Global Estimates of Exposure to Fine Particulate Matter Concentrations from Satellite-based Aerosol Optical Depth

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Global Estimates of Exposure to Fine Particulate Matter Concentrations from Satellite-based Aerosol Optical Depth

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** primary location of this work
Running Title: Global Estimates of PM$_{2.5}$ Exposure from Satellite AOD

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List of Abbreviations:

AOD - Aerosol Optical Depth

AQS - Air Quality Guideline

CALIPSO - Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation satellite

CTM - Chemical Transport Model

MISR - Multiangle Imaging Spectroradiometer

MODIS - Moderate Resolution Imaging Spectroradiometer

PM$_{2.5}$ - Fine particulate matter with diameter less than 2.5 µm.

RH - Relative Humidity (%)

SEDAC - Socioeconomic Data and Applications Center

IT - Interim Target

WHO - World Health Organisation
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Introduction

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Background: Epidemiologic and health impact studies of fine particulate matter (PM$_{2.5}$) are limited by the lack of monitoring data, especially in developing countries. Satellite-observations offer valuable global information about PM$_{2.5}$ concentrations.

Methods: Global ground-level PM$_{2.5}$ concentrations were mapped using total column aerosol optical depth (AOD) from the MODIS and MISR satellite instruments and coincident aerosol vertical profiles from the GEOS-Chem global chemical transport model.

Results: Global estimates of long-term average (2001-2006) PM$_{2.5}$ at ~10 km x 10 km resolution indicates that the World Health Organization Air Quality PM$_{2.5}$ Interim Target-1 (35 µg/m$^3$ annual average) is exceeded over central and eastern Asia for 35% and 45% of the population, respectively. While concentrations have decreased in Western Europe and Eastern North America between 2001 and 2006, Eastern Asia shows a large upward trend, with PM$_{2.5}$ values in the top 10$^{th}$ percentile increasing by 6.5 µg/m$^3$ each year. The highest mean PM$_{2.5}$ concentrations (> 80 µg/m$^3$) are over Eastern China.

Evaluation of the satellite-derived estimate with ground-based in-situ measurements indicates significant spatial agreement with North American measurements ($r = 0.78$, slope = 1.02, n = 1083) and with non-coincident measurements elsewhere ($r = 0.83$, slope = 0.86, n = 244). The one-sigma uncertainty in the satellite-derived PM$_{2.5}$ is 25%, inferred from the AOD retrieval and aerosol vertical profiles errors and sampling. The global population-weighted mean uncertainty is 6.7 µg/m$^3$.

Conclusions: Satellite-derived total-column AOD, when combined with an aerosol transport model, provides estimates of global PM$_{2.5}$ exposure.
Chronic exposure to airborne fine particulate matter with diameter less than 2.5 µm (PM$_{2.5}$) is associated with adverse human health impacts including morbidity and mortality (e.g., Dockery et al. 1993; McDonnell et al. 2000; 2002; Pope et al. 2009). Several national environmental agencies in North America and Europe monitor PM$_{2.5}$ concentrations at numerous sites throughout their jurisdictions, but even these relatively dense networks provide only limited geographical coverage. Few long-term measurement sites exist elsewhere in the world, particularly in rapidly developing countries where concentrations and estimated health impacts are greatest (Cohen et al. 2003). Point measurements collected at monitoring sites are not necessarily representative of regional concentration, and regional variability is difficult to assess from point measurements alone. Application of satellite observation to surface air quality has advanced considerably in recent years (Martin 2008; Hoff and Christopher 2009). Global aerosol observations from satellite could substantially improve estimates of population exposure to PM$_{2.5}$.

Since mid-2000, the Moderate Resolution Imaging Spectroradiometer (MODIS) and Multiangle Imaging Spectroradiometer (MISR) instruments onboard the Terra satellite have provided global observations of aerosol optical depth (AOD), a measure of light extinction by aerosol over the atmospheric column above the earth’s surface. Terra’s Sun-synchronous orbit encircles the Earth approximately 15 times each day, with each pass crossing the Equator at approximately 10:30, local time. Observations of AOD from Terra provide daily insight into the global distribution of column-integrated aerosol. However, because these retrievals do not exclusively correspond to near-ground aerosol,
the applicability of AOD to surface air quality depends upon the vertical structure and scattering properties of atmospheric aerosol.

Many studies have investigated the relationship between total-column AOD and surface PM\textsubscript{2.5} measurements. Most have developed simple empirical relationships between these two variables (e.g. Wang and Christopher 2003; Engel-Cox et al. 2004a), while more recent works often using local meteorological conditions to improve agreement (e.g. Liu et al. 2005; Koelemeijer et al. 2006; Kumar et al. 2007) or to filter the data (e.g. Gupta et al. 2006). Some studies have also employed LIDAR instruments to capture the vertical aerosol distribution at specific locations (e.g. Engel-Cox et al. 2006; Schaap et al. 2008). As noted by Schaap et al. (2008), locally derived AOD-PM\textsubscript{2.5} relationships cannot be easily extended to other regions due to variation in meteorology and aerosol composition. Unique, local, time-dependent AOD-PM\textsubscript{2.5} relationships are necessary for AOD to provide global estimates of PM\textsubscript{2.5}. Due to lack of coverage, ground-based measurements of aerosol vertical profiles and properties are insufficient to estimate local AOD-PM\textsubscript{2.5} relationships at the global scale.

Global chemical transport models (CTMs) resolve the atmosphere at a resolution of hundreds of kilometres horizontally by hundreds of meters vertically, with a temporal frequency of tens of minutes. Liu et al. (2004) first estimated surface-level PM\textsubscript{2.5} from MISR observations by using CTM output to represent local AOD-PM\textsubscript{2.5} conversion factors over the contiguous United States. Van Donkelaar et al. (2006) extended the approach of Liu et al. (2004) to estimate PM\textsubscript{2.5} from both MODIS and MISR observations and investigated the factors affecting the agreement between AOD and surface-level PM\textsubscript{2.5}. Statistical models have also been used to relate AOD to PM\textsubscript{2.5}, using
in addition to model-derived vertical distribution, MISR spherical vs. non-spherical particle property retrievals to separate mineral dust from spherical species that include aerosol pollution (Liu et al. 2007). Some recent work also probed the limitations of using AOD without accounting for vertical distribution or speciation, and concluded that agreement with ground-based monitors based on this approach might depend on factors other than the satellite observations (Paciorek and Liu 2009).

