Supporting Information

Global Chemical Composition of Ambient Fine Particulate Matter for Exposure Assessment

Sajeev Philip[1*], Randall V. Martin[1, 2], Aaron van Donkelaar[1], Jason Wai-Ho Lo[1], Yuxuan Wang[3], Dan Chen[4], Lin Zhang[5], Prasad S. Kasibhatla[6], Siwen Wang[7], Qiang Zhang[8], Zifeng Lu[9], David G. Streets[9], Shabtai Bittman[10] and Douglas J. Macdonald[11]

[1]Department of Physics and Atmospheric Science, Dalhousie University, Halifax, Nova Scotia, Canada

[2]Also at Harvard-Smithsonian Center for Astrophysics, Cambridge, Massachusetts, USA

[3]Ministry of Education Key Laboratory for Earth System Modeling, Center for Earth System Science, Institute for Global Change Studies, Tsinghua University, Beijing, China

[4]Department of Atmospheric and Oceanic Sciences, University of California, Los Angeles, California, USA.

[5]Department of Atmospheric and Oceanic Sciences, School of Physics, Peking University, China

[6]Nicholas School of the Environment and Earth Sciences, Duke University, Durham, North Carolina, USA.

[7]State Key Joint Laboratory of Environment Simulation and Pollution Control, School of Environment, Tsinghua University, Beijing, China.

[8]Center for Earth System Science, Tsinghua University, Beijing, China.

[9]Decision and Information Sciences Division, Argonne National Laboratory, Argonne, IL, USA

[10]Agriculture and Agri-Food Canada, Agassiz, British Columbia, Canada

[11]Environment Canada, Canada

Author Information

Corresponding Author

[*]Phone: +19024941820; Fax: +19024945191; e-mail: philip.sajeev@dal.ca

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Description of the GEOS-Chem aerosol simulation

We used the GEOS-Chem global three-dimensional chemical transport model (version 9-01-03; http://geos-chem.org) to calculate the local conversion factors (ratio of component to AOD) coincident with each satellite observation of AOD. The GEOS-Chem uses assimilated meteorological data from the Goddard Earth Observing System (GEOS-5) at the NASA Global Modeling Assimilation Office (GMAO). The meteorological data includes instantaneous fields, surface variables (e.g., mixed layer depth) at a temporal resolution of 3 hours, and other variables at 6 hours. We reduced the stratospheric layers of the native GEOS-5 vertical grids (72 hybrid eta levels) to 48 for computational expediency. The vertical layers of the current model extend from the Earth's surface to the top of the atmosphere (0.01 hPa) with 47 vertical levels. The lowest layer of the model is centered at approximately 70 meters, and used here to represent the ground-level aerosol concentrations.

The native horizontal resolution for the GEOS-5 meteorological data is 0.5^o x 0.667^o. First, we used data regridded to a coarser resolution of 2^o x 2.5^o for computational expediency, and performed the global simulation at this resolution. Second, we conducted three regional (nested) simulations at the native horizontal resolution of 0.5^o x 0.667^o for three regions of the globe: North America (140^oW–40^oW, 10^oN–70^oN), Europe (30^oW–50^oE, 30^oN–70^oN) and East Asia (70^oW–150^oW, 11^oS–55^oN). This higher resolution simulation preserves the finer spatial patterns of the chemical components (1, 2). The global simulation outputs were used as boundary conditions for the regional grids. We spun up the model for one month before each global and regional simulation to remove the effects of initial conditions on the aerosol simulation. The dynamical processes (transport and convection) have a temporal resolution of 10 minutes for the nested simulations and 15 minutes for the global simulation. We used a timestep of 60 minutes for chemical processes and emissions for both nested and global resolutions. We used full mixing of species below the mixed layer, with a correction to the GEOS-5 predicted nocturnal mixed layer depth as described in Heald et al. (3) and Walker et al. (4).

GEOS-Chem simulates HO_x-NO_x-VOC-ozone-aerosol chemistry in detail (5, 6). Park et al. (6) describe the simulation of secondary inorganic ions coupled to gas phase chemistry. The simulation of aerosol-gas interactions are through the aerosol extinction effects on photolysis rates (7), and heterogeneous chemistry (8) with updated N_2O_5 (9) and HO_2 (10) uptake by aerosols. The ISORROPIA II thermodynamic scheme (11) is used for partitioning gases and aerosols (12). GEOS-Chem uses in-cloud sulfate formation using the cloud liquid water content and cloud volume fractions of the GEOS-5 data (13). We artificially limited the nitric acid to two thirds of its value for each timestep to correct for an overestimation in HNO_3 found in comparison with measurements over the eastern U.S. (3). GEOS-Chem calculates AOD based on

66 the relative humidity dependent aerosol optical properties as described in Martin et al. (7) with
67 an updated growth factor for organic matter, and updates to ammonium sulfate optics.
68 Modification to dust optics is described in Ridley et al. (14).

