

Figure S1: Modeled (background) and observed (circles) PM_{2.5} annual mean concentrations in µg/m³ for a. Africa, b. Asia, c. Australia, d. Europe, e. N. America and f. S. America. These show the exact same information as in Figure 3a but in subregions.

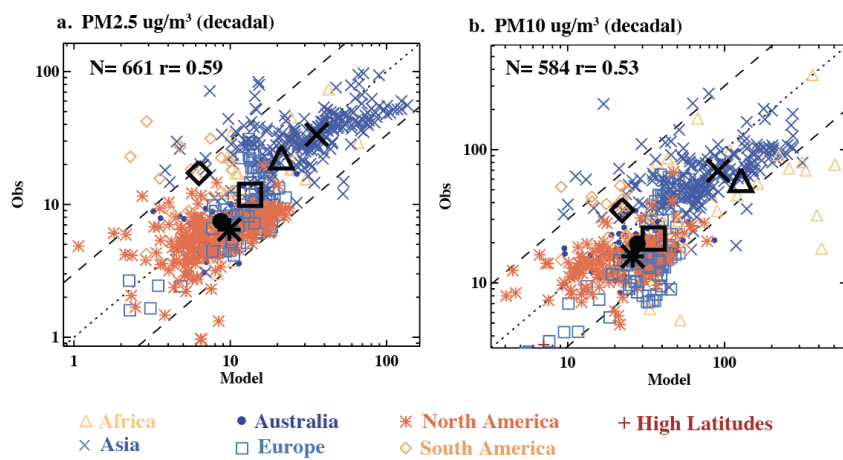


Figure S2: Scatterplots showing the model-data comparison of PM2.5 (a) and PM10 (b) using the decadal 2010-2019 averages instead of the climatological averages of the observations (otherwise the same as Figure 3d and 6c).

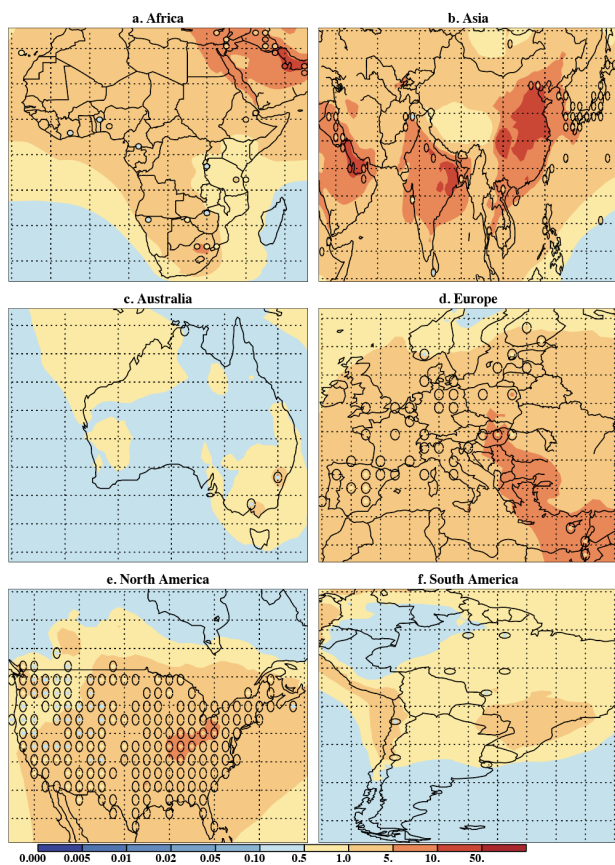


Figure S3: Modeled (background) and observed (circles) $\text{SO}_4 \text{PM}_{2.5}$ annual mean concentrations in $\mu\text{g}/\text{m}^3$ for a. Africa, b. Asia, c. Australia, d. Europe, e. N. America and f. S. America These show the exact same information as in Figure 5a but in subregions.