In this paper, we develop a global satellite-based estimate of surface PM$_{2.5}$ at a spatial resolution of 0.1° x 0.1°, or approximately 10 km x 10 km at mid-latitudes. The methods section develops an approach for combining MODIS and MISR AOD into a single, improved estimate of AOD. AOD-PM$_{2.5}$ conversion factors, calculated with a global chemical transport model, are produced and applied to these AOD in the results section. We present a global estimate of PM$_{2.5}$ and validate with ground-based (in-situ) observations. We estimate global population exposure to PM$_{2.5}$ using our satellite-derived product to demonstrate the potential of satellite-estimated PM$_{2.5}$ for global health studies. We then examine sources of error.

Methods

Satellite Observations

The MODIS instrument measures a wide range of spatial and spectral information from its orbit aboard the Terra satellite. The near-daily global coverage from the MODIS AOD retrieval (Levy et al. 2007) is advantageous. The MISR instrument, also aboard Terra, offers smaller spatial and spectral ranges, but views each scene on the Earth from
nine different angles. This additional angular information allows the MISR AOD retrieval (Martonchik et al. 2002; Diner et al. 2005; Martonchik et al. 2009) to reduce some algorithmic assumptions and retrieval bias, as well as obtain some information about microphysical properties, and plume heights in aerosol source regions (Kahn et al. 2007).

We use the MODIS BRDF/Albedo product (MOD43, Collection 5) to distinguish surface types and, in conjunction with ground-based retrievals of AOD, identify regions of high bias in both MODIS and MISR AOD. The supplemental material describes in detail the satellite retrievals, and our bias filtration.

Estimating $PM_{2.5}$ from Aerosol Optical Depth

A local conversion factor ($\eta$) is necessary to estimate ground-level concentrations of $PM_{2.5}$ (in $\mu g/m^3$) from total-column AOD (unitless):

$$PM_{2.5} = \eta \cdot AOD \quad \quad [1]$$

$\eta$ is a function of aerosol size, aerosol type, diurnal variation, relative humidity and the vertical structure of aerosol extinction. Following Liu et al. (2004; 2007) and van Donkelaar et al. (2006), we use a global 3-D chemical transport model (GEOS-Chem; Supplemental Material) to calculate the daily global distribution of $\eta$. The GEOS-Chem model solves for the temporal and spatial evolution of aerosol (sulfate, nitrate, ammonium, carbonaceous, mineral dust and sea salt) and gaseous compounds using
meteorological data sets, emission inventories, and equations that represent the physics and chemistry of atmospheric constituents. We calculate daily values of $\eta$ as the ratio of 24-h ground-level PM$_{2.5}$ for a relative humidity of 35% (U.S. and Canadian surface-measurement gravimetric analysis standard), and at 50% (European surface-measurement standard), to total-column AOD at ambient relative humidity at Terra overpass time.

We compare the original MODIS and MISR total-column AOD with coincident ground-based measurements of daily mean PM$_{2.5}$. Canadian sites are part of the National Air Pollution Surveillance Network, maintained by Environment Canada (http://www.etc.cte.ec.gc.ca/NAPS/index_e.html). U.S. data are from the Interagency Monitoring of Protected Visual Environments (IMPROVE) network (http://vista.cira.colostate.edu/improve/Data/data.htm) and the U.S. Environmental Protection Agency Air Quality System Federal Reference Method sites (http://www.epa.gov/air/data/index.html). Validation of global satellite-derived PM$_{2.5}$ estimates is hindered by the lack of available surface-measurement networks in many parts of the world. We collect annually representative, ground-based PM$_{2.5}$ data from both published and unpublished field measurements outside of North America, as described in the supplemental material.

Results

The top and middle panels of Figure 1 show mean AOD for 2001-2006 over North America from MODIS and MISR. Both datasets exhibit similar AOD values of 0.15-0.25 over the eastern United States, reflecting a combination of anthropogenic and biogenic sources. Several individual cities can be clearly identified in mean MODIS
AOD for the Great Lakes region. A large AOD enhancement over the southwestern United States appears in the MODIS retrievals, but is absent from the MISR retrievals. The bottom panel of Figure 1 shows the mean combined MODIS and MISR AOD over North America. Our combination of these two AOD products removes the biased AOD observed by MODIS over the western United States. The combined product is dominated by MODIS in the east due to finer temporal sampling. MISR dominates in the west due to its accuracy.

Table 1 gives statistics comparing the spatial variation in six-year mean AOD retrievals with measurements of daily 24h average PM$_{2.5}$ sampled on the same days. Both the MODIS and MISR instruments indicate some relationship between retrieved total-column AOD and in-situ PM$_{2.5}$, with spatial correlation coefficients of 0.40 for MODIS and 0.54 for MISR. A simple average of the daily AOD from both instruments yields a correlation of 0.45. Combining retrievals from these instruments as described in the methods section increases the correlation to 0.63. Additional information is required to quantitatively estimate PM$_{2.5}$ exposure from AOD as presented below.

Figure 2 shows the annual mean distribution of daily $\eta$ values used to relate satellite-observed total-column AOD to PM$_{2.5}$ at 35% relative humidity. Average values of $\eta$ are typically 20-130 $\mu$g/m$^3$. High values of $\eta$ over regions of large dust concentration (Prospero et al. 2002) reflect in part its low hygroscopicity. Values of $\eta$ are lower for hygroscopic aerosols as their dry volume is significantly smaller than under ambient conditions. Ground-level aerosol sources in industrial regions lead to vertical profiles that peak near ground, and moderate values of $\eta$. Western North America is characterized by low $\eta$, providing additional insight into the poor AOD-PM$_{2.5}$
correlations (Engel-Cox et al. 2004b; Hu 2009) associated with this region, and in agreement with Liu et al. (2007) who found that transported dust aloft affects the western N. America AOD-PM$_{2.5}$ relationship. $\eta$ is related to land types only insomuch as these are typified by particular aerosol types, meteorology and vertical structures. Temporal variation in $\eta$ is also considerable.

The top panel of Figure 3 shows the six-year mean of 24-h average satellite-derived surface PM$_{2.5}$ over North America as calculated from equation 1 at a daily timescale. A large-scale PM$_{2.5}$ enhancement is apparent over the eastern United States and extending into southern Canada. The western and northern parts of the continent are generally characterized by low concentrations, with a few exceptions. Mean PM$_{2.5}$ concentrations over eastern and western North America are 8.6 µg/m$^3$ and 7.3 µg/m$^3$, respectively, although major city centers can exceed annual mean concentrations of 15 µg/m$^3$.

Application of $\eta$ (Figure 2) increased the spatial contrast in Figure 1, reflecting large aerosol sources in the east and aerosols aloft in the north and west.