69 The GEOS-Chem simulation uses emission inventories of aerosol and its precursor gases as
70 input. We used regional anthropogenic emission inventories of NO_x and SO_2 over Canada (CAC;
71 http://www.ec.gc.ca/inrp-npri/), the U.S. (Environmental protection Agency-National Emissions
72 Inventory 2005; http://www.epa.gov/ttnchie1/net/2005inventory.html), Mexico (BRAVO;(15)),
73 Europe (EMEP; http://www.emep.int/), and East Asia ((16) for NO_x; (17) for SO_2. Elsewhere,
74 we used anthropogenic emissions from EDGAR v32-FT2000 global inventory for 2000 (18), and
75 scaled it based on the energy statistics to subsequent years (19). Anthropogenic NO_x emissions
76 were scaled from 2006 to subsequent years based on the NO_2 column density data retrieved from
77 the OMI satellite sensor (20). GEOS-Chem includes soil NO_x (21, 22), lightning NO_x (23-27),
78 ship SO_2 from the ICOADS inventory (28, 29), and volcanic emissions (13). Seasonality for NO_x
79 and SO_2 is based on the statistics from regional inventories. GEOS-Chem includes a diurnal
80 variation for NO_x as described in van Donkelaar et al. (19). Global ammonia emission in GEOS-
81 Chem is from Bouwman et al. (30) with a seasonality imposed by Park et al. (6). Spatial and
82 seasonal NH_3 variation over Canada is based on monthly varying agricultural activity statistics
83 provided by the Agriculture Canada (31). Other regional inventories are over the U.S. (EPA-
84 NEI), Europe (EMEP), and East Asia (32). East Asian annual emissions are superimposed with a
85 relative seasonal variation (13, 33), and a reduction of 30% (33) motivated by comparison with
86 other inventories (34, 35). We doubled NH_3 emissions over California as suggested by Heald et
87 al. (3) and Walker et al. (4).

GEOS-Chem carbonaceous aerosols include black carbon (BC), organic carbon (OC), and secondary organic aerosols (SOA) (3, 36, and 37). The global anthropogenic OC and BC inventory is from Bond et al. (38), with the Cooke et al. (39) inventory over North America (40), and the Lu et al. (17) inventory over East Asia. We doubled the East Asian OC and BC emissions based on a comparison with top-down inversion of regional inventories by Fu et al. (41) and recognize there is ongoing discussion on this topic (42). GEOS-Chem simulates the formation of SOA from the oxidation of volatile organic compounds (43-46). Global biomass burning emission is from the GFED-3 inventory at 3-day temporal resolution (47, 48), and global biofuel emission is from Yevich and Logan (49) superimposed with the regional inventories mentioned above. We calculated OM as the sum of model OC and SOA. We used the OM/OC ratio estimated from observations by the OMI satellite sensor and Aerosol Mass Spectrometer to convert OC to OM to account for the presence of non-carbon elements (50).

GEOS-Chem includes the simulation of natural particles such as mineral dust and sea salt. The mineral dust simulation is described by Fairlie et al. (51). We used the first dust size bin and 37% of the second dust size bin (out of the 4 bins) to get a $PM_{2.5}$ size range. Sea salt emission in the model is described by Alexander et al. (52) with updates by Jaegle et al. (53). We use sea salt accumulation mode size range from 0.1 to 1 μm, which in normal coastal conditions represents the approximate $PM_{2.5}$ size range.

GEOS-Chem includes dry deposition (22), and wet deposition (37, 54, 55).

Numerous studies have evaluated the GEOS-Chem ground-level aerosol concentrations and its seasonal variation (e.g., 3, 4, 6, 12, 36, 40, 41, 51, 56, and 57). Vertical profiles of aerosol composition were also compared with various aircraft observations (e.g., (19, 58, 59), and with

CALIOP satellite observations (60-62). Six-year coincident comparisons of GEOS-Chem and CALIOP suggest simulated near-surface to column extinction ratios are often within 25%, but can approach a factor of two in certain seasons and locations (61).

