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

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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 $0.1^{o} \times 0.1^{o}$ 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 2º × 2.5º 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 (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 (±70.5º, ±60.0º, ±45.6º, ±25.1º 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 $0.1^{o} \times 0.1^{o}$.

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 0.1º × 0.1º 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 0.1º × 0.1º versus 0.59 for 2º × 2.5º), 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 0.1º × 0.1º (0.67 versus 0.53).

Figure S-4 shows global coincidently sampled satellite-derived and simulated $PM_{2.5}$ at the simulation resolution of 2º × 2.5º. 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 $\mu g/m^3$. Satellite-derived $PM_{2.5}$ deviate from simulated concentrations over east Asia and northern India by as much as 30 $\mu g/m^3$. Satellite-derived $PM_{2.5}$ over Mexico has an enhancement of 5-10 $\mu g/m^3$ 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 $PM_{2.5}$ ($\mu g/m^3$)	Satellite-derived $PM_{2.5}$ ($\mu g/m^3$)	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		(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	Sept 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	1;3	(Oanh et al. 2006);(Chuersuwan et al. 2008)
BEIJING	CHINA	114.1	97.3	39.9º	116.4º	Aug 2001-Sep 2002;2000;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)
BEIJING	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)

CAIRO	EGYPT	67.4	51.8	30º	31.4º	Fall/Winter 1999 Summer 2002	3	(Abu-Allaban et al. 2007)
CAIRO	EGYPT	79.3	41.6	30º	31.3º	2001-2002	14	(Zakey et al. 2008)
CHENNAI	INDIA	42.2	18	13.1º	80.3º	2001-2004;2002-2003	1;1	(Oanh et al. 2006);(Kumar and Joseph 2006)
DELHI	INDIA	97	56.8	28.4º	77.1º	Mar 2001 - Jan 2002;Jul-Dec 2003	1;1;113	(The World Bank 2004); (Chowdhury 2004);(Kumar et al. 2007)
KOLKATA	INDIA	107.8	26.2	18.7º	72.8º	Mar 2001 - Jan 2002	1;1	(The World Bank 2004); (Chowdhury 2004)
MUMBAI	INDIA	43	26.2	19º	72.8º	Apr 2003- Mar 2004	1	(Kumar et al. 2007)
MUMBAI	INDIA	40.5	23	19.1º	72.9º	2002-2005	1	(Hopke et al. 2008)
MUMBAI	INDIA	52.5	38.8	22.6º	88.3º	Mar 2001 - Jan 2002	1;1	(The World Bank 2004); (Chowdhury 2004)
NAVI MUMBAI (VASHI)	INDIA	44	26.2	18.8º	73º	Annual	1	(Kothai et al. 2008)
BANDUNG	INDONESIA	29.2	16.8	-6.5º	107.4º	2001-2004;2002-2005	1;1	(Oanh et al. 2006);(Hopke et al. 2008)
LEMBANG	INDONESIA	12.9	19.6	-6.2º	107.2º	2002-2005;	1	(Hopke et al. 2008)
TOKYO	JAPAN	23	23.2	35.7º	139.7º	2001-2004	1	(Minoura et al. 2006)
DAEJEON	KOREA	10.8	20.3	36.4º	127.4º	2002-2005	1	(Hopke et al. 2008)
SEOUL	KOREA	44.3	30	37.4º	126.8º	2002-2004	2	(Kim et al. 2006)
SEOUL	KOREA	37.2	39.5	37.6º	126º	2005-2006	1	(Park et al. 2008)
KUWAIT	KUWAIT	38	37.5	29.3º	48º	Feb 2004 - Jul 2005	3	(Brown et al. 2008)
BEIRUT	LEBANON	35.5	24	33.9º	35.5º	Feb-May 2003;2004;Feb 2004-Jan 2005	1;1;1	(Sheehan and Bowman 2001);(Kouyoumdjian and Saliba 2006);(Saliba et al. 2007)
KUALA LUMPUR	MALAYSIA	29.3	17.1	3.2º	101.7º	2005	1	(Hopke et al. 2008)
KATHMANDU VALLEY	NEPAL	30.7	22.8	27.7º	85.5º	Dec 1998-Oct 2000	2	(Carrico et al. 2003)
CABAUW	NETHERLANDS	18.2	19.1	52º	4.9º	Aug 2006 - May 2007	1	(Schaap et al. 2008)
ISLAMABAD	PAKISTAN	14.4	30.8	33.7º	73.3º	2002-2004	1	(Hopke et al. 2008)
ATENEO DE MANILLA	PHILIPPINES	35.3	15.9	14.6º	121.1º	2002-2005;2001-2004;Apr - Dec 2001	1;1;1	(Hopke et al. 2008);(Oanh et al. 2006);(Cohen et al. 2002)
SINGAPORE	SINGAPORE	27.2	25.7	1.3º	104º	Jan-Dec 2000	1	(Balasubramanian et al. 2003)
BOTSALANAO	SOUTH AFRICA	10.5	4.9	-25.5º	25.8º	July 2006-July2007	1	(Laakso et al. 2008)
AEA	SRI LANKA	28.4	12.1	6.9º	79.9º	2002-2005	1	(Hopke et al. 2008);
KAOHSIUNG	TAIWAN	68	32.6	22.6º	120.3º	Nov 1998-Apr 1999	6	(Lin 2002)
BANGKOK	THAILAND	23.8	24.1	13.8º	100.5º	2002-2005	1	(Hopke et al. 2008)
PATHUM THANI	THAILAND	20	19.4	14º	100.5º	2003-2005	1	(Hopke et al. 2008)
ISTANBUL	TURKEY	20.8	17.6	41º	28.6º	Jul 2002 - Jul 2003	1	(Karaca et al. 2005; Karaca et al. 2008)