We evaluate the satellite-derived PM$_{2.5}$ with surface monitors. The bottom right panel of Figure 3 shows the annual mean of 24-h PM$_{2.5}$ measured with the surface monitors and sampled on the same days as the satellite-derived PM$_{2.5}$. Ground-level measurements show similar features to our satellite-derived product. The bottom left panel of Figure 3 compares satellite-derived and ground-level measured PM$_{2.5}$, quantitatively. We find significant spatial correlation between coincidently sampled satellite-derived and ground-based PM$_{2.5}$ across North America ($r = 0.78$, slope = 1.02, bias = -1.07 µg/m$^3$). Many factors contribute to the scatter of points, including significant differences between what
satellite and *in situ* surface sampling represent that do not necessarily indicate errors in either measurement.

Global Estimates of PM$_{2.5}$ Concentrations

Figure 4 presents the six-year mean of our global satellite-derived PM$_{2.5}$. This figure is at 50% RH for comparison with European ground-based measurements. The satellite-derived PM$_{2.5}$ include a small adjustment for discontinuous sampling as described below. PM$_{2.5}$ concentrations over eastern North America and western Europe are typically 10-20 µg/m$^3$, with the exception of the Po Valley in northern Italy, where regional PM$_{2.5}$ concentrations are 25-35 µg/m$^3$. PM$_{2.5}$ concentrations are typically 60-90 µg/m$^3$ over eastern China, with values in excess of 100 µg/m$^3$ for major industrial regions of China. Enhanced SO$_2$ and NO$_2$ columns observed from satellite over the same region of eastern China (Martin et al. 2006; Lee et al. in press), highlight the influence of combustion and other regional aerosol pollution sources on surface air quality. The Indo-Gangetic plain, from New Delhi eastward contains the highest PM$_{2.5}$ concentrations in India, with values of 80-100 µg/m$^3$, especially in winter (e.g. Di Girolamo et al. 2004). Concentrations elsewhere in northern India are typically 35-60 µg/m$^3$. The effects of biomass burning in central Africa and central South America on PM$_{2.5}$ levels are visible. Dust transport in the fine mode is significant (Jones and Christopher 2007), contributing to large-scale PM$_{2.5}$ in the Middle East of approximately 20-50 µg/m$^3$.

Figure 4 also shows locations of ground-based measurements and values outside North America that were used for comparison. Despite increased uncertainty due to temporal sampling differences, significant overall agreement exists ($r = 0.83$, slope = 0.86,
intercept = 1.15 µg/m³, n = 244). Similar agreement is obtained when all sites except
Europe and North America are considered (r = 0.91, slope = 0.84, intercept = -2.52 µg/m³,
n = 84).

The left two columns of Figure 5 overlay contours of population density and surface
elevation onto satellite-derived PM$_{2.5}$ for regions of major anthropogenic sources: eastern
North America, western Europe and eastern Asia. Low-lying regions are often both
heavily populated and highly polluted, as can be seen throughout eastern China and in
the Po Valley of northern Italy. By contrast, high altitude regions, such as the
Appalachian Mountains in eastern North America, are associated with decreased
population and PM$_{2.5}$. Some PM$_{2.5}$ enhancements are associated with urban areas.

We bin satellite-derived PM$_{2.5}$ for each year between 2001 and 2006 onto a 0.5º x 0.5º
grid to investigate trends in these regions, as shown in the right column of Figure 5 and
summarized in Table 2. Both eastern North America and western Europe exhibit a slight
decreasing trend in PM$_{2.5}$. This is consistent with ground-based measurements for the
United States that show an annual mean decrease of 1.3% per year (U.S. Environmental
Protection Agency 2008). The European trend is greatest in parts of Germany and the Po
Valley. Eastern Asia shows a large upward trend, with PM$_{2.5}$ concentrations in the top
10$^{th}$ percentile increasing by 6.5 µg/m³ each year. The sign of these regional trends is
also consistent with trends in emissions of aerosol precursors (Ohara et al. 2007) and in
satellite observations of combustion products (Richter et al. 2005), and in ground-based
measurements of aerosol radiative effects (Wild et al. 2009). Increased oceanic AOD in
coastal regions of Asia from the late 1980s to early 2000 (Mishchenko and Geogdzhayev
2007) suggest the satellite-derived PM$_{2.5}$ trends are part of an overall longer-term trend.
Error Analysis

The dominant sources of error in satellite-derived PM$_{2.5}$ arise from uncertainties in both AOD measurement and vertical structure (van Donkelaar et al. 2006). We evaluate the GEOS-Chem simulation of the aerosol vertical profile using observations from the CALIPSO satellite. The GEOS-Chem simulation generally captures to within 5% the fraction of AOD within the boundary layer (Supplemental Material). We combine errors in regional vertical structure with the residual satellite AOD bias, as evaluated from ground-based AERONET measurements. We estimate the error in satellite-derived PM$_{2.5}$ as the change that occurs when η and AOD are adjusted by their individual errors.

Figure 6 shows the error distribution of coincidently sampled satellite-derived PM$_{2.5}$. Arid regions are typically over-predicted, and populated regions of East Asia under-predicted. We find that one standard deviation of the global error distribution is within ±15% of the satellite-derived value. We test this uncertainty estimate by comparison with coincident PM$_{2.5}$ observations for North America (Figure 2) and find that one standard deviation of the data lies within ±(1 µg/m$^3$ + 15%). The necessary inclusion of a small absolute term suggests that our uncertainty estimate may be underestimated at low PM$_{2.5}$ values, and supports the presence of a small negative bias, as indicated by the line of best fit.

Non-uniform and incomplete sampling by satellites have the potential to create bias in long-term mean observations (Levy et al. 2009; Paciorek and Liu 2009). Here we investigate how non-random sampling of AOD by satellite observations affect the representation of annual mean PM$_{2.5}$. Figure 7 shows the percent difference in between a
GEOS-Chem simulation of PM$_{2.5}$ sampled coincidently with daily satellite-derived PM$_{2.5}$ versus a complete annual mean of the simulated values. Most regions exhibit a sampling-induced uncertainty (one-sigma) within $\pm$20% of simulated PM$_{2.5}$. Sampling of satellite-derived PM$_{2.5}$ causes larger errors in regions influenced by seasonal biomass burning or mineral dust. Validation of these errors is inhibited by the lack of in-situ measurements in the regions most significantly affected by intermittent sampling. Statistical comparison over the United States and Canada of non-coincident satellite-derived and in-situ PM$_{2.5}$ decreases the agreement relative to a coincident comparison (non-coincident: slope = 1.08, $r = 0.71$ versus coincident: slope = 1.02, $r = 0.78$), supporting the need for sampling error correction. Uncertainties derived from both the PM$_{2.5}$ estimate and sampling can vary substantially on the regional scale. Testing the combined uncertainty of $\pm$25% from both sources reveals that approximately one standard deviation of the North American data falls within this overall error envelope. Globally, the population-weighted mean uncertainty in satellite-derived PM$_{2.5}$ is 6.7 $\mu$g/m$^3$.

