Global Estimates of Ambient Fine Particulate Matter Concentrations from Satellite-based Aerosol Optical Depth: Development and Application

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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.

SEDAC - Socioeconomic Data and Applications Center

IT - Interim Target

WHO - World Health Organisation
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$_{2.5}$ concentrations at ~10 km × 10 km resolution indicate a global population-weighted geometric mean PM$_{2.5}$ concentration of 20 µg/m$^3$. 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 38% and 50% of the population, respectively. Annual mean PM$_{2.5}$ concentrations exceed 80 µg/m$^3$ over Eastern China.

Evaluation of the satellite-derived estimate with ground-based in-situ measurements indicates significant spatial agreement with North American measurements ($r = 0.77$, slope = 1.07, n = 1057) 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 long-term average PM$_{2.5}$ concentrations.
Introduction

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 have 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$_{2.5}$.

The Moderate Resolution Imaging Spectroradiometer (MODIS) and Multiangle Imaging Spectroradiometer (MISR) instruments onboard the Terra satellite have provided since mid-2000 global observations of aerosol optical depth (AOD), a measure of light extinction by aerosol in 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, the applicability of AOD to surface air quality depends upon several factors, including the vertical structure, composition, size distribution and water content of atmospheric aerosol.

Many studies have investigated the relationship between total-column AOD and surface PM$_{2.5}$ 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 used local meteorological information to improve agreement (e.g. Liu et al. 2005; Koelemeijer et al. 2006) or to filter the data (e.g. Gupta et al. 2006). Some studies have 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$_{2.5}$ relationships cannot be easily extended to other regions due to variation in meteorology and aerosol composition. Unique, local, time-dependent AOD-PM$_{2.5}$ relationships are necessary to infer global estimates of PM$_{2.5}$. Ground-based measurements of aerosol vertical profiles and properties have insufficient coverage to estimate global AOD-PM$_{2.5}$ relationships.

Global chemical transport models (CTMs) resolve atmospheric composition 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$_{2.5}$ from MISR observations by using CTM output to represent local AOD-PM$_{2.5}$ conversion factors over the contiguous United States. Van Donkelaar et al. (2006) extended the approach of Liu et al. (2004) to estimate PM$_{2.5}$ from both MODIS and MISR observations and investigated the factors affecting the agreement between AOD and surface-level PM$_{2.5}$. Statistical models have also been used to relate AOD to PM$_{2.5}$,
using MISR-retrieved spherical vs. non-spherical particle fraction, in addition to model-derived vertical distribution, to separate mineral dust from other aerosol species (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 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° × 0.1°, or approximately 10 km × 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$_{2.5}$ concentrations and validate this with ground-based (in-situ) observations. We estimate global exposure to outdoor ambient PM$_{2.5}$ using our satellite-derived product to demonstrate potential application 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 (Diner et al. 2005; Martonchik et al. 2009) to reduce algorithmic assumptions and retrieval bias, as well as obtain information about microphysical properties, and plume heights in aerosol source regions (Kahn et al. 2007). Neither instrument can retrieve AOD in cloudy conditions.

We use the MODIS BRDF/Albedo product (MOD43, Collection 5) to distinguish surface types and, in conjunction with ground-based retrievals of AOD, to identify regions of high bias in both MODIS and MISR AOD. We define these surface types for each month according to the ratio of surface albedo for different wavelengths, similar to assumptions inherent in the MODIS AOD retrieval. We remove AOD retrieved from either instrument with an anticipated bias greater than ±(0.1 or 20%), based on comparison with AERONET sun photometer measurements of AOD. Remaining MODIS and MISR AOD are averaged to produce a single value at a given grid cell. The supplemental material describes in detail the satellite retrievals, and this bias filtration. We restrict our subsequent analysis to locations with at least 50 successful satellite retrievals over 2001-2006, to yield near-complete (95%) global geographic coverage.