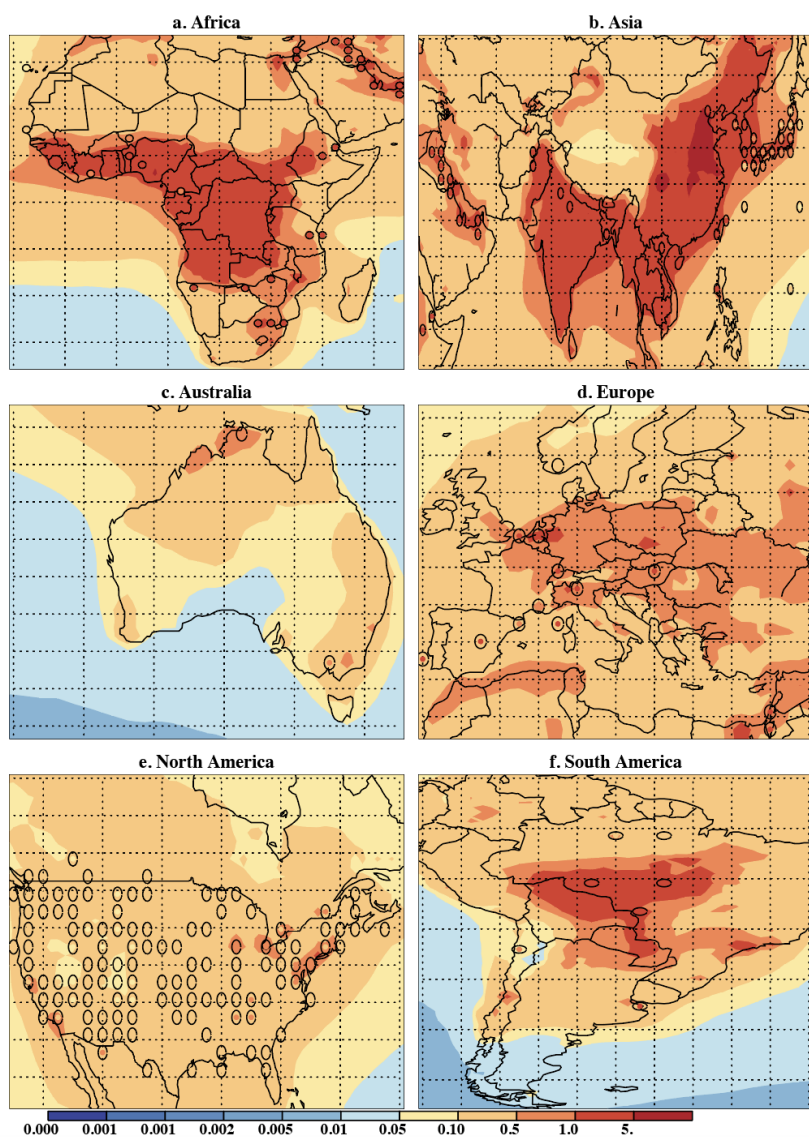


Figure S4: Modeled (background) and observed (circles) BC PM_{2.5} annual mean concentrations in µg/m³ for a. Africa, b. Asia, c. Australia, d. Europe, e. N. America and f. S. America. These show the exact same information as in Figure 5c but in subregions.

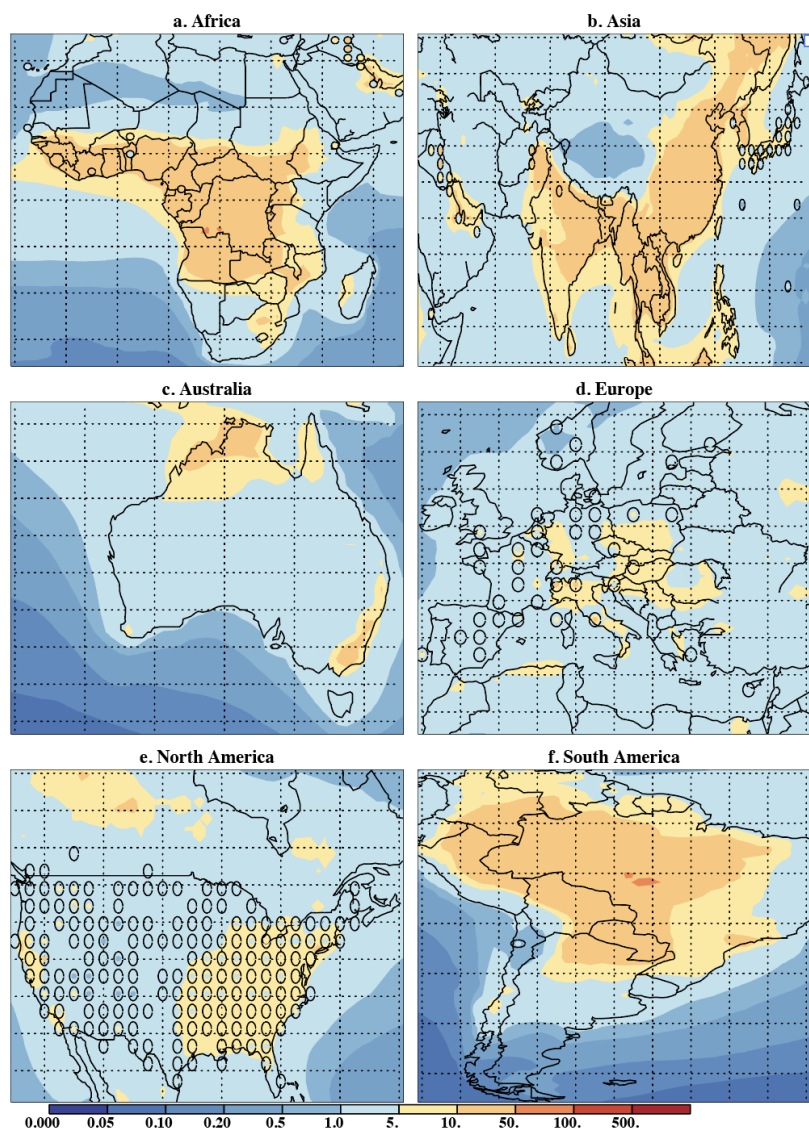


Figure S5: Modeled (background) and observed (circles) OM PM_{2.5} annual mean concentrations in µg/m³ for a. Africa, b. Asia, c. Australia, d. Europe, e. N. America and f. S. America These show the exact same information as in Figure 5e but in subregions.