**Global PM$_{2.5}$ Exposure**

*Pope et al.* [2009] estimate that a long-term PM$_{2.5}$ exposure decrease of 10 $\mu$g/m$^3$ increases life expectancy by 0.61 $\pm$ 0.30 years for the United States. We estimate global PM$_{2.5}$ exposure at a spatial resolution of 0.1° using our satellite-derived values for 2001-2006 and the Gridded Population of the World (Tobler et al. 1997) data for 2005 from the Socioeconomic Data and Applications Center (SEDAC; GPW v3; http://sedac.ciesin.columbia.edu/). Figure 8 shows the global distribution of exposure, and regional distributions over North America, Europe and Asia. Table 3 summarizes
these results. All regions exhibit non-linear relationships between population and exposure. Eastern and central Asia have the highest levels of PM$_{2.5}$ exposure, with 38-49% of regional population exceeding the WHO Air Quality Interim Target-1 (WHO 2005) of 35 µg/m$^3$. According to the WHO Guidelines, concentrations at this level and higher are associated with approximately a 15% increased mortality risk, relative to the Air Quality Guideline of 10 µg/m$^3$. Globally 80% of the population lives in regions that exceed the Air Quality Guideline. These exposure estimates should be of considerable value for assessing the chronic health impacts of air pollution, especially in regions with sparse ground-based monitoring.

Discussion

A major challenge for global epidemiological studies and assessments of air pollution health impacts is the lack of representative exposure estimates (Cohen et al. 2003). While extensive ground-based monitoring networks exist in some parts of the world, major portions of the globe are not covered. The situation is especially acute in developing countries with large populations and high pollution levels, but limited monitoring with traditional ground-based sampling techniques. While measurements form ground monitors are currently the “gold-standard” for epidemiological studies, that are not only sparse, but are also potentially unrepresentative of the larger region depending on the influence of nearby localized emission sources (e.g. major roadways or industrial emitters). Satellite observations offer area-integrated values with frequent global coverage, providing valuable additional information for global health studies.
In this work, we produced a satellite-derived climatology of PM$_{2.5}$ concentrations and applied it to estimate the global distribution of long-term average exposure. These estimates should facilitate studies of chronic exposure to particulate matter, similar to those already conducted in Europe and North America (e.g. Dockery et al. 1993; Pope et al. 2002; Beelen et al. 2008; Pope et al. 2009), in regions of the world currently without extensive ground-based monitoring networks. Although there are a growing number of studies of the impacts of short-term exposure to particulate matter have in previously under-represented regions of the world (e.g. Gouveia et al. 2008; Wong et al. 2008; Romieu et al. 2009), studies of long-term exposure also incorporate impacts related to chronic disease and therefore provide a more comprehensive estimate of overall health effects (Kunzli et al. 2001). Our estimates suggest that 80% of the global population resides in locations where ambient concentrations exceed the WHO Air Quality Guideline of 10 µg/m$^3$. Application of the satellite-derived PM$_{2.5}$ dataset also identifies global regions and areas of specific concern; nearly half (49%) of the eastern Asian population lives in regions that exceed the WHO Air Quality Interim Target 3 of 35 µg/m$^3$, and are therefore at increased risk from air pollution-related health impacts. These results highlight the potential of satellite aerosol observations to contribute to chronic effects studies at regional and global scales.

Several notable developments over previous work were included in these estimates. Combining AOD from two satellite instruments (MODIS and MISR) improved the correlation of AOD versus ground-based PM$_{2.5}$ measurements. Extending the satellite data over six years (2001-2006) reduces sampling issues. The unprecedented global spatial resolution of 0.1° x 0.1° retains important details in the exposure estimate. A
chemical transport model (GEOS-Chem) was applied to account for aerosol vertical
distribution, a key factor affecting the relationship between satellite-retrieved, total
column AOD and near-surface PM$_{2.5}$. We found significant spatial agreement between
mean coincident satellite-derived and ground-based PM$_{2.5}$ for North America (slope =
1.02, r = 0.78, n = 1083), as well as evidence of global agreement with non-coincident
measurements from published and unpublished data (slope = 0.86, r = 0.83, n = 244).
Notably, this level of agreement with ground-based PM$_{2.5}$ is significantly better than that
obtained using a global chemical transport model (GEOS-Chem) without satellite data
(Supplemental Material). Detailed spatial structure in the satellite-derived PM$_{2.5}$
concentrations reflects the influences of emissions and topography.

We assessed the uncertainty in the satellite-derived product through comparison with
independent observations and error propagation. We estimated our coincident satellite-
derived PM$_{2.5}$ to be accurate at the one-sigma level to within ±15% of the satellite-
derived value using the relative AOD vertical structure measured by the CALIPSO
satellite and the total column AOD from ground-based measurements (AERONET). We
found evidence that the effect of non-uniform satellite sampling typically biases annual
mean satellite-derived PM$_{2.5}$ by less than ±20% of the satellite-derived value. Larger
effects are expected over regions influenced by substantial seasonal variation in biomass
burning, dust transport and monsoonal activity, or for individual, severe pollution events.
Globally, the overall combined PM$_{2.5}$ uncertainty of ±25% indicates a mean uncertainty
in exposure of 6.7 µg/m$^3$.

Additional refinements to the analysis process could continue to reduce error in the
satellite-derived PM$_{2.5}$ chronic exposure estimates presented here. Further improvements
to the AOD retrieval (e.g. Drury et al. 2008) would improve accuracy and reduce sampling bias by reducing data rejection. Simulating the AOD-PM$_{2.5}$ conversion factors at finer spatial resolution would better capture their variability, which is especially important in regions of sharp topographic or emissions gradients. Further development of aerosol speciation capability (e.g. Liu et al. 2007) and satellite-based estimates of additional species, such as NO$_2$ (Lamsal et al. 2008) would be valuable to more specifically estimate pollutant exposure. We encourage deployment of additional ground-based monitors, and undertaking of cohort studies and impact assessment of acute changes in exposure, in the regions of high chronic exposure identified by our analysis.
References


Jones AJ, Christopher SA. 2007. Modis derived fine mode fraction characteristics of marine, dust, and anthropogenic aerosols over the ocean, constrained by gocart, mopitt, and toms. Journal of geophysical research 112(D22204).