Estimating PM$_{2.5}$ from Aerosol Optical Depth

Estimating ground-level concentrations of dry 24-h PM$_{2.5}$ (in µg/m$^3$) from satellite observations of total-column AOD (unitless) requires a conversion factor that accounts for their spatially and temporally varying relationship.
\[ \text{PM}_{2.5} = \eta \cdot \text{AOD} \]  

\[ \eta \] is a function of the factors that relate 24-h dry aerosol mass to satellite observations of ambient AOD: aerosol size, aerosol type, diurnal variation, relative humidity and the vertical structure of aerosol extinction (van Donkelaar et al. 2006). 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. The model calculates the global 3-D distribution of aerosol mass and AOD with a transport timestep of 15 minutes. We apply the modelled relationship between aerosol mass and relative humidity for each aerosol type to calculate \( \text{PM}_{2.5} \) for relative humidity values that correspond to surface-measurement standards (35% for U.S. and Canada; 50% for Europe). We calculate daily values of \( \eta \) as the ratio of 24-h ground-level \( \text{PM}_{2.5} \) 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. The AOD is averaged between 10 a.m. and 12 p.m. local time, corresponding to the Terra overpass period. We interpolate values of \( \eta \) from \( 2^\circ \times 2.5^\circ \), the resolution of the GEOS-Chem simulation, to \( 0.1^\circ \times 0.1^\circ \) for application to satellite AOD values.
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 NAPS Network, maintained by Environment Canada (http://www.etc.cte.ec.gc.ca/NAPS/index_e.html). U.S. data are from the 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 244 annually representative, ground-based PM$_{2.5}$ data from both published and unpublished field measurements outside the United States and Canada, 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 24-h average PM$_{2.5}$ sampled on the same days as successful satellite observations. Both the MODIS and MISR instruments indicate some relationship between retrieved total-column AOD and in-situ PM$_{2.5}$, both with spatial correlation coefficients of 0.39. A simple average of the daily AOD from both instruments yields a correlation of 0.44. Combining retrievals from these instruments as described in the methods section increases the correlation to 0.61. Additional information is required to quantitatively estimate PM$_{2.5}$ concentrations 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 North 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 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. The western and northern parts of the continent are generally characterized by low concentrations, with a few exceptions. Geographic mean PM$_{2.5}$ concentrations over eastern and western North America are 6.9 $\mu$g/m$^3$ and 6.2 $\mu$g/m$^3$, respectively. Application of $\eta$ (Figure 2) increased the spatial contrast in Figure 1, reflecting ground-level 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$_{2.5}$ concentrations 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 quantitatively compares satellite-derived and ground-level measured PM$_{2.5}$. We find significant cross-sectional correlation between average coincidently sampled satellite-derived and ground-based PM$_{2.5}$ across North America ($r = 0.77$, slope = 1.07, bias = -1.75 $\mu$g/m$^3$). Many factors contribute to the scatter of points, including differences between what satellite and in situ measurements 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, and all subsequent, are at 50% RH in agreement with European ground-based measurements. We reject points created with less than 50 values, enabling 95% global geographic coverage. The satellite-derived PM$_{2.5}$ include an adjustment for discontinuous sampling as described in the error analysis. Annual mean PM$_{2.5}$ concentrations vary spatially by more than an order of magnitude. Values are less than 10 µg/m$^3$ for large regions of the earth. In contrast, PM$_{2.5}$ concentrations of 60-90 µg/m$^3$ are found over eastern China, with values in excess of 100 µg/m$^3$ for its major industrial regions. 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 15-60 µg/m$^3$. Biomass burning effects on PM$_{2.5}$ levels are visible in central South America and central Africa, where concentrations of 10-17 µg/m$^3$ are estimated. Dust transport in the fine mode is substantial (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/m$^3$, n = 244). Similar agreement is obtained when all sites except Europe and North America are considered ($r = 0.83$, slope = 0.91, intercept = -2.64 µg/m$^3$, n = 84).