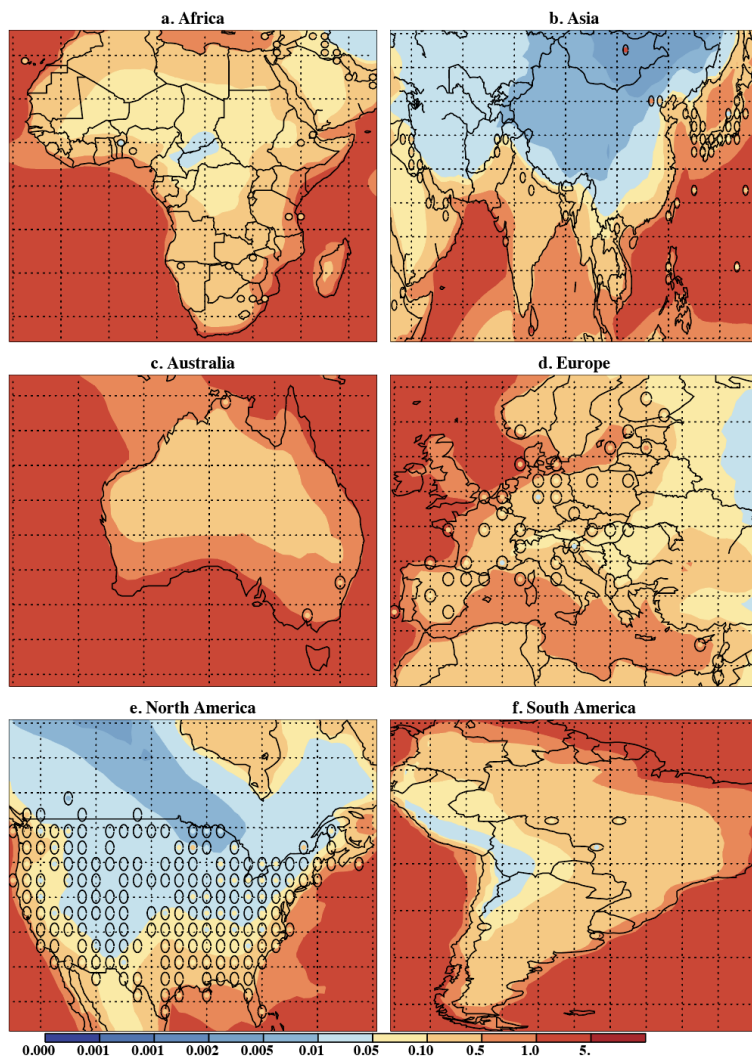


Figure S6: Modeled (background) and observed (circles) Na PM_{2.5} annual mean concentrations in µg/m³ for a. Africa, b. Asia, c. Australia, d. Europe, e. N. America and f. S. America. These show the exact same information as in Figure 5g but in subregions.

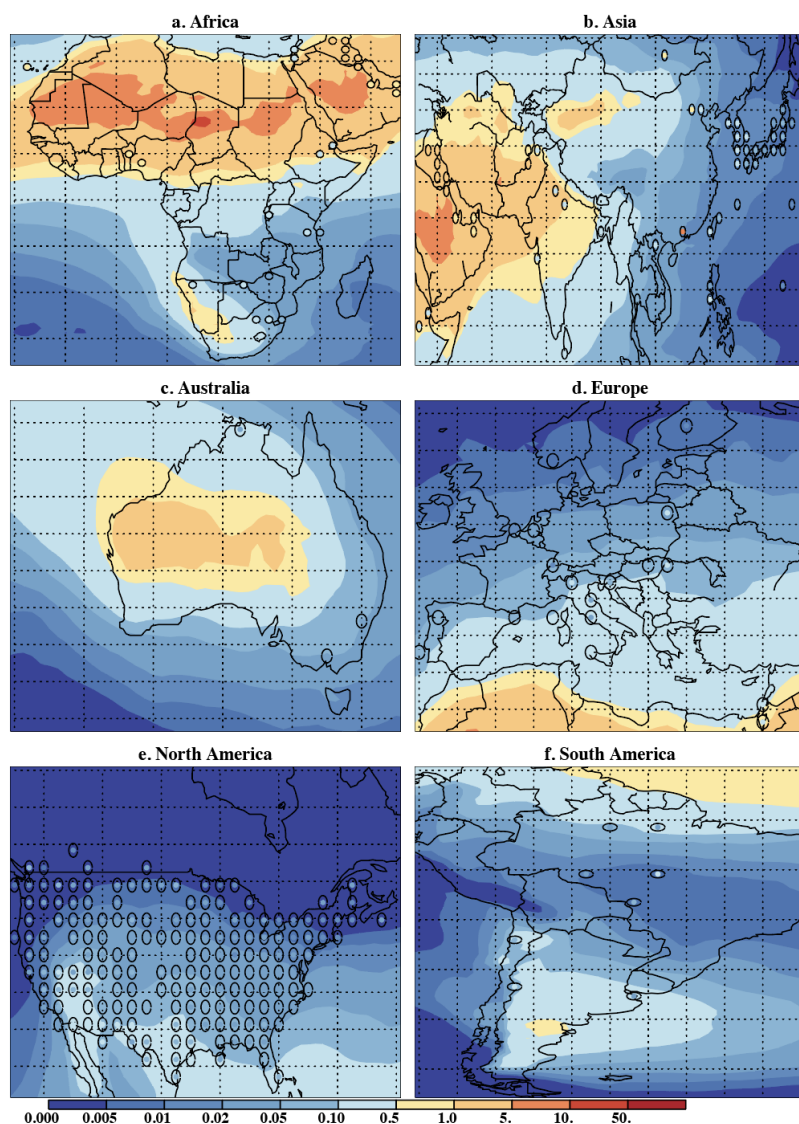


Figure S7: Modeled (background) and observed (circles) Al PM_{2.5} annual mean concentrations in µg/m³ for a. Africa, b. Asia, c. Australia, d. Europe, e. N. America and f. S. America. These show the exact same information as in Figure 5i but in subregions.