Table 1: Comparison of coincidently sampled six-year mean measurements\textsuperscript{a} of daily 24h average PM\textsubscript{2.5} with AOD and satellite-derived PM\textsubscript{2.5}.

<table>
<thead>
<tr>
<th></th>
<th>slope\textsuperscript{b}</th>
<th>intercept</th>
<th>r</th>
<th>n</th>
</tr>
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<tbody>
<tr>
<td>MODIS AOD</td>
<td>0.020</td>
<td>0.10</td>
<td>0.40</td>
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<td>MISR AOD</td>
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<td>0.09</td>
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<td>0.10</td>
<td>0.45</td>
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<tr>
<td>Combined AOD</td>
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<td>0.63</td>
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<tr>
<td>Satellite-derived PM\textsubscript{2.5}</td>
<td>1.016</td>
<td>-1.07</td>
<td>0.78</td>
<td>1083</td>
</tr>
</tbody>
</table>

\textsuperscript{a} A minimum of 5 measurements, 3 years out of 6 are required for each point.
\textsuperscript{b} calculated with reduced major-axis linear regression (Hirsh and Golroy 1984)
Table 2: Annual trends in satellite-derived PM$_{2.5}$ of major industrialized regions from 2001-2006. Spatial details are presented in Figure 5.

<table>
<thead>
<tr>
<th>Region</th>
<th>Percent with significant trend [%]</th>
<th>Overall trend [µg/m$^3$]</th>
<th>Trend in 10$^{th}$ percentile [µg/m$^3$]</th>
</tr>
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<tbody>
<tr>
<td>E. North America</td>
<td>39.1</td>
<td>-0.7</td>
<td>-1.0</td>
</tr>
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<td>W. Europe</td>
<td>60.7</td>
<td>-0.9</td>
<td>-1.4</td>
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<tr>
<td>E. Asia</td>
<td>41.8</td>
<td>3.1</td>
<td>6.5</td>
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Table 3: Regional population in excess of WHO Air Quality Guideline (AQG) and Interim Targets (IT)\textsuperscript{a}. Regions are defined in Figure 6.

<table>
<thead>
<tr>
<th>Region</th>
<th>Population (million people)</th>
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</thead>
<tbody>
<tr>
<td></td>
<td>Total</td>
</tr>
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<td>World</td>
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\textsuperscript{a} (WHO 2005)
Figure Legends

Figure 1: Mean aerosol optical depth (AOD) over 2001-2006 from the MODIS and MISR satellite instruments. The bottom panel indicates data from a combined product developed here. White space denotes water or fewer than 5 measurements per year for at least 3 of 6 years.

Figure 2: Annual mean $\eta$ (ratio of PM$_{2.5}$ to AOD) for 35% relative humidity. White space indicates water.

Figure 3: Satellite-derived PM$_{2.5}$ and comparison with surface measurements. The top panel shows overall mean satellite-derived PM$_{2.5}$ between 2001-2006. White space denotes water or fewer than 5 AOD measurements per year for at least 3 of 6 years. The bottom right panel shows positions and mean values of coincidently measured surface sites. The bottom left panel compares average coincident values of both measured and satellite-estimated PM$_{2.5}$ in $\mu$g/m$^3$. The solid black line denotes unity. Thin dotted lines show measured uncertainty of $\pm(1 \, \mu$g/m$^3 + 15\%)$. The line of best fit (Hirsh and Golroy 1984) is dashed.

Figure 4: Global satellite-derived PM$_{2.5}$ averaged over 2001-2006. White space indicates water or locations containing less than 5 measurements per year, for 3 of 6 years. Satellite-derived PM$_{2.5}$ is adjusted to 50% relative humidity. Circles correspond to values and locations of comparison sites outside Canada and the United States. The black rectangle outlines European sites.

Figure 5: Regional satellite-derived PM$_{2.5}$ sources and trends. Left and center columns show mean satellite-derived PM$_{2.5}$ for 2001-2006 at locations containing at least 5 measurements per year, for 3 years within 2001-2006. Contours denote population density in the left column and surface elevation in the center column. The color scale varies by region. The right column shows PM$_{2.5}$ trends from 2001-2006, with white space being statistically insignificant ($r < 0.62$) or water. Crosses indicate city centers in all three columns. Population density data are based upon 0.25º x 0.25º 2005 Gridded Population of the World (Tobler et al. 1997) data from the Socioeconomic Data and Applications Center (SEDAC; GPW v3; http://sedac.ciesin.columbia.edu/). Altitude data are from the United States Geological Survey.

Figure 6: Estimate of the satellite-derived PM$_{2.5}$ bias, defined as (satellite-derived PM$_{2.5}$ - truth) / truth. Boxed areas define the regions used in Figure 8.

Figure 7: Estimated sampling-induced uncertainty. White space denotes regions where the simulation, sampled coincidently with satellite-estimated PM$_{2.5}$, shows negligible change relative to the annual mean or lack of data.

Figure 8: Cumulative distribution of regional, annual mean exposure estimated from satellite-derived PM$_{2.5}$ at a resolution of 0.1º x 0.1º for 2001-2006. The top axis identifies WHO Air Quality Guidelines (AQG) and Interim Targets (IT-#) associated with each exposure level. Regions are defined in Figure 6.
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Figure 2: Annual mean $\eta$ (ratio of PM2.5 to AOD) for 35% relative humidity. White space indicates water.

152x76mm (300 x 300 DPI)
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Figure 6: Estimate of the satellite-derived PM2.5 bias, defined as \((\text{satellite-derived PM2.5} - \text{truth}) / \text{truth}\). Boxed areas define the regions used in Figure 8.
203x101mm (300 x 300 DPI)
Figure 7: Estimated sampling-induced uncertainty. White space denotes regions where the simulation, sampled coincidently with satellite-estimated PM2.5, shows negligible change relative to the annual mean or lack of data.
Figure 8: Cumulative distribution of regional, annual mean exposure estimated from satellite-derived PM2.5 at a resolution of 0.1° x 0.1° for 2001-2006. The top axis identifies WHO Air Quality Guidelines (AQS) and Interim Targets (IT-#) associated with each exposure level. Regions are defined in Figure 6.
Supplemental Material: Global Estimates of Exposure to Fine Particulate Matter

Concentrations from Satellite-based Aerosol Optical Depth

Collection of Global Ground-based PM$_{2.5}$ measurements

Satellite-derived and simulated global PM$_{2.5}$ concentrations require validation against surface measurements. We combine values from numerous sources for the purpose of comparison. We use European data from a combination of the European Monitoring and Evaluation Programme (EMEP; http://www.emep.int/) and the European Air quality database (AIRBASE; http://air-climate.eionet.europa.eu/databases/airbase/). Australian data were collected from the Environment Protection and Heritage Council (http://www.ephc.gov.au/). New Zealand data were collected from the New Zealand Ministry for the Environment website (http://www.mfe.govt.nz/). Mexican data are from the ESCALA project (Gouveia et al. 2008; Romieu et al. 2009). Columbian data were provided by Victor Miranda and Isabelle Romieu and from the Instituto de Hidrologia Meteorologia y Estudios Ambientales (www.ideal.gov.co). Some Brazilian data for Sao Paulo are from the secretary of State for the Environment, Sao Paulo (http://www.cetesb.sp.gov.br/). Chilean data were provided by CENMA, the Chilean National Environment Center (http://www.cenma.cl/). Additional sources are described in Table S-1. We exclude sites from all sources that are suspected to be spatially or temporally biased.