Figure 5 overlays 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. Some relationships are apparent between PM$_{2.5}$, topography and populationl. Heavily populated and highly polluted, low-lying regions of eastern China and the Po Valley of northern Italy contrast sharply with neighboring higher altitude regions. The Appalachian Mountains in eastern North America emerge as a relatively clean region. Many PM$_{2.5}$ enhancements are associated with urban or industrial areas, but these relationships are complex.

Error Analysis

The dominant sources of error in satellite-derived PM$_{2.5}$ arise from uncertainties in both AOD retrieval and aerosol vertical structure (van Donkelaar et al. 2006). The residual AOD bias after data filtering in within $\pm(0.1 + 20\%)$, as evaluated with ground-based AERONET measurements. 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 estimate the error in satellite-derived PM$_{2.5}$ as the change in PM$_{2.5}$ that occurs when $\eta$ and AOD are adjusted by their uncertainty, approximated as the GEOS-Chem vertical profile bias and residual satellite AOD bias, respectively.

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 $\pm15\%$ 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$_{2.5}$. The total number of successful satellite retrievals are shown in the top panel of Figure 7 and summarized regionally as population-weighted mean in Table 2. Lower sampling is fortuitously collocated with lower population. The global population-weighted mean number of observations per 0.1° × 0.1° box is 297. The bottom panel of Figure 7 shows the percent difference 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 ±20% of simulated PM$_{2.5}$. Regions of low sampling do not necessarily demonstrate enhanced uncertainty and vice versa. Sampling error of satellite-derived PM$_{2.5}$ is larger in regions influenced by biomass burning, mineral dust, or persistent cloud due to a combination of large seasonal variability and non-representative sampling. We apply the ratio of complete to coincident mean simulated PM$_{2.5}$ to reduce uncertainty from sampling variability.

Validation of this 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 = 1.13, r = 0.70 versus coincident: slope = 1.07, r = 0.77), 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 ±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 µg/m$^3$.

*Global Ambient PM$_{2.5}$: Application to Population Exposure*

Pope et al. [2009] estimate that a long-term PM$_{2.5}$ exposure decrease of 10 µg/m$^3$ increases life expectancy by 0.61 ± 0.30 years for the United States. We estimate global long-term exposure to ambient PM$_{2.5}$ 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 and regional distributions of long-term ambient PM$_{2.5}$ exposure. Table 3 summarizes these results. All regions exhibit non-linear relationships between population and PM$_{2.5}$ concentrations. Eastern and central Asia have the highest levels of PM$_{2.5}$ concentrations with 38-50% of the 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 PM$_{2.5}$ 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
from ground monitors are currently the “gold-standard” for epidemiological studies, these
are not only sparse, but may represent only a small spatial extent in heterogeneous
regions (Chen et al. 2006). Satellite observations offer area-integrated values with global
coverage, providing valuable additional information for global health studies.

In this work, we produced a satellite-derived climatology of PM$_{2.5}$ concentrations. 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. 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.
Our estimates suggest the global population-weighted geometric mean PM$_{2.5}$ concentration is 20 µg/m$^3$ and 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; half (50%) 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 0.1º × 0.1º retains variation relevant to population distribution. 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 = 1.07, r = 0.77, n = 1057), 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 reflect multiple influences.

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 profile 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, by persistent cloud, or for individual, severe pollution events. The overall combined PM$_{2.5}$ uncertainty of ±25% indicates a mean global, population-weighted uncertainty in PM$_{2.5}$ concentration of 6.7 µg/m$^3$.

Additional developments could continue to reduce error in the satellite-derived PM$_{2.5}$ estimates presented here. Increased satellite coverage would reduce sampling concerns and may allow for satellite-derived PM$_{2.5}$ to be applied to studies of temporal or spatiotemporal variation. 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 concentrations.
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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}.

|                      | slope\textsuperscript{b} | intercept | r      | n       |
|----------------------|--------------------------|-----------|--------|---------|
| MODIS AOD            | 0.020                    | 0.10      | 0.39   | 1218    |
| MISR AOD             | 0.010                    | 0.11      | 0.39   | 353     |
| Average AOD          | 0.015                    | 0.06      | 0.44   | 1236    |
| Combined AOD         | 0.017                    | 0.10      | 0.61   | 1057    |
| Satellite-derived PM\textsubscript{2.5} | 1.066                   | -1.75     | 0.77   | 1057    |

