<td>13.9                                                     1.6</td>
<td>12.3</td>
<td>10,645</td>
</tr>
<tr>
<td>Saskatchewan</td>
<td>8,515</td>
<td>7.5                                                      2.4</td>
<td>5.2</td>
<td>678</td>
</tr>
<tr>
<td>Yukon Territory</td>
<td>193</td>
<td>1.1                                                      0.9</td>
<td>0.3</td>
<td>1</td>
</tr>
</tbody>
</table>

$^a$ Annual number of deaths attributed to long term exposure to PM$_{2.5}$ generated by fossil fuel combustion.

$^b$ Mean proportion of deaths attributed to long term exposure to fossil-fuel related PM$_{2.5}$.

$^c$ Includes South Sudan

$^d$ Estimates derived after applying a 43.7% reduction to PM$_{2.5}$ from all sources for China
References


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