We combine measurements onto the same 0.1° x 0.1° grid as the satellite dataset. We average colocated studies/sites, weighted by the product of their temporal range (years)
and number of monitors (to a maximum of 5), such that long-term, multi-monitor studies have greater influence on final comparison values. Any surface PM$_{2.5}$ grid cell with an overall weight of less than 1 monitor-year is considered unrepresentative and is not used for evaluation of satellite-derived PM$_{2.5}$.

*Description of the GEOS-Chem model*

We use v8-01-04 of the GEOS-Chem chemical transport model (http://acmg.seas.harvard.edu/geos/index.html). The GEOS-Chem model is driven by assimilated meteorology from the Goddard Earth Observing System (GEOS-4) at the NASA Global Modeling Assimilation Office (GMAO). Our simulation is run at $2^\circ \times 2.5^\circ$ with 42 vertical levels ranging between the surface and approximately 80 km. The thickness of the lower layer is approximately 100 meters.

The GEOS-Chem aerosol simulation includes the sulfate-nitrate-ammonium system (World Bank 2004; Park et al. 2006), primary (Park et al. 2003) and secondary (Liao et al. 2007) carbonaceous aerosols, mineral dust (Fairlie et al. 2007) and sea-salt (Alexander et al. 2005). Formation of sulfate and nitrate (Park et al. 2004), heterogeneous chemistry (Jacob 2000) and photolysis rates (Martin et al. 2003) are all coupled with oxidant simulation. Dry and wet deposition are described in Liu et al. (2001), and include both washout and rainout. The emission inventory has been recently updated to 2005, following van Donkelaar et al. (2008b). We use the eight day Global Fire Emission Database version 2 (GFEDv2) biomass burning emissions (van der Werf et al. 2006), as implemented by Nassar et al. (2009).
The GEOS-Chem aerosol simulation has been extensively evaluated with ground-based measurements (e.g. Park et al. 2006; Fairlie et al. 2007; Pye et al. in press) and aircraft measurements (e.g. Heald et al. 2005; van Donkelaar et al. 2008a; Dunlea et al. 2009).

**Description of Satellite Retrievals**

The MODIS instrument provides near-daily global AOD coverage in the absence of clouds. The MODIS AOD retrieval algorithm over land (Levy et al. 2007) applies three spectral bands at 0.47 µm, 0.66 µm and 2.1 µm plus those used for cloud masking, and requires that surface-reflected radiation makes little contribution to total radiation leaving the top of the atmosphere. Dark surfaces are first detected using the infrared (2.1 µm) spectral band, where atmospheric absorption and scattering from aerosols is generally weak. Surface reflection at visible wavelengths (0.47 µm and 0.66 µm) is then estimated through specified relationships with the 2.1 µm reflectivity. Pre-computed lookup tables (LUT) that combine likely aerosol scenarios with surface reflectivities are then matched with top-of-atmosphere observations to determine AOD values representing 10 km x 10 km retrieval regions. Quality assured collection (version 5 MODIS AOD over land has been validated such that at least two-thirds of retrievals are within ±(0.05 + 15%) using AERONET measurements of AOD (Levy et al. 2009; Remer et al. 2008). The ratio of two spectral bands is used estimate the contribution of non-dust (fine) aerosol to total AOD, but this product is highly uncertain (Remer et al. 2005), especially over land, where it is considered an algorithm diagnostic rather than a retrieval quantity (Anderson et al. 2005; Levy et al. 2009).
The MODIS BRDF/Albedo product (MOD43 V5, Lucht et al. 2000) estimates 16-day average land surface albedo through an algorithm that is separate from the surface reflectivity estimate used by the MODIS AOD retrieval. Albedo, the hemispheric integration of directional surface reflectance, is separated into black-sky and white-sky albedo, where these refer to the albedo under purely direct and diffuse conditions, respectively. The true albedo varies between these two extremes.

The MISR instrument observes radiation leaving the top of the atmosphere in four spectral bands (0.446, 0.558, 0.672 and 0.866 µm), each at nine viewing angles (±70.5°, ±60.0°, ±45.6°, ±25.1° and nadir). MISR takes 9 days for complete global coverage at the equator, and two days near the poles, in the absence of clouds. The MISR AOD retrieval algorithm (Martonchik et al. 2002; Diner et al. 2005; Martonchik et al. 2009) uses same-scene, multi-angle, multi-spectral observations to infer AOD and aerosol microphysical property information over 18 km x 18 km retrieval regions, assuming only approximate spectral invariance of the surface angular reflectance, via pre-calculated LUTs. MISR AOD has been validated such that two-thirds of retrievals fall within the maximum of ±(0.05 or 20%) of ground truth observations (Kahn et al. 2005). The MISR aerosol product also provides estimates of AOD contribution according to aerosol size, dividing AOD into the fraction of particles of radius < 0.35 µm, between 0.35-0.7 µm and > 0.7 µm. The aerosol-size retrieval is most reliable when AOD is greater than 0.2 (Kahn et al. 2009).

We explored using satellite retrievals of aerosol fine mode fraction (FMF) in lieu of the GEOS-Chem simulation of this quantity in the calculation of η, but found that simulated FMF was more accurate for our application due to retrieval uncertainties, temporal
coverage and consistency of fine mode definition. We determine FMF from the GEOS-Chem simulation of fine AOD (sulfate, organic carbon, black carbon, and fine dust and fine sea salt) and coarse AOD (coarse dust and coarse sea salt).