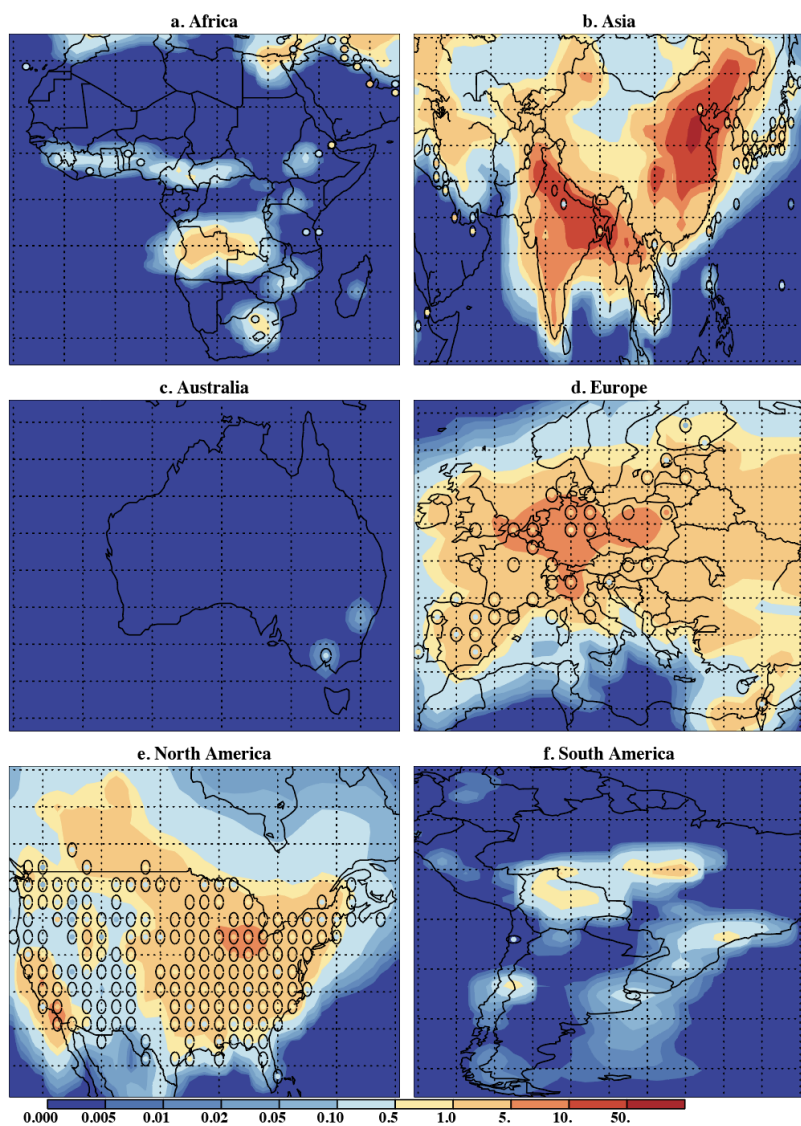


Figure S8: Modeled (background) and observed (circles) $\text{NO}_3 \text{PM}_{2.5}$ annual mean concentrations in $\mu\text{g}/\text{m}^3$ for a. Africa, b. Asia, c. Australia, d. Europe, e. N. America and f. S. America. These show the exact same information as in Figure 5k but in subregions.

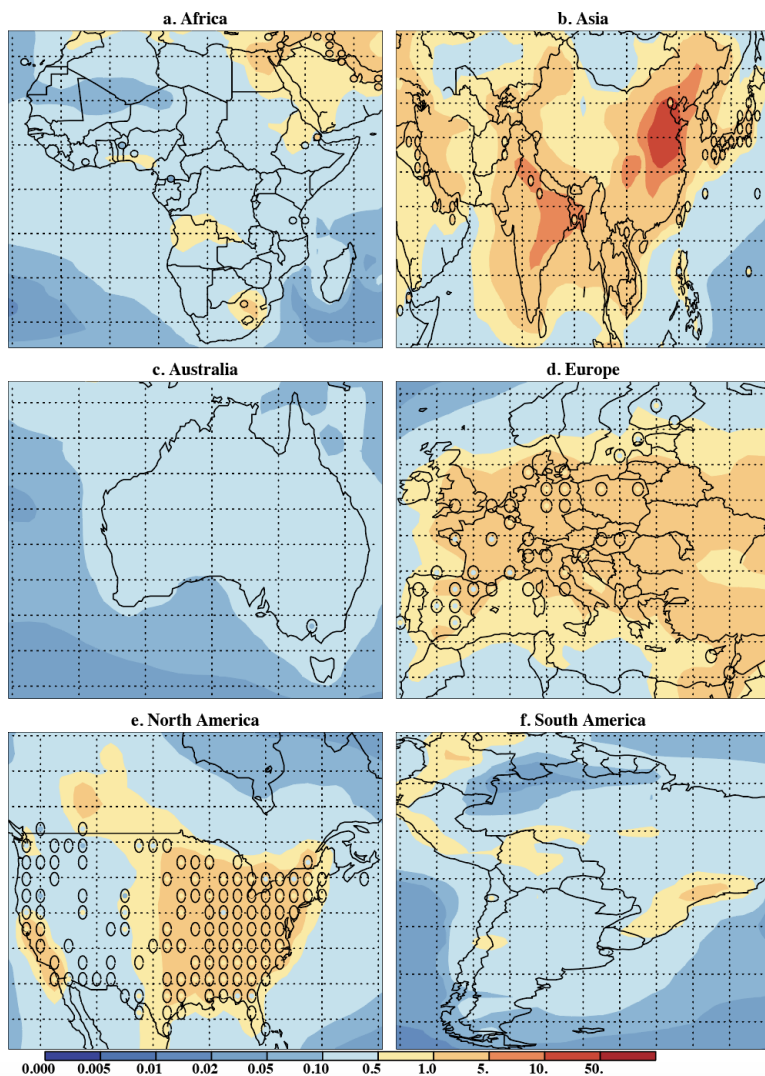


Figure S9: Modeled (background) and observed (circles) $\text{NH}_4 \text{PM}_{2.5}$ annual mean concentrations in $\mu\text{g}/\text{m}^3$ for a. Africa, b. Asia, c. Australia, d. Europe, e. N. America and f. S. America These show the exact same information as in Figure 51 but in subregions.