Description of the ground-based $PM_{2.5}$ composition measurements

We utilized filter-based in situ measurements by several networks, such as, the National Air Pollution Surveillance Network (NAPS) and the Canadian Air and Precipitation Monitoring Network (CAPMoN) over Canada (http://www.on.ec.gc.ca/natchem). The U.S. measurement networks include the Clean Air Status and Trends Network (CASTNET, http://java.epa.gov/castnet/epa_jsp/sites.jsp), the Interagency Monitoring of Protected Visual Environments (IMPROVE, http://vista.cira.colostate.edu/improve/), and the U.S. Environmental Protection Agency Air Quality System (EPA-AQS, http://www.epa.gov/ttn/airs/airsaqs/). The NAPS network provides 24-hr composition every third day across Canada as described in Dabek-Zlotorzynska et al. (63). We used weekly average sulfate and ammonium ion measurements from the CAPMoN and the CASTNET networks even though they are devoid of $PM_{2.5}$ filters (64). The IMPROVE network provides 24-hr $PM_{2.5}$ composition data except for ammonium for every third day from several national parks in the U.S. The EPA-AQS network mainly operates over rural areas which report 24-hr averages of all the major composition measurements every consecutive third or sixth day. Here, we used the $PM_{2.5}$ composition data from EPA-AQS and IMPROVE networks (2005-2008 mean) reported by Hand et al. (65). We calculated ammonium from sulfate and nitrate measurements of EPA-AQS and IMPROVE networks by assuming a fully neutralized sulfuric acid by ammonia gas. We averaged the data reported by Hand et al. (65), and the long-term (2004-2008) in situ

132 measurements from other networks into the 0.1^0 x 0.1^0 grid for comparison with satellite-model
133 composition over North America.

134 We treated these in situ data as 'truth' to evaluate our product. However, it is worth noting some
135 uncertainties. Carbon measurements are prone to errors due to filter contamination (e.g., 66). The
136 ratio of OM to OC varies from 1.2 to 2.6 depending on the spatial and seasonal differences (e.g.,
137 67, 68). Mineral dust concentrations were from the elemental measurements of IMPROVE and
138 EPA-AQS following Malm et al. (69) even though measurements of five elements alone are
139 inadequate to determine the ambient mineral dust (65, 70). Sea salt were from elemental chlorine
140 or chlorine ion measurements, by accounting for 55% chlorine by weight; the selection of sea
141 salt marker as sodium or chlorine is also uncertain (65, 71). Hand et al. (72) obtained relative
142 errors for $PM_{2.5}$ and its chemical components from a comparison of collocated IMPROVE and
143 AQS measurements (17% for $PM_{2.5}$, 5% for ammonium sulfate, 11% for ammonium nitrate, 10%
144 for OC, 12% for EC, 33% for dust, and 77% for sea salt). Finally, the gridded in situ data are
145 prone to representation error as an individual measurement is used to represent a 10 km x 10 km
146 area.

147 In addition, we collected annually representative inorganic and organic composition
148 measurements from the European Monitoring and Evaluation Programme (EMEP;
149 http://www.emep.int/), the Acid Deposition Monitoring Network in East Asia (EANET;
150 http://www.eanet.cc/), and several field measurements around the world from published papers
151 e.g., (41, 73, 74) (for organic measurements) and several others (Table S2). We used this dataset
152 to evaluate the global satellite-model composition. We included only sites with all SIA
153 components to achieve a consistent evaluation.