Combining MODIS and MISR observations

Here we describe our approach to combine AOD retrievals from both MODIS and MISR. We translate daily AOD measurements between Jan. 1 2001 and Dec. 31 2006 from MODIS level 2, version 5, best quality and MISR level 2 (F09_0017-F11_0021, best estimate) onto a global 0.1º x 0.1º grid. MODIS AOD retrievals exhibit a high bias over deserts and coastal sites due to surface brightness and subpixel water contamination (Abdou et al. 2005) partially explaining the poor agreement between MODIS AOD and surface PM$_{2.5}$ observed the western United States (e.g. Engel-Cox et al. 2004; Liu et al. 2007; Hu 2009). Systematic regional differences between MODIS and MISR AOD are also found over north-central Africa, northern India and Bangladesh, and the Patagonia Desert region of South America (Kahn et al. 2009).

We use the MODIS BRDF/Albedo product to distinguish surface types and identify regional error in AOD retrieval. Two ratios of six-year monthly mean black-sky albedo (0.47 µm / 0.66 µm and 0.66 µm / 2.1 µm) are used to divide the Earth’s surface into nine albedo-based domains, as defined by the combinations of each ratio being < 0.4, 0.4 - 0.6, and > 0.6. Four surface types dominate, as shown for July in the top panel of Figure S-1. MODIS and MISR AOD are then compared against ground-based retrievals of AOD from the Aerosol Robotic Network (AERONET) (Holben et al. 1998) to calculate an average monthly bias for each instrument within each domain. Local AERONET
comparisons are combined according to surface type. We reject all satellite AOD
retrievals with a local estimated monthly bias in excess of the maximum of ±(0.1 or 20%).
Data from regions that cannot be confirmed to be within these bounds are rejected.
Nearby AERONET sites are weighted more heavily in the comparison to allow more
representative measurements to dominate the filtration process. The bottom row of
Figure S-1 shows monthly mean comparisons with AERONET by zone, plus comparison
of unfiltered locations. MODIS AOD over zone 2 (470/660: >0.6; 660/2100: 0.4-0.6)
and zone 9 (470/660: >0.6; 660/2100: >0.6) show more scatter than other zones. Figure
S-2 shows the total number of months allowed for each instrument as a result of this
filtration process. MODIS AOD are frequently rejected over bright surfaces, such as
deserts, and are more heavily filtered than MISR.

To reduce the influence of large particles, we also exclude individual MODIS and MISR
AOD with less than 20% fine mode fraction based upon their respective retrievals of this
quantity. The albedo-filtered, fine-mode-filtered AOD from MODIS and MISR are
averaged to produce daily of AOD at 0.1º x 0.1º.

Comparison of GEOS-Chem vertical structure with CALIPSO measurements

The Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO)
satellite has been providing aerosol backscatter and extinction profiles from orbit since
June 2006 (Vaughan et al. 2004). Extinction profiles obtained from CALIPSO are
presently unvalidated, beta-quality products. This dataset, however, is the most complete
measurement-based representation of global aerosol profiles currently available and a
valuable source of information for the validation of simulated vertical profiles and their impact on satellite-derived PM$_{2.5}$. We therefore compare simulated and measured AOD relative vertical profiles from GEOS-Chem and CALIPSO.

Figure S-3 shows average relative vertical profiles from CALIPSO for various land regions, for June-December 2006, the period of overlap with GEOS-4 meteorological fields. The fraction of AOD within the simulated lower mixed layer ranges from about 30% over Europe to 50% over North Africa. This represents a lower bound for fully sampled mean conditions, as profiles taken during high pollution events are unlikely to reach the ground due to attenuation of the CALIPSO beam. Figure S-3 also shows the mean of coincidently sampled profiles from the GEOS-Chem simulation. Simulated and retrieved profiles are consistent. The largest regional differences occur at approximately 5 km. The fraction of AOD in the mixed layer typically differ by less than 5%, with the exception of South America and Polynesia, where this difference is within 15%. There are concerns about an error in the CALIPSO data below 800 m (Ray Hoff, personal communication). Differences in the mixed layer fraction of simulated and observed AOD remain within the above percentages when excluding these values.

Comparison of simulated and satellite-derived PM$_{2.5}$

Of interest is whether the satellite-derived PM$_{2.5}$ improves over the GEOS-Chem simulation of PM$_{2.5}$. Table S-2 compares satellite-derived and simulated PM$_{2.5}$ with ground-based PM$_{2.5}$ over North America and the rest of the world. PM$_{2.5}$ data are sampled coincidently over North America. Annual average measurements are used for
the rest of the world. The slope between ground-based measurements and satellite-
derived PM$_{2.5}$ at 0.1° x 0.1° is consistently nearer to unity as compared to the simulation.
The bias is also smaller between the satellite-data and ground-based measurements.
Much of the global improvement in slope is driven by the finer resolution of satellite-
derived PM$_{2.5}$ (0.86 for 0.1° x 0.1° versus 0.59 for 2° x 2.5°), but correlation is higher with
the satellite product than for the simulation regardless (satellite-derived: 0.69-0.75 versus
simulated: 0.52). By contrast, coarse resolution comparisons over western North
America have an improved slope relative to simulation (0.84 versus 0.51), but a poorer
correlation than at 0.1° x 0.1° (0.62 versus 0.42).

Figure S-4 shows global coincidently sampled satellite-derived and simulated PM$_{2.5}$ at
the simulation resolution of 2° x 2.5°. Both PM$_{2.5}$ estimates agree with each other (r =
0.82), with major enhancements associated with dust, biomass burning and industrial
activities. The magnitude of the concentrations, however, have pronounced differences.
Simulated values of PM$_{2.5}$ over the Sahara exceed satellite-derived estimates by 20-150
µg/m$^3$. Satellite-derived PM$_{2.5}$ deviate from simulated concentrations over east Asia and
northern India by as much as 30 µg/m$^3$. Satellite-derived PM$_{2.5}$ over Mexico has an
enhancement of 5-10 µg/m$^3$ relative to simulation The large population present in the
latter three regions make differences of particular epidemiological significance and may
indicate regional bias in current emission inventories.
References


Mariani RL, de Mello WZ. 2007. Pm2.5-10, pm2.5 and associated water-soluble inorganic species at a coastal urban site in the metropolitan region of rio de janeiro. Atmospheric Environment 41(13): 2887-2892.


Table S-1: Additional PM$_{2.5}$ surface measurements used for comparison and their combined values. Source indicates all sources used to determine location value.