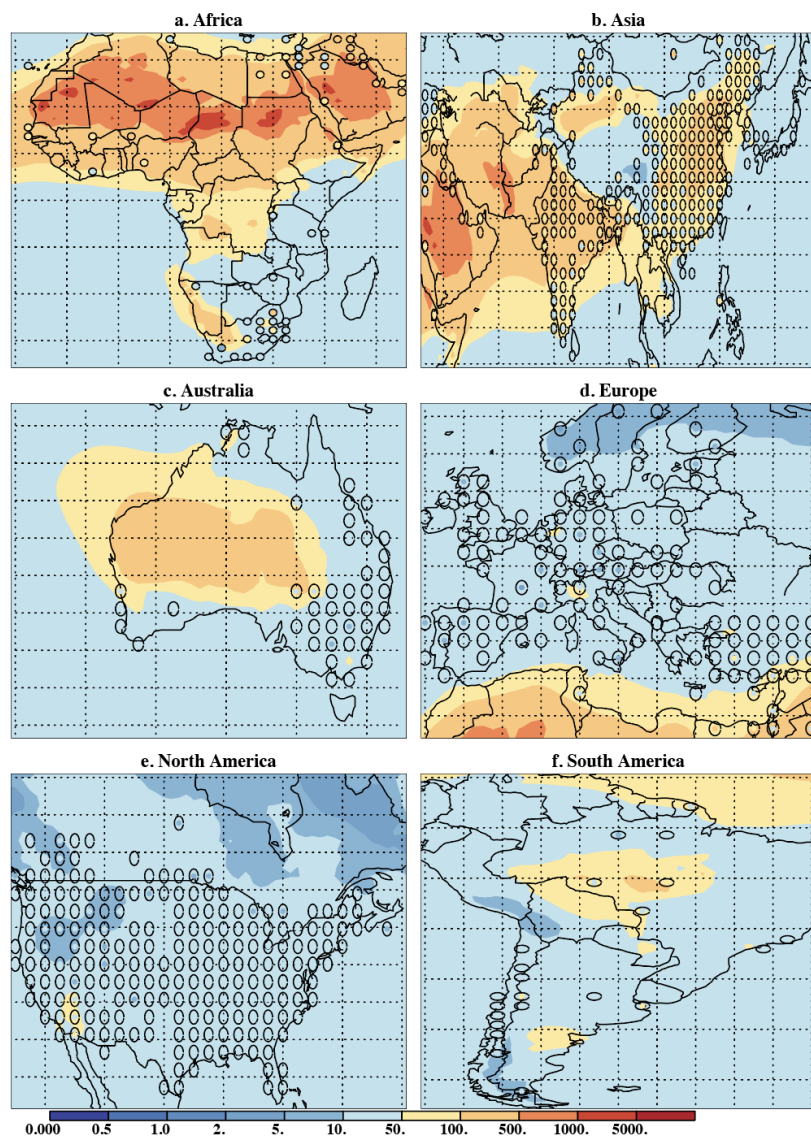


Figure S10: Modeled (background) and observed (circles) PM₁₀ annual mean concentrations in µg/m³ for a. Africa, b. Asia, c. Australia, d. Europe, e. N. America and f. S. America. These show the exact same information as in Figure 6a but in subregions.