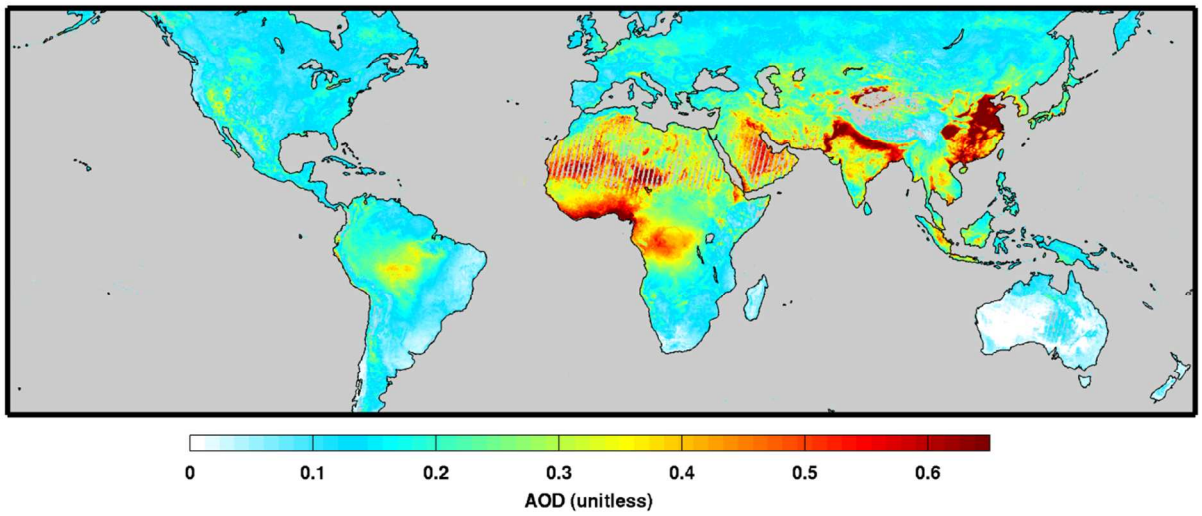
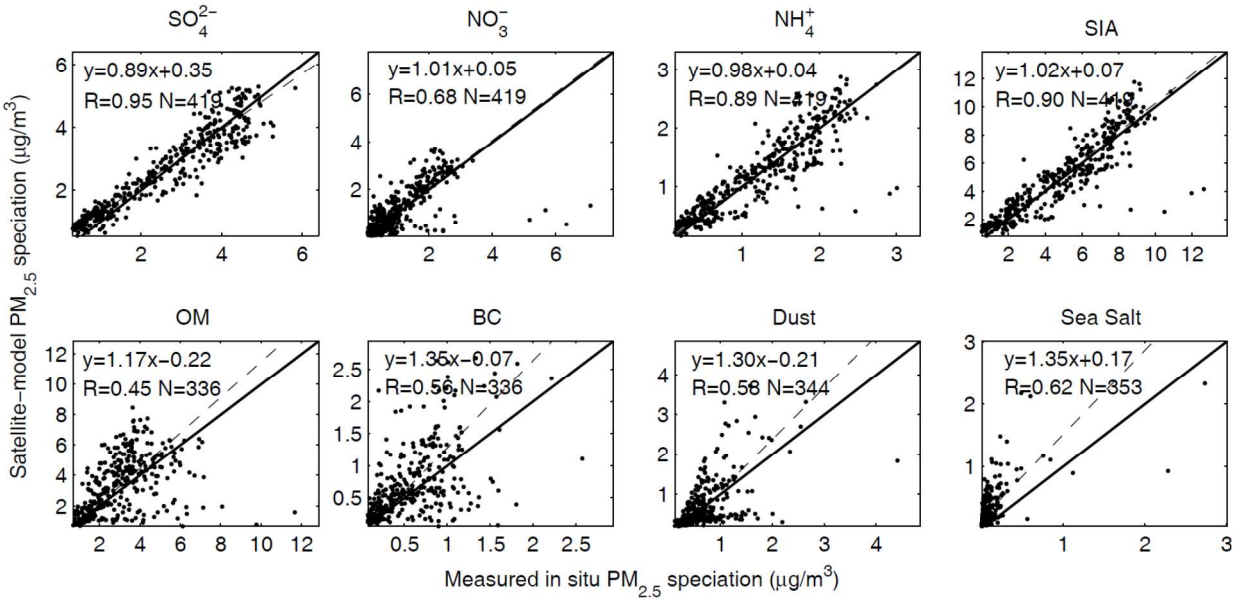


Figure S1. Combined aerosol optical depth (AOD) from the MODIS and MISR satellite instruments for 2004-2008. Gray denotes water or missing satellite AOD observations.



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167 **Figure S2.** Comparison of $PM_{2.5}$ composition from the satellite-model product versus in situ
168 observations across North America. The solid black line is the 1:1 line, and the dashed line is the
169 best fit line. The inset contains the Pearson correlation coefficient as well as the slope and
170 intercept from reduced major axis regression.