\textsuperscript{a} A minimum of 50 measurements are required for each point. 
\textsuperscript{b} calculated with reduced major-axis linear regression (Hirsh and Gilroy 1984)
Table 2: Regional PM$_{2.5}$ statistics, number of observations and population in excess of WHO Air Quality Guideline (AQG) and Interim Targets (IT). Regions are defined in Figure 6.

| Region            | Population-weighted statistics [µg/m$^3$] | Population-weighted total observations | Population (million people) |
|-------------------|------------------------------------------|----------------------------------------|-----------------------------|
|                   | mean | std | geo. mean | geo. std. | Total | AQG (10 µg/m$^3$) | IT-3 (15 µg/m$^3$) | IT-2 (25 µg/m$^3$) | IT-1 (35 µg/m$^3$) |
| World             | 27   | 23  | 20        | 2.3       | 297   | 6,400            | 5,100 (81%) | 4,000 (63%) | 2,300 (37%) | 1,600 (25%) |
| E. Asia           | 44   | 29  | 34        | 2.2       | 270   | 1,900            | 1,800 (93%) | 1,600 (83%) | 1,200 (65%) | 940 (50%)   |
| C. Asia           | 31   | 16  | 27        | 1.8       | 230   | 1,500            | 1,400 (94%) | 1,300 (86%) | 780 (54%)  | 560 (38%)   |
| N. Africa         | 26   | 10  | 24        | 1.5       | 195   | 540              | 530 (98%)  | 450 (85%)  | 250 (47%)  | 8.7 (17%)   |
| S. Africa         | 11   | 6   | 9         | 1.9       | 368   | 540              | 240 (47%)  | 110 (21%)  | 7.7 (1.5%) | 0.1 (0.0%)  |
| E. Europe         | 15   | 5   | 14        | 1.4       | 437   | 450              | 400 (88%)  | 210 (47%)  | 14 (3.0%)  | 1.8 (0.4%)  |
| S. America        | 7    | 4   | 5         | 2.0       | 361   | 400              | 52 (13%)   | 18 (4.5%)  | 5.3 (1.3%) | 0.0 (0.0%)  |
| E. North America  | 13   | 5   | 12        | 1.5       | 476   | 350              | 250 (72%)  | 110 (32%)  | 17 (4.7%)  | 0.0 (0.0%)  |
| S. Asia/Australia | 12   | 6   | 10        | 1.8       | 304   | 310              | 180 (58%)  | 72 (24%)   | 13 (4.4%)  | 1.3 (0.4%)  |
| W. Europe         | 17   | 5   | 16        | 1.4       | 311   | 260              | 250 (94%)  | 170 (63%)  | 15 (5.9%)  | 3.7 (1.4%)  |
| W. North America  | 11   | 5   | 10        | 1.6       | 366   | 120              | 56 (46%)   | 22 (18%)   | 0.3 (0.2%) | 0.0 (0.0%)  |

* (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 the combined product developed here. White space denotes water or fewer than 50 measurements.

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 mean satellite-derived PM$_{2.5}$ between 2001-2006. White space denotes water or fewer than 50 AOD measurements. 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 uncertainty of $\pm (1 \mu g/m^3 \pm 15\%)$. The line of best fit (Hirsh and Gilroy 1984) is dashed.

Figure 4: Global satellite-derived PM$_{2.5}$ averaged over 2001-2006. White space indicates water or locations containing less than 50 measurements. 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$_{2.5}$ concentrations. Columns show mean satellite-derived PM$_{2.5}$ for 2001-2006 at locations containing at least 50 measurements. Contours denote population density in the left column and surface elevation in the right. The color scale varies by region. Crosses indicate city centers in both columns. 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: Satellite-derived PM$_{2.5}$ sampling and its estimated induced uncertainty. The top panel shows the total number of values used from satellite per 0.1º grid box. The bottom panel shows the percent change in average coincidently-sampled simulated PM$_{2.5}$ concentrations relative to a full year average.