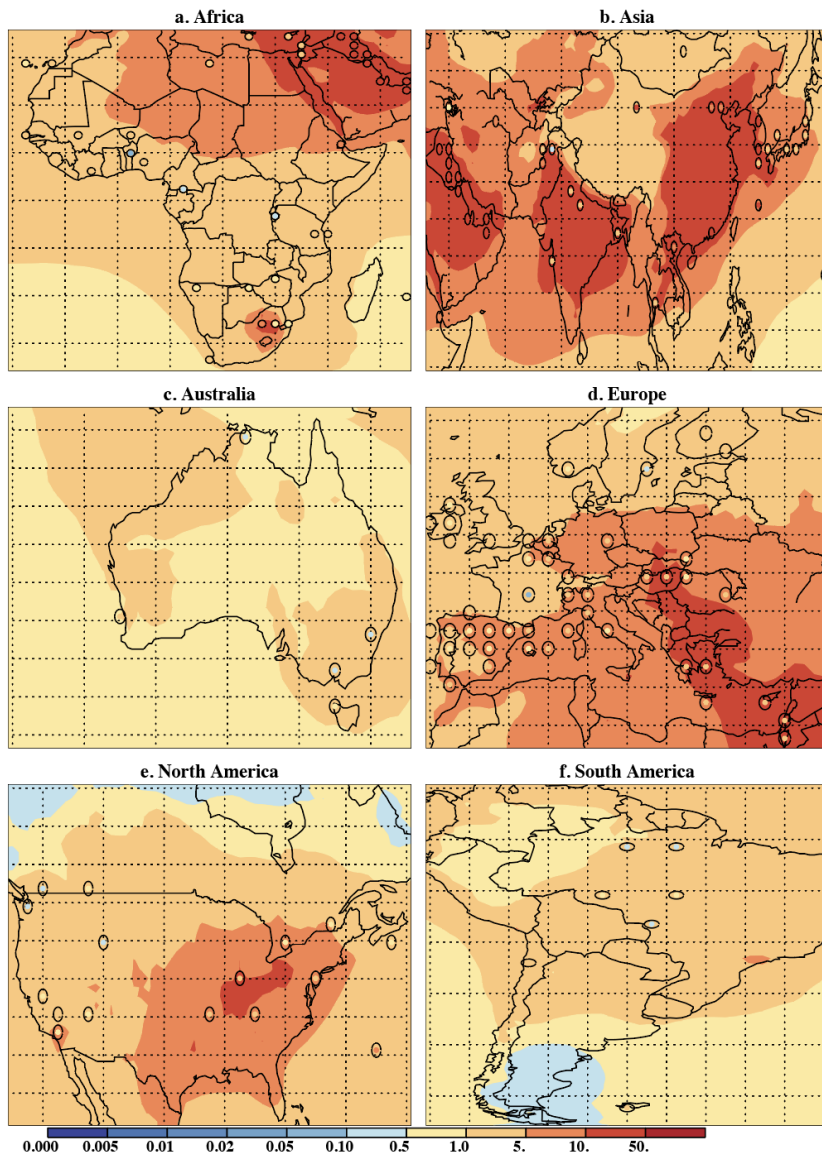


Figure S11: Modeled (background) and observed (circles) $\text{SO}_4 \text{PM}_{10}$ annual mean concentrations in $\mu\text{g}/\text{m}^3$ for a. Africa, b. Asia, c. Australia, d. Europe, e. N. America and f. S. America. These show the exact same information as in Figure 6a but in subregions.

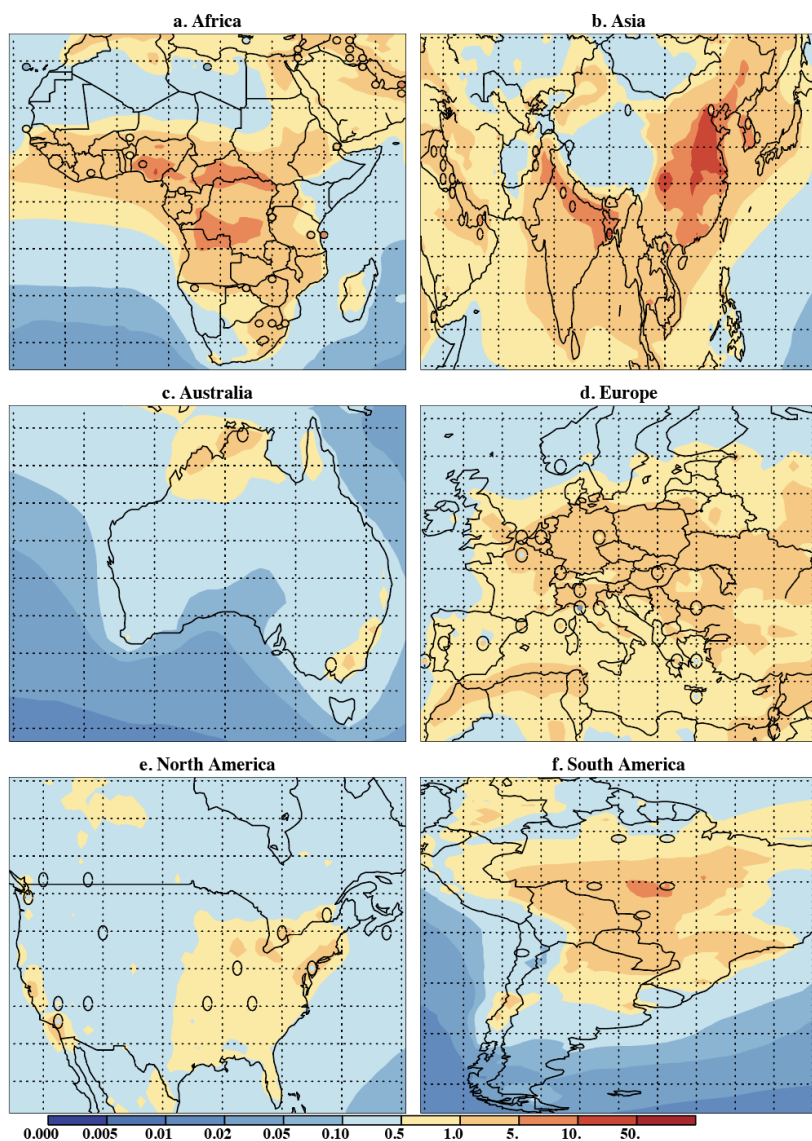


Figure S12: Modeled (background) and observed (circles) BC PM₁₀ annual mean concentrations in µg/m³ for a. Africa, b. Asia, c. Australia, d. Europe, e. N. America and f. S. America These show the exact same information as in Figure 6c but in subregions.