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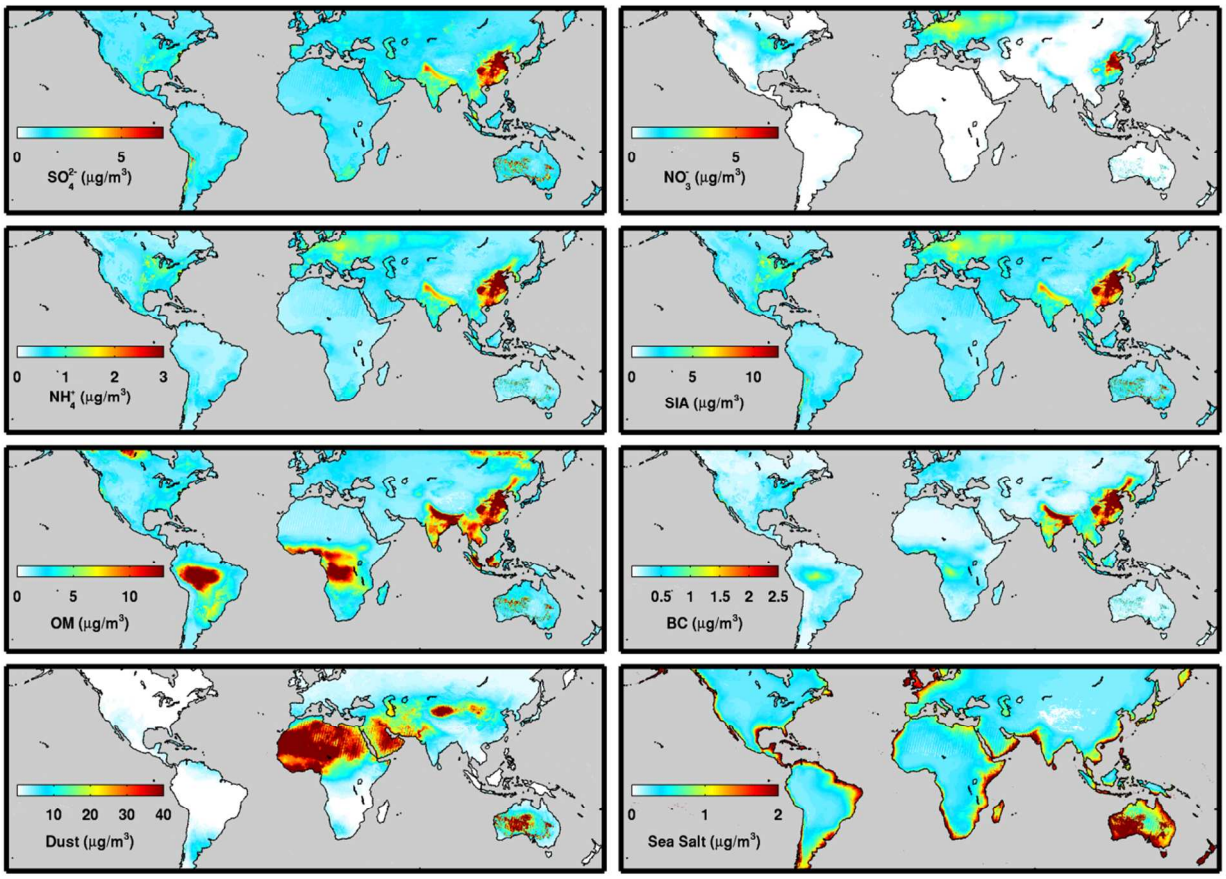

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179 **Figure S3.** Absolute uncertainty of satellite-model $PM_{2.5}$ composition determined by propagation

180 of error. Gray denotes water.

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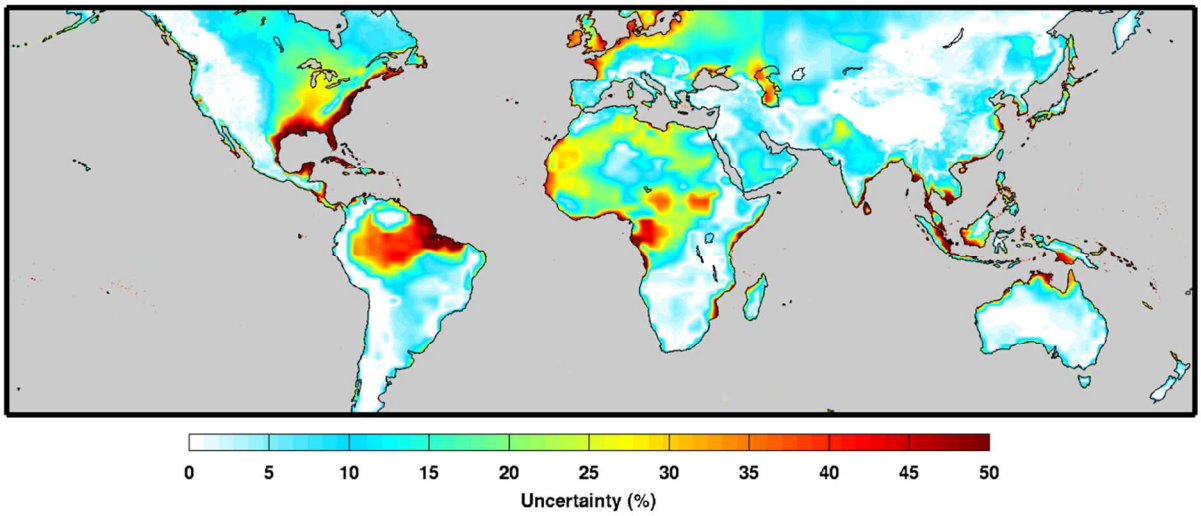