HANOI	VIETNAM	53.3	49.5	21º	105.8º	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	Sampling Resolution	slope	bias [μg/m^3]	r	n
North America[b,e]	Satellite	0.1º × 0.1º	1.07	-1.75	0.77	1057
		2º × 2.5º	0.94	0.38	0.82	190
	Simulation	2º × 2.5º	1.26	-3.14	0.87	190
E. North America[b,e]	Satellite	0.1º × 0.1º	1.20	-3.40	0.74	798
		2º × 2.5º	1.04	-1.54	0.83	117
	Simulation	2º × 2.5º	1.34	-3.80	0.92	117
W. North America[b,e]	Satellite	0.1º × 0.1º	0.69	1.39	0.67	259
		2º × 2.5º	0.83	0.76	0.53	73
	Simulation	2º × 2.5º	0.49	2.40	0.40	73
Global[c,e]	Satellite	0.1º × 0.1º	0.86	1.15	0.83	244
		2º × 2.5º	0.59	4.37	0.75	244
	Simulation	2º × 2.5º	0.54	8.89	0.63	244
Global (non-EU)[d,e]	Satellite	0.1º × 0.1º	0.91	-2.64	0.83	84
		2º × 2.5º	0.64	0.78	0.76	84
	Simulation	2º × 2.5º	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.

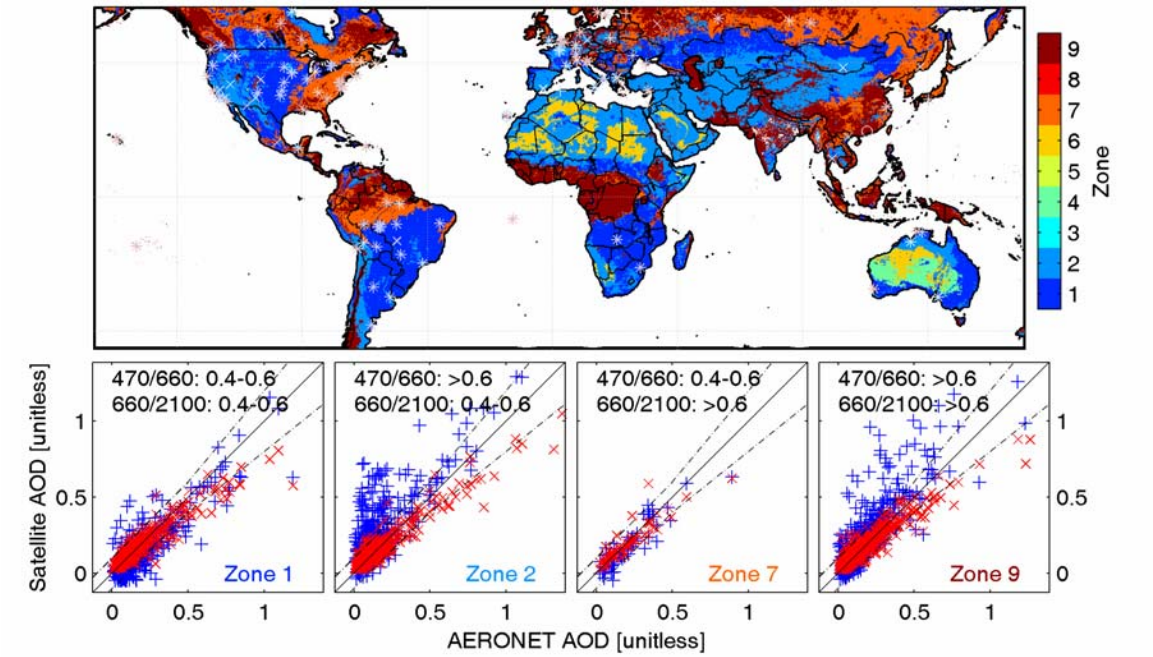
Figure Legends

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.