Figure 8: Cumulative distribution of regional, annual mean PM$_{2.5}$ estimated from satellite-derived PM$_{2.5}$ at a resolution of 0.1º $\times$ 0.1º for 2001-2006. The top axis identifies WHO Air Quality Guidelines (AQG) and Interim Targets (IT-#) associated with each concentration level. Regions are defined in Figure 6.
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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.ideam.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 $0.1^\circ \times 0.1^\circ$ 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 model timestep for transport is 15 minutes.

The GEOS-Chem aerosol simulation includes the sulfate-nitrate-ammonium system (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. (2008). 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. 2008; 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 seasonally and spatially varying 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 × 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 Aerosol Robotic Network (AERONET, Holben et al. 1998) 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 × 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 as the ratio of fine AOD (sulfate, organic carbon, black carbon, and fine dust and fine sea salt) to total AOD (fine 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 × 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 AERONET 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 compares unfiltered satellite and AERONET AOD by zone for all months. 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 included from each instrument after this filtration process. MODIS AOD are frequently rejected over bright surfaces, such as deserts, and are more heavily filtered than MISR. Regions with few months are more susceptible to sampling bias as discussed in the main text. Fortunately most of the regions with poor seasonal sampling tend to have low population.

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 0.1° × 0.1°.
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° × 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° × 0.1° versus 0.59 for 2° × 2.5°), but correlation is higher with the satellite product than for the simulation regardless (satellite-derived: 0.75-0.83 versus simulated: 0.63). By contrast, coarse resolution comparisons over western North America have an improved slope relative to simulation (0.83 versus 0.49), but a poorer correlation than at 0.1° × 0.1° (0.67 versus 0.53).

Figure S-4 shows global coincidently sampled satellite-derived and simulated PM$_{2.5}$ at the simulation resolution of 2° × 2.5°. Both PM$_{2.5}$ estimates agree with each other (r = 0.77), 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.
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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.