Figure S4. Uncertainty in $PM_{2.5}$ due to GEOS-Chem model vertical profile bias determined by comparison with CALIOP satellite observations of aerosol extinction following van Donkelaar et al. (61). Gray denotes water.

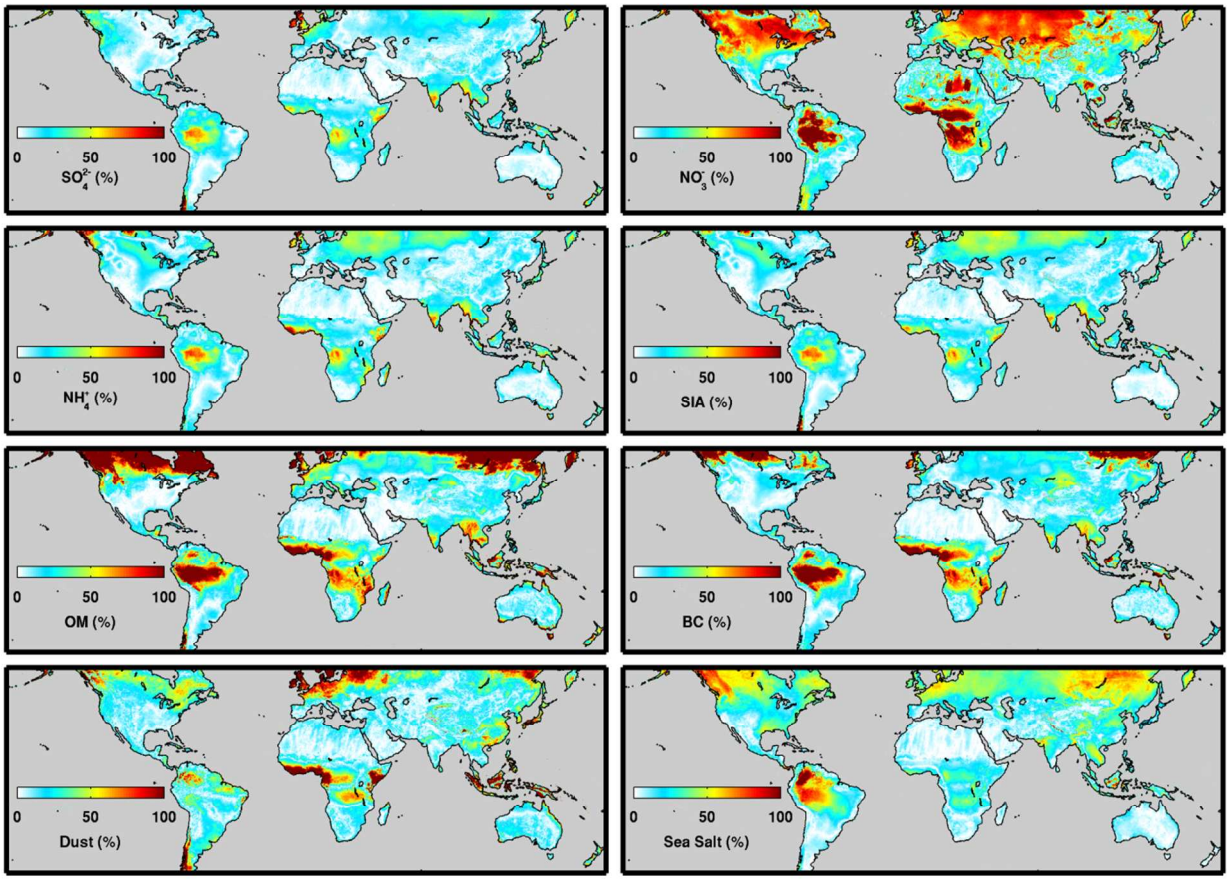

Figure S5. Uncertainty in satellite-model $PM_{2.5}$ composition due to incomplete sampling estimated as the difference between the long-term mean simulated $PM_{2.5}$ composition sampled continuously versus coincidently with the satellite observations. Gray denotes water.