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<tr>
<th>City/Site</th>
<th>Country</th>
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<th>Satellite-derived PM$_{2.5}$ (µg/m$^3$)</th>
<th>Lat</th>
<th>Lon</th>
<th>Study Period</th>
<th>Source</th>
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<td>(Hopke et al. 2008);Environment Protection and Heritage Council</td>
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<td>37.4°</td>
<td>44.3°</td>
<td>Summer</td>
<td>2001</td>
<td>2002-2004</td>
<td>2002-2004</td>
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<td>2001</td>
<td>2003-2004-Feb 2004-Jan 2005</td>
<td>2004-Feb 2005</td>
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<tr>
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<td>29.3°</td>
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<td>2001</td>
<td>2005</td>
<td>2005</td>
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<td>ATENEO DE MANILLA</td>
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<td>14.6°</td>
<td>53.5°</td>
<td>Summer</td>
<td>2001</td>
<td>2002-2005; 2001-2004; Apr - Dec 2004</td>
<td>2005</td>
</tr>
<tr>
<td>SINGAPORE</td>
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<td>1.3°</td>
<td>27.2°</td>
<td>Summer</td>
<td>2001</td>
<td>Jan-May 2000</td>
<td>Jan-Dec 2000</td>
</tr>
<tr>
<td>AEA</td>
<td>Sri Lanka</td>
<td>6.9°</td>
<td>28.4°</td>
<td>Summer</td>
<td>2001</td>
<td>2002-2005</td>
<td>2002-2005</td>
</tr>
<tr>
<td>City</td>
<td>Country</td>
<td>Latitude</td>
<td>Longitude</td>
<td>Date Range</td>
<td>References</td>
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<tr>
<td>Hanoi</td>
<td>Vietnam</td>
<td>53.3</td>
<td>49.5</td>
<td>21°</td>
<td>105.8°</td>
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<td></td>
<td>Jan - Dec 2001-2004; 2002-2005</td>
<td>(Cohen et al. 2002); (Oanh et al. 2006); (Hopke et al. 2008)</td>
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Table S-2: Comparison of simulated and satellite-derived PM$_{2.5}$ with ground-based measurements.$^a$

<table>
<thead>
<tr>
<th>Region</th>
<th>Data Source</th>
<th>Sampling Resolution</th>
<th>slope</th>
<th>bias [µg/m$^3$]</th>
<th>r</th>
<th>n</th>
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<tr>
<td>North America$^{b,e}$</td>
<td>Satellite</td>
<td>0.1° x 0.1°</td>
<td>1.02</td>
<td>-1.07</td>
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<td></td>
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<td>0.79</td>
<td>199</td>
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<tr>
<td></td>
<td>Simulation</td>
<td>2° x 2.5°</td>
<td>1.22</td>
<td>-2.62</td>
<td>0.88</td>
<td>199</td>
</tr>
<tr>
<td></td>
<td>Satellite</td>
<td>0.1° x 0.1°</td>
<td>1.18</td>
<td>-3.12</td>
<td>0.74</td>
<td>760</td>
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<td></td>
<td>2° x 2.5°</td>
<td>1.01</td>
<td>-1.12</td>
<td>0.81</td>
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<tr>
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<td>Simulation</td>
<td>2° x 2.5°</td>
<td>1.33</td>
<td>-3.63</td>
<td>0.91</td>
<td>115</td>
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<td>E. North America$^{b,e}$</td>
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<td>1.22</td>
<td>-2.62</td>
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<td>760</td>
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<td>0.81</td>
<td>115</td>
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<tr>
<td>W. North America$^{b,e}$</td>
<td>Simulation</td>
<td>2° x 2.5°</td>
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<td>-3.63</td>
<td>0.91</td>
<td>115</td>
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<td>0.62</td>
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<td>84</td>
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<td>Global$^{c,e}$</td>
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<tr>
<td>Global (non-EU)$^{d,e}$</td>
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<td>8.51</td>
<td>0.63</td>
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<td>0.85</td>
<td>0.76</td>
<td>84</td>
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<td>Simulation</td>
<td>2° x 2.5°</td>
<td>0.56</td>
<td>2.75</td>
<td>0.71</td>
<td>84</td>
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</tbody>
</table>

$^a$ All PM$_{2.5}$ data are averaged within the sampling resolution. A minimum of 5 measurements, 3 years out of 6 are required for each point.

$^b$ North American ground measurements are coincidently sampled with both satellite and simulated values.

$^c$ Global excludes North American sites.

$^d$ Global (non-EU) additionally excludes European sites.

$^e$ NA and Global comparisons are conducted at 35% and 50% relative humidity, respectively, for appropriate comparison with ground measurements.
**Figure Legends**

Figure S-1: Sample of albedo ratio zones, or surface types, used for AOD filtration. The top panel shows zone definitions for July. Marker positions and colors indicate AERONET locations and zones. Acceptable agreement (within 0.1 or 20%) of AERONET and MODIS (+), MISR (x) or both (*) AOD retrievals is shown at each site. An ‘o’ indicates neither satellite retrieval meets this criteria. The bottom row compares AERONET and unfiltered satellite AOD for all months within the predominant zones. MODIS AOD are denoted by blue ‘+’ and MISR AOD by red ‘x’. Agreement of 0.1 or 20% lie within the black dotted lines.

Figure S-2: Number of months remaining from the MODIS and MISR AOD retrievals after filtering to remove bias. Points denote AERONET stations used for bias identification.

Figure S-3: Vertically-resolved aerosol optical depth (AOD) from the top of the atmosphere to the given altitude (z). Red lines show values retrieved from the CALIPSO (CAL) satellite instrument over June-December 2006. Blue lines show values simulated with GEOS-Chem (GC) and sampled coincidently with CALIPSO. Cyan lines denote simulated mixed layer height. Percentages give fraction of AOD within the mixed layer. Regions are defined in Figure 6 of the main article. Error bars give one standard deviation.

Figure S-4: Comparison of coincidently sampled satellite-estimated and simulated PM\textsubscript{2.5}. Satellite-estimated PM\textsubscript{2.5} has been degraded to a resolution of 2° x 2.5°.
Figure S-1: Sample of albedo ratio zones, or surface types, used for AOD filtration. The top panel shows zone definitions for July. Marker positions and colors indicate AERONET locations and zones. Acceptable agreement (within 0.1 or 20%) of AERONET and MODIS (+), MISR (x) or both (*) AOD retrievals is shown at each site. An ‘o’ indicates neither satellite retrieval meets this criteria. The bottom row compares AERONET and unfiltered satellite AOD for all months within the predominant zones. MODIS AOD are denoted by blue ‘+’ and MISR AOD by red ‘x’. Agreement of 0.1 or 20% lie within the black dotted lines.
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Figure S-4: Comparison of coincidently sampled satellite-estimated and simulated PM$_{2.5}$.
Satellite-estimated PM$_{2.5}$ has been degraded to a resolution of 2° x 2.5°.