| City/Site       | Country      | In-situ PM$_{2.5}$ (µg/m$^3$) | Satellite-derived PM$_{2.5}$ (µg/m$^3$) | Lat  | Lon   | Study Period       | Number of Stations | Source                                                                 |
|-----------------|--------------|-------------------------------|-----------------------------------------|------|-------|--------------------|-------------------|------------------------------------------------------------------------|
| LIVERPOOL       | AUSTRALIA    | 8.2                           | 5                                       | -33.9º | 150.9º | 2002-2005;2005;2005 | 1;1;1             | (Hopke et al. 2008); Environment Protection and Heritage Council; Environment Protection and Heritage Council |
| LUCAS HEIGHTS   | AUSTRALIA    | 5.7                           | 3.1                                     | -34º  | 151º  | 2002-2005;2005     | 1;1               | (Hopke et al. 2008); Environment Protection and Heritage Council      |
| DHAKA           | BANGLADESH   | 33.7                          | 23.9                                    | 23.8º | 90.4º  | 2000-2003;2005     | 1;1;1             | (Begum et al. 2006); (Begum et al. 2008)                             |
| DHAKA           | BANGLADESH   | 28.7                          | 26.2                                    | 23.7º | 90.4º  | 2002-2005          | 1;1               | (Hopke et al. 2008)                                                  |
| CUIABA          | BRAZIL       | 10.5                          | 10.7                                    | -15.6º | -56.1º | Jul 1991 - Feb 1993 | 1                 | (Artaxo et al. 1994)                                                 |
| RIO DE JANEIRO  | BRAZIL       | 17                            | 7.1                                     | -22.9º | -43.1º | Oct 1998 - Sep 1999 | 1                 | (Mariani and de Mello 2007)                                           |
| RIO DE JANEIRO  | BRAZIL       | 10                            | 5                                       | -22.9º | -43.4º | Sep 2003 - Sept 2004 | 10                | (Soluri et al. 2007)                                                 |
| SAO PAULO       | BRAZIL       | 22.6                          | 8.8                                     | -23.5º | -46.5º | Jul 1997-March 1998 | 2                 | (Castanho and Artaxo 2001)                                            |
| SERRA DO NAVIO  | BRAZIL       | 9.9                           | 6.4                                     | 1º    | -52º   | Nov 1991 - Apr 1993 | 1                 | (Artaxo et al. 1994)                                                 |
| BANGKOK         | CHINA        | 36.6                          | 23.6                                    | 13.8º | 100.5º | 2001-2004;Feb 2002 - Jan 2003 Aug 2001-Sep 2002;2002;2000;2001-2004 | 1;3               | (Oanh et al. 2006); (Chuersuwan et al. 2008)                          |
| BEIJING         | CHINA        | 114.1                         | 97.3                                    | 39.9º  | 116.4º | 2001-2004          | 2;5;1             | (Duan et al. 2006); (Zheng et al. 2005); (Oanh et al. 2006)            |
| BEIJING         | CHINA        | 121                           | 99.6                                    | 39.8º  | 116.5º | Jul 1999- Sep 2000 | 2                 | (He et al. 2001)                                                     |
| BEIJEING        | CHINA        | 35.6                          | 96.1                                    | 39.9º  | 116.3º | 2002-2004          | 1                 | (Hopke et al. 2008)                                                  |
| BEIJING         | CHINA        | 127.5                         | 97                                       | 39.9º  | 116.5º | unknown            | 8                 | (Zhang et al. 2009)                                                  |
| BEIJING         | CHINA        | 87.7                          | 90.4                                    | 39.9º  | 116.3º | 2005-2007          | 1                 | (Zhao et al. 2009)                                                   |
| BEIJING         | CHINA        | 54.2                          | 29.6                                    | 40.6º  | 117.1º | 2005-2007          | 1                 | (Zhao et al. 2009)                                                   |
| BEIJING-SUBURBAN| CHINA        | 42.1                          | 87.2                                    | 39.7º  | 116º   | 2003-2004          | 1                 | (Hopke et al. 2008)                                                  |
| GUANGZHOU       | CHINA        | 97.3                          | 79.3                                    | 23.1º  | 113.1º | unknown            | 2                 | (Zhang et al. 2009)                                                  |
| HONG KONG       | CHINA        | 42.8                          | 45.4                                    | 22.3º  | 114.2º | Nov 2000-Feb 2001; Jun-Aug 2001 | 2                 | (Ho et al. 2006)                                                     |