$PM_{2.5}$ Composition	Data	North America				Global			
		r	Slope	Offset ($\mu g/m^3$)	N	r	Slope	Offset ($\mu g/m^3$)	N
SO_4^{2-}	Satellite	0.95	0.89	0.35	419	0.92	0.88	0.93	55
	Simulation	0.97	0.73	0.23	419	0.94	0.61	0.80	55
NO_3^-	Satellite	0.68	1.01	0.05	419	0.73	0.83	-0.26	55
	Simulation	0.63	0.80	0.11	419	0.69	0.64	-0.24	55
NH^+	Satellite	0.89	0.98	0.04	419	0.92	1.08	0.13	55
	Simulation	0.89	0.79	0.04	419	0.93	0.77	0.12	55
SIA	Satellite	0.90	1.02	0.07	419	0.93	0.91	0.85	55
	Simulation	0.90	0.82	0.10	419	0.92	0.64	0.70	55
OM	Satellite	0.45	1.17	-0.22	336	0.67	0.42	-0.73	56
	Simulation	0.42	0.99	-0.34	336	0.61	0.30	-0.47	56
BC	Satellite	0.56	1.35	-0.07	336	0.65	1.05	-1.83	65
	Simulation	0.53	1.04	-0.04	336	0.56	0.68	-1.15	65

Table S1. Comparison of $PM_{2.5}$ composition from the simulation and satellite-model product versus in situ observations across North America, and globally (non-North American). Abbreviations are Secondary Inorganic Aerosol (SIA; the sum of SO_4^{2-}, NO_3^- and NH_4^+), Organic Mass (OM), and Black Carbon (BC). "Satellite" and "Simulation" in the second column represents satellite-model composition, and complete model simulation respectively at a spatial resolution of 0.1o x 0.1o. Correlation statistics are calculated with reduced major axis regression.