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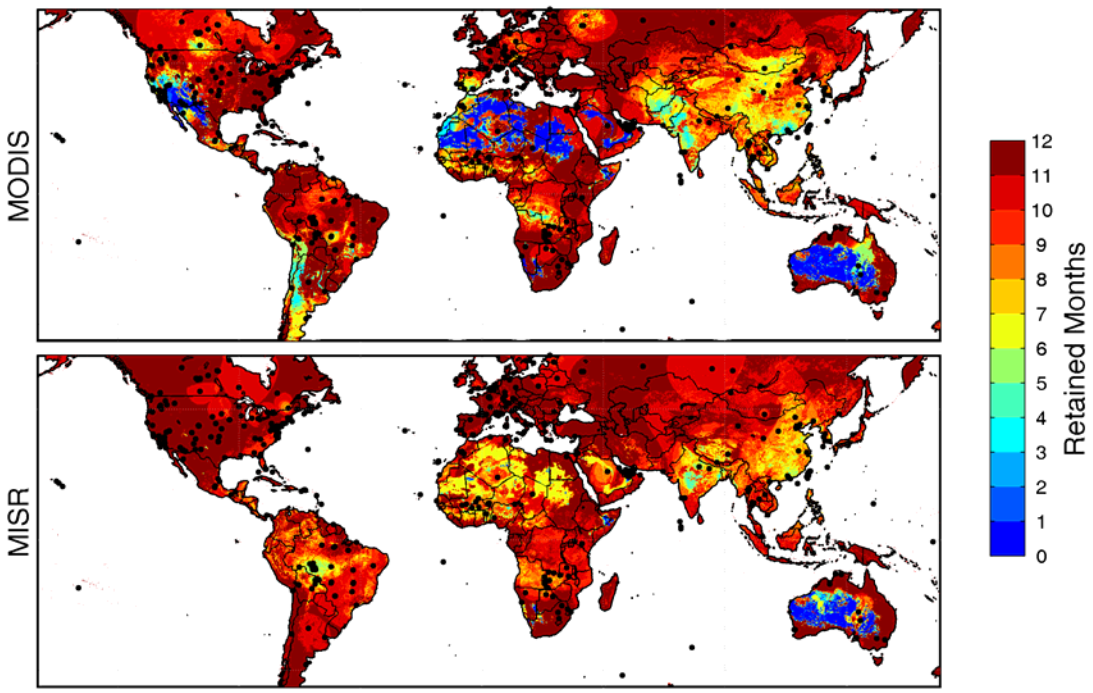
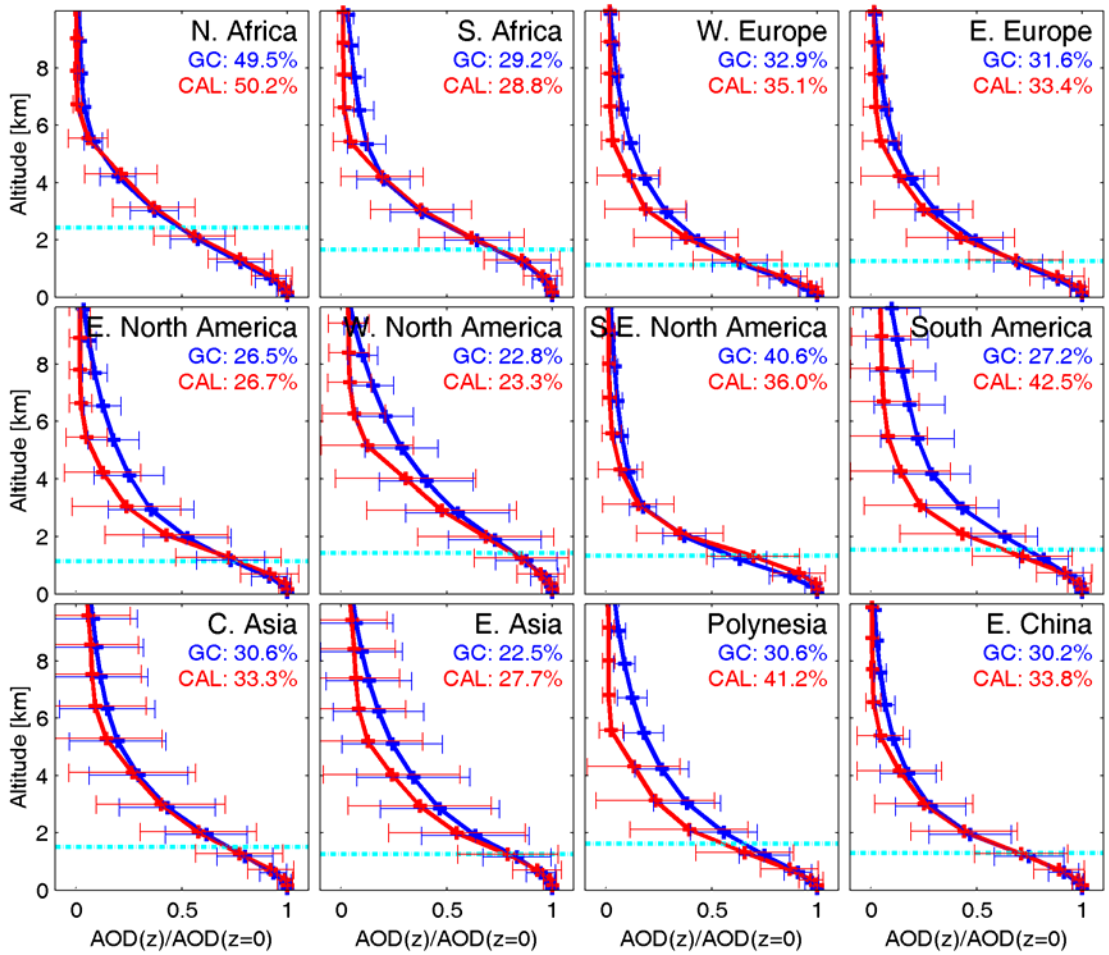