| HONG KONG       | CHINA        | 47.4                          | 44.5                                    | 25.2º  | 115.1º | unknown            | 2                 | (Zhang et al. 2009)                                                  |
| SHANGHAI        | CHINA        | 92.9                          | 95.8                                    | 31.3º  | 121.3º | 2005-2006          | 2                 | (Feng et al. 2009)                                                   |
| SHANGHAI        | CHINA        | 59.6                          | 65.9                                    | 31.2º  | 121.5º | 1999               | 2                 | (Ye et al. 2003)                                                     |
| SHANGHAI        | CHINA        | 78.6                          | 98.1                                    | 31.1º  | 121.3º | unknown            | 3                 | (Zhang et al. 2009)                                                  |
| City          | Country   | Latitude | Longitude | Temperature  | Dates                      | Authors                                      |
|--------------|-----------|----------|-----------|--------------|----------------------------|----------------------------------------------|
| Cairo        | Egypt     | 30° 31.4° | 30° 31.3° | Fall/Winter 1999; Summer 2002 | 2001-2002, 2001-2004, 2002-2003 | (Abu-Allaban et al. 2007)                   |
| Cairo        | Egypt     | 30° 31.3° | 30° 31.4° | Mar 2001 - Jan 2002, Jul-Dec 2003 | 1; 1; 1, 113 | (Zakey et al. 2008)                     |
| Chennai      | India     | 13° 80.3° | 13° 80.3° | Mar 2001 - Jan 2002, Jul-Dec 2003 | 1; 1 | (Oanh et al. 2006); (Kumar and Joseph 2006) |
| Delhi        | India     | 28° 77.1° | 28° 77.1° | Mar 2001 - Jan 2002, Jul-Dec 2003 | 1; 1 | (The World Bank 2004); (Chowdhury 2004); (Kumar et al. 2007) |
| Kolkata      | India     | 18° 72.8° | 18° 72.8° | Apr 2003 - Mar 2004 | 1 | (Kumar et al. 2007)                   |
| Mumbai       | India     | 19° 72.8° | 19° 72.8° | 2002-2005 | 1 | (Hopke et al. 2008)                   |
| Mumbai       | India     | 19° 72.9° | 19° 72.9° | 2002-2005 | 1 | (The World Bank 2004); (Chowdhury 2004) |
| Mumbai       | India     | 22° 88.3° | 22° 88.3° | Mar 2001 - Jan 2002 | 1; 1 | (Kothai et al. 2008)                  |
| Mumbai       | India     | 18° 73° | 18° 73° | Annual | 1 | (Kothai et al. 2008)                  |
| Bandung      | Indonesia | -6° 107° | -6° 107° | 2001-2004, 2002-2005 | 1; 1 | (Oanh et al. 2006); (Hopke et al. 2008) |
| Lembang      | Indonesia | -6° 107° | -6° 107° | 2002-2005 | 1 | (Hopke et al. 2008)                   |
| Tokyo        | Japan     | 35° 139° | 35° 139° | 2001-2004 | 1 | (Minoura et al. 2006)                  |
| Daegu        | Korea     | 36° 127° | 36° 127° | 2002-2005 | 1 | (Hopke et al. 2008)                   |
| Seoul        | Korea     | 37° 126° | 37° 126° | 2002-2004 | 2 | (Kim et al. 2006)                     |
| Seoul        | Korea     | 39° 126° | 39° 126° | 2005-2006 | 1 | (Park et al. 2008)                    |
| Kuwait       | Kuwait    | 29° 101° | 29° 101° | Feb 2004 - Jul 2005 | 3 | (Brown et al. 2008)                   |
| Beirut       | Lebanon   | 33° 104° | 33° 104° | 2003-2004 | 1; 1 | (Sheehan and Bowman 2001); (Kouyoumdjian and Saliba 2006); (Saliba et al. 2007) |
| Kuala Lumpur | Malaysia  | 3° 101° | 3° 101° | 2005 | 1 | (Hopke et al. 2008)                   |
| Kathmandu    | Nepal     | 27° 85.5° | 27° 85.5° | Dec 1998-Oct 2000 | 2 | (Carrico et al. 2003)                 |
| Cabauw       | Netherlands | 52° 4° | 52° 4° | Aug 2006 - May 2007 | 1 | (Schaap et al. 2008)                  |
| Islamabad    | Pakistan  | 33° 73° | 33° 73° | 2002-2004 | 1 | (Hopke et al. 2008)                   |
| Ateneo        | Philippines | 14° 121° | 14° 121° | 2002-2005, 2004-Apr-Dec 2001 | 1; 1 | (Hopke et al. 2008); (Oanh et al. 2006); (Cohen et al. 2002) |
| Singapore    | Singapore | 1.3° 104° | 1.3° 104° | Jan-Dec 2000 | 1 | (Balasubramanian et al. 2003)         |
| Boitsalano    | South Africa | -25.5° 25.8° | -25.5° 25.8° | July 2006-July 2007 | 1 | (Laakso et al. 2008)                  |
| AEA          | Sri Lanka | 6° 79.9° | 6° 79.9° | 2002-2005 | 1 | (Hopke et al. 2008); (Lin 2002)       |
| Kaohsiung    | Taiwan    | 22.6° 120.3° | 22.6° 120.3° | Nov 1998-Apr 1999 | 6 | (Hopke et al. 2008)                   |
| Bangkok      | Thailand  | 13.8° 100.5° | 13.8° 100.5° | 2002-2005 | 1 | (Hopke et al. 2008)                   |
| Pathum Thani | Thailand  | 14° 100.5° | 14° 100.5° | 2003-2005 | 1 | (Hopke et al. 2008)                   |
| Istanbul     | Turkey    | 41° 28.6° | 41° 28.6° | Jul 2002 - Jul 2003 | 1 | (Karaca et al. 2005; Karaca et al. 2008) |
| City     | Country     | Lat. | Long. | Latitude | Longitude | Year Range   | Data Source         |
|----------|-------------|------|-------|----------|-----------|--------------|---------------------|
| HANOI    | VIETNAM     | 53.3 | 49.5  | 21°      | 105.8°    | Jan - Dec 2001; 2001-2004; 2002-2005 | 1:1:1               |