Sl. No.	Site/City	Country	Latitude	Longitude	Study Period	SO_4^{2-}	NO_3^-	NH_4^+	OC	BC	Size	Source
			degree	degree		μg/m^3	μg/m^3	μg/m^3	μg/m^3	μg/m^3		
1	Beijing	China	40.3	116.3	Mar 2005 - Feb 2006	15.8	10.1	7.3	34.4	8.3	$PM_{2.5}$	75
2	Miyun Resevoir	China	40.5	116.8	Mar 2005 - Feb 2006	13.0	6.4	6.1	21.9	3.8	"	
3	Chongqing	China	29.6	106.5	Mar 2005 - Feb 2006	25.5	5.3	7.9	42.2	6.5	"	
4	Dadukou	China	29.5	106.5	Mar 2005 - Feb 2006	23.4	5.1	7.6	47.2	6.4	"	
5	Jinyun	China	29.8	106.4	Mar 2005 - Feb 2006	24.0	4.8	7.3	41.2	4.7	"	
6	Hok Tsui	Hong Kong	22.2	114.3	Nov 2004 - Oct 2005	11.9	0.8	3.1	4.3	2.1	"	76
7	Tsuen Wan	Hong Kong	22.4	114.1	Nov 2004 - Oct 2005	13.2	1.6	4.1	7.4	6.0	"	"
8	Mong Kok	Hong Kong	22.3	114.1	Nov 2004 - Oct 2005	12.8	2.4	4.4	11.9	13.7	"	"
9	Delhi	India	28.4	77.1	Mar 2001 - Jan 2002	10.9	6.1	6.4	40.3	10.5	"	77
10	Mumbai	India	22.6	88.3	Mar 2001 - Jan 2002	6.9	1.6	2.0	12.6	4.6	"	"
11	Kolkata	India	18.7	72.8	Mar 2001 - Jan 2002	7.2	2.9	3.6	37.1	12.1	"	"
12	Lahore	Pakistan	31.5	74.3	Jan 2007 - Jan 2008	10.5	6.6	3.6	64.4	11.2	"	78
13	Brazil	Brazil	23.0	43.2	Sep 2003 - Sep 2004	1.5	0.3	0.6	NA	2.1	"	79
14	Brazil	Brazil	22.9	43.2	Sep 2003 - Sep 2004	1.9	0.4	0.7	NA	3.1	"	"
15	Brazil	Brazil	22.9	43.2	Sep 2003 - Sep 2004	2.2	0.5	0.7	NA	2.9	"	"
16	Brazil	Brazil	23.0	43.4	Sep 2003 - Sep 2004	1.4	0.3	0.4	NA	1.3	"	"
17	Brazil	Brazil	23.0	43.3	Sep 2003 - Sep 2004	1.8	0.5	0.5	NA	2.7	"	"
18	Brazil	Brazil	22.8	43.4	Sep 2003 - Sep 2004	1.5	0.4	0.5	NA	3.2	"	"
19	Brazil	Brazil	23.0	43.6	Sep 2003 - Sep 2004	1.5	0.4	0.5	NA	1.2	"	"
20	Brazil	Brazil	22.9	43.6	Sep 2003 - Sep 2004	1.7	0.4	0.6	NA	2.4	"	"
21	Brazil	Brazil	22.9	43.4	Sep 2003 - Sep 2004	1.7	0.4	0.6	NA	2.5	"	"
22	Brazil	Brazil	22.9	43.7	Sep 2003 - Sep 2004	1.7	0.3	0.5	NA	1.7	"	"
23	Tel Aviv	Israel	32.1	34.8	Jan 2007 - Dec 2007	5.2	0.7	NA	4.7	1.5	"	80
24	Haifa	Israel	32.8	35.0	Jan 2007 - Dec 2007	5.2	1.4	NA	3.0	1.0	"	"
25	W. Jerusalem	Israel	31.8	35.2	Jan 2007 - Dec 2007	4.6	1.0	NA	4.1	1.1	"	"
26	Hebron	Palestine	31.5	35.1	Jan 2007 - Dec 2007	3.8	0.9	NA	5.3	1.8	"	"
27	E. Jerusalem	Palestine	31.8	35.2	Jan 2007 - Dec 2007	4.4	0.9	NA	5.2	2.2	"	"
28	Nablus	Palestine	32.2	35.2	Jan 2007 - Dec 2007	4.3	1.0	NA	8.2	5.6	"	"
29	Amman	Jordan	32.0	35.8	Jan 2007 - Dec 2007	4.5	1.0	NA	6.2	2.4	"	"
30	Balearic islands	Spain	39.6	2.6	Jan 2004 - Feb 2005	3.9	0.9	2.0	2.9	0.5	"	81
31	Kuwait	Kuwait	29.3	48.0	Feb 2004 - Jan 2005	9.9	1.8	NA	3.7	2.5	"	82
32	Chengdu	China	30.7	104.0	Jan 2006 - Dec 2007	40.5	NA	14.0	36.3	10.8	PM_{10}	83
33	Dalian	China	38.9	121.6	Jan 2006 - Dec 2007	23.3	NA	7.7	20.2	5.3	"	"
34	Dunhuang	China	40.2	94.7	Jan 2006 - Dec 2007	6.6	NA	0.4	26.7	3.6	"	"
35	Gaolanshan	China	36.0	105.9	Jan 2006 - Dec 2007	16.7	NA	6.5	19.1	3.8	"	"
36	Gucheng	China	39.1	115.8	Jan 2006 - Dec 2007	35.5	NA	14.4	38.5	11.0	"	"
37	Jinsha	China	29.6	114.2	Jan 2006 - Dec 2007	26.6	NA	7.6	15.3	3.0	"	"
38	Lhasa	China	29.7	91.1	Jan 2006 - Dec 2007	2.9	NA	0.2	21.7	3.8	"	"
39	LinAn	China	30.3	119.7	Jan 2006 - Dec 2007	21.7	NA	6.8	15.1	4.3	"	"
40	Longfengshan	China	44.7	127.6	Jan 2006 - Dec 2007	10.0	NA	2.5	15.9	2.3	"	"
41	Nanning	China	22.8	108.4	Jan 2006 - Dec 2007	21.6	NA	5.8	17.9	4.0	"	"
42	Panyu	China	23.0	113.4	Jan 2006 - Dec 2007	26.8	NA	8.6	22.3	7.9	"	"
43	Taiyangshan	China	29.2	111.7	Jan 2006 - Dec 2007	28.8	NA	7.9	13.8	2.7	"	"
44	Xian	China	34.4	109.0	Jan 2006 - Dec 2007	46.7	NA	14.4	42.6	12.7	"	"
45	Zhengzhou	China	34.8	113.7	Jan 2006 - Dec 2007	45.0	NA	16.5	29.2	9.2	"	"
46	Akdala	China	47.1	88.0	Jul 2004 - Mar 2005	3.3	NA	0.6	2.9	0.4	"	84,85
47	Shangri-La, Zhuzhang	China	28.0	99.7	Jul 2004 - Mar 2005	1.6	NA	0.2	3.1	0.3	"	"

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221 **Table S2.** Global in situ data collected from publications. "NA" represents data not available or

222 filtered out.

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