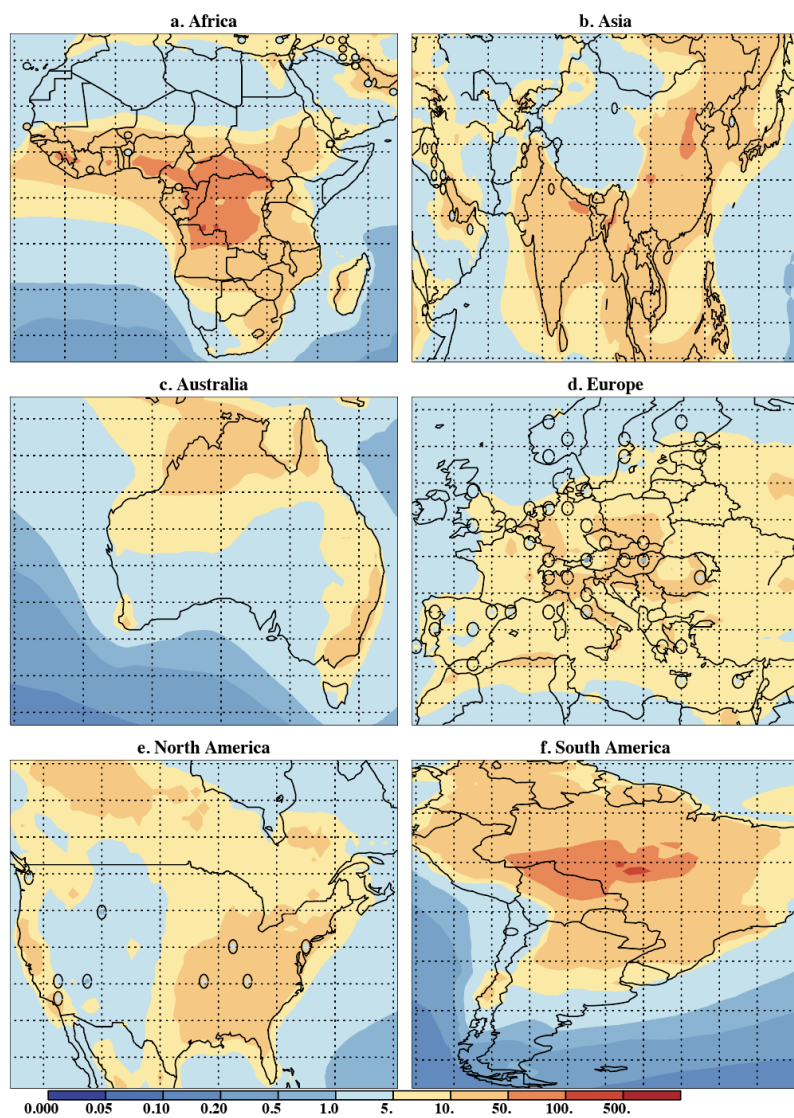


Figure S13: Modeled (background) and observed (circles) OM PM₁₀ annual mean concentrations in µg/m³ for a. Africa, b. Asia, c. Australia, d. Europe, e. N. America and f. S. America. These show the exact same information as in Figure 7e but in subregions.

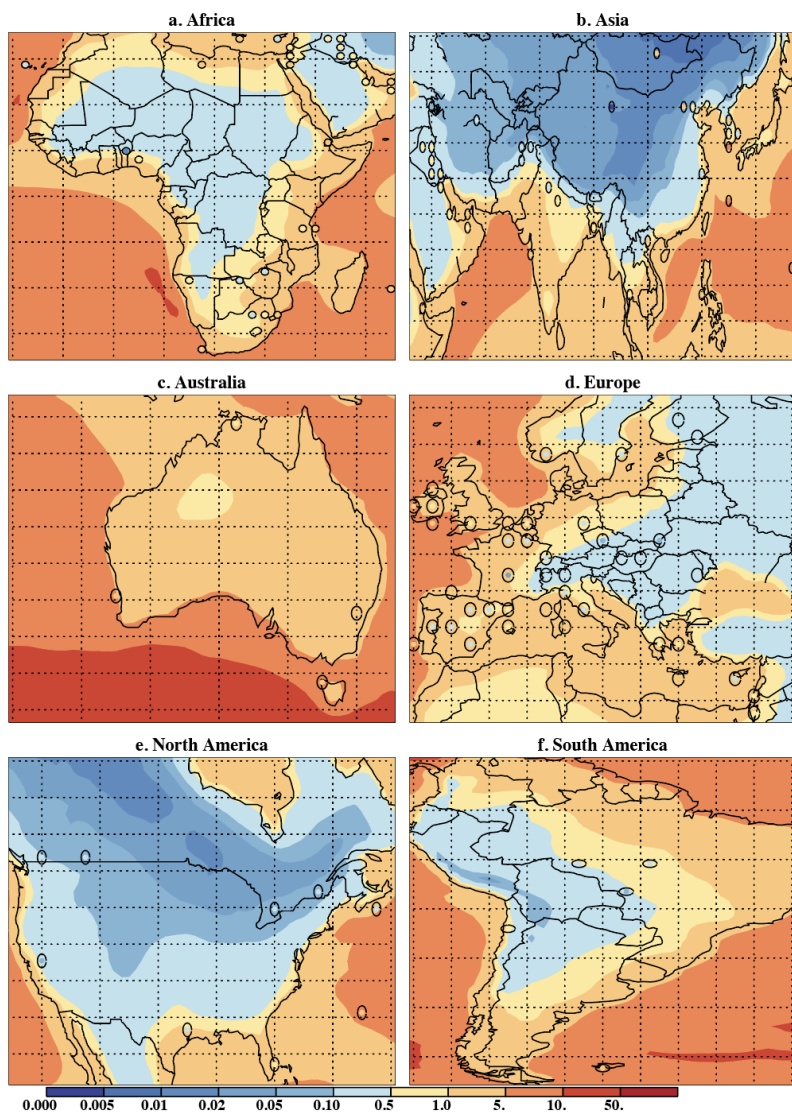


Figure S14: Modeled (background) and observed (circles) Na PM₁₀ annual mean concentrations in µg/m³ for a. Africa, b. Asia, c. Australia, d. Europe, e. N. America and f. S. America. These show the exact same information as in Figure 7g but in subregions.