Figure S-2: Number of months of MODIS and MISR AOD included in satellite-derived $PM_{2.5}$ estimate. Points denote AERONET stations used for bias identification.



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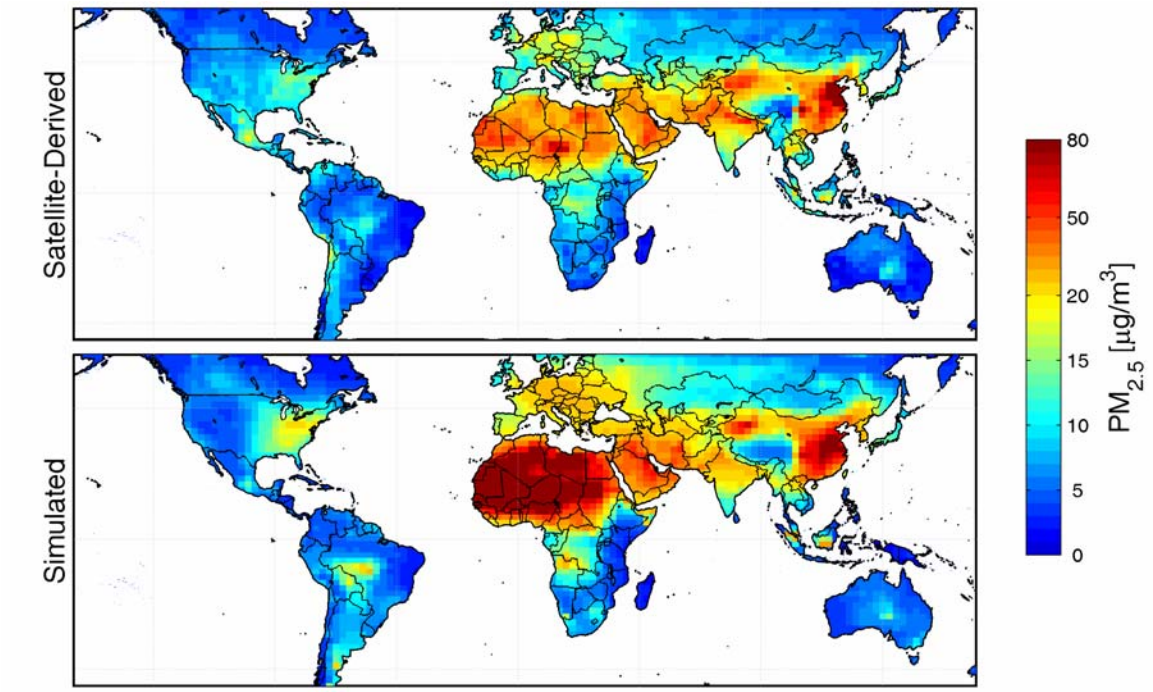

Figure S-4: Comparison of coincidently sampled satellite-estimated and simulated $PM_{2.5}$. Satellite-estimated $PM_{2.5}$ has been degraded to a resolution of 2º × 2.5º.