(Cohen et al. 2002); (Oanh et al. 2006); (Hopke et al. 2008)
### Table S-2: Comparison of simulated and satellite-derived PM$_{2.5}$ with ground-based measurements.$^a$

| Region                     | Data Source | Resolution | slope | bias [µg/m$^3$] | r   | n   |
|----------------------------|-------------|------------|-------|-----------------|-----|-----|
| North America$^{b,e}$      | Satellite   | $0.1^\circ \times 0.1^\circ$ | 1.07  | -1.75           | 0.77 | 1057|
|                            | Satellite   | $2^\circ \times 2.5^\circ$   | 0.94  | 0.38            | 0.82 | 190 |
|                            | Simulation  | $2^\circ \times 2.5^\circ$   | 1.04  | -1.54           | 0.83 | 117 |
| E. North America$^{b,e}$   | Satellite   | $0.1^\circ \times 0.1^\circ$ | 1.20  | -3.40           | 0.74 | 798 |
|                            | Satellite   | $2^\circ \times 2.5^\circ$   | 1.34  | -3.80           | 0.92 | 117 |
|                            | Simulation  | $2^\circ \times 2.5^\circ$   | 0.69  | 1.39            | 0.67 | 259 |
| W. North America$^{b,e}$   | Satellite   | $0.1^\circ \times 0.1^\circ$ | 0.63  | 0.76            | 0.53 | 73  |
|                            | Satellite   | $2^\circ \times 2.5^\circ$   | 0.49  | 2.40            | 0.40 | 73  |
|                            | Simulation  | $2^\circ \times 2.5^\circ$   | 0.83  | 2.40            | 0.40 | 73  |
| Global$^{c,e}$              | Satellite   | $0.1^\circ \times 0.1^\circ$ | 0.86  | 1.15            | 0.83 | 244 |
|                            | Satellite   | $2^\circ \times 2.5^\circ$   | 0.59  | 4.37            | 0.75 | 244 |
|                            | Simulation  | $2^\circ \times 2.5^\circ$   | 0.54  | 8.89            | 0.63 | 244 |
| Global (non-EU)$^{d,e}$    | Satellite   | $0.1^\circ \times 0.1^\circ$ | 0.91  | -2.64           | 0.83 | 84  |
|                            | Satellite   | $2^\circ \times 2.5^\circ$   | 0.64  | 0.78            | 0.76 | 84  |
|                            | Simulation  | $2^\circ \times 2.5^\circ$   | 0.60  | 2.45            | 0.72 | 84  |

$^a$ All PM$_{2.5}$ data are averaged within the sampling resolution. A minimum of 50 measurements 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 (×) 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 ‘×’. 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$_{2.5}$. Satellite-estimated PM$_{2.5}$ has been degraded to a resolution of 2º × 2.5º.
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 (×) 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 ‘×’. Agreement of 0.1 or 20% lie within the black dotted lines.
Figure S-2: Number of months of MODIS and MISR AOD included in satellite-derived PM$_{2.5}$ estimate. 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$_{2.5}$. Satellite-estimated PM$_{2.5}$ has been degraded to a resolution of 2º × 2.5º.