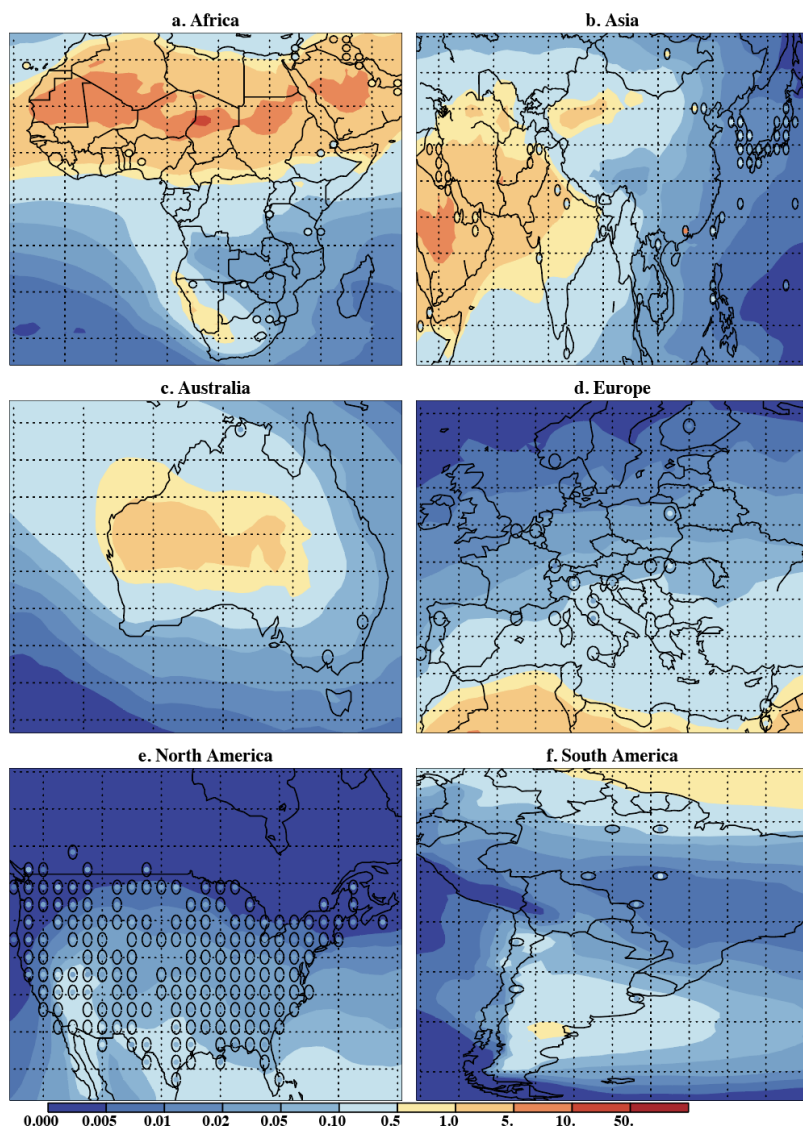


Figure S15: Modeled (background) and observed (circles) Al PM₁₀ annual mean concentrations in µg/m³ for a. Africa, b. Asia, c. Australia, d. Europe, e. N. America and f. S. America These show the exact same information as in Figure 7i but in subregions.

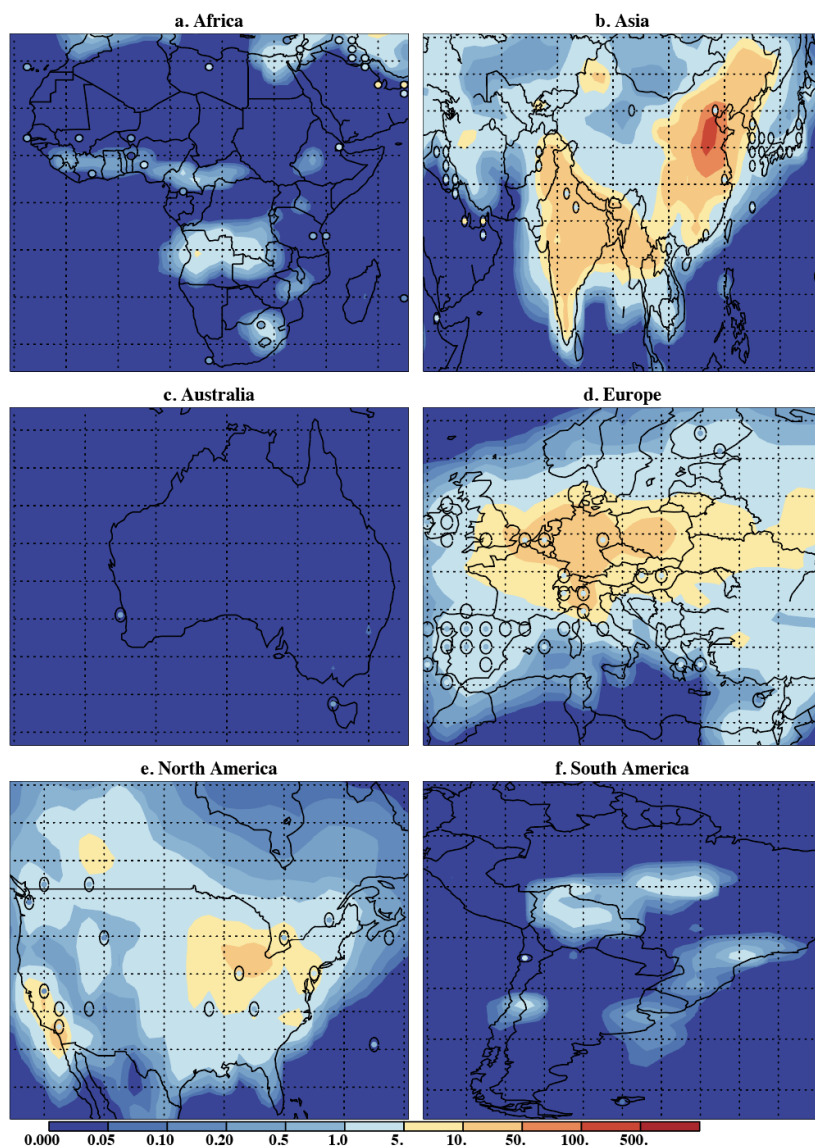


Figure S16: Modeled (background) and observed (circles) NO_3 PM_{10} annual mean concentrations in $\mu\text{g}/\text{m}^3$ for a. Africa, b. Asia, c. Australia, d. Europe, e. N. America and f. S. America. These show the exact same information as in Figure 7k but in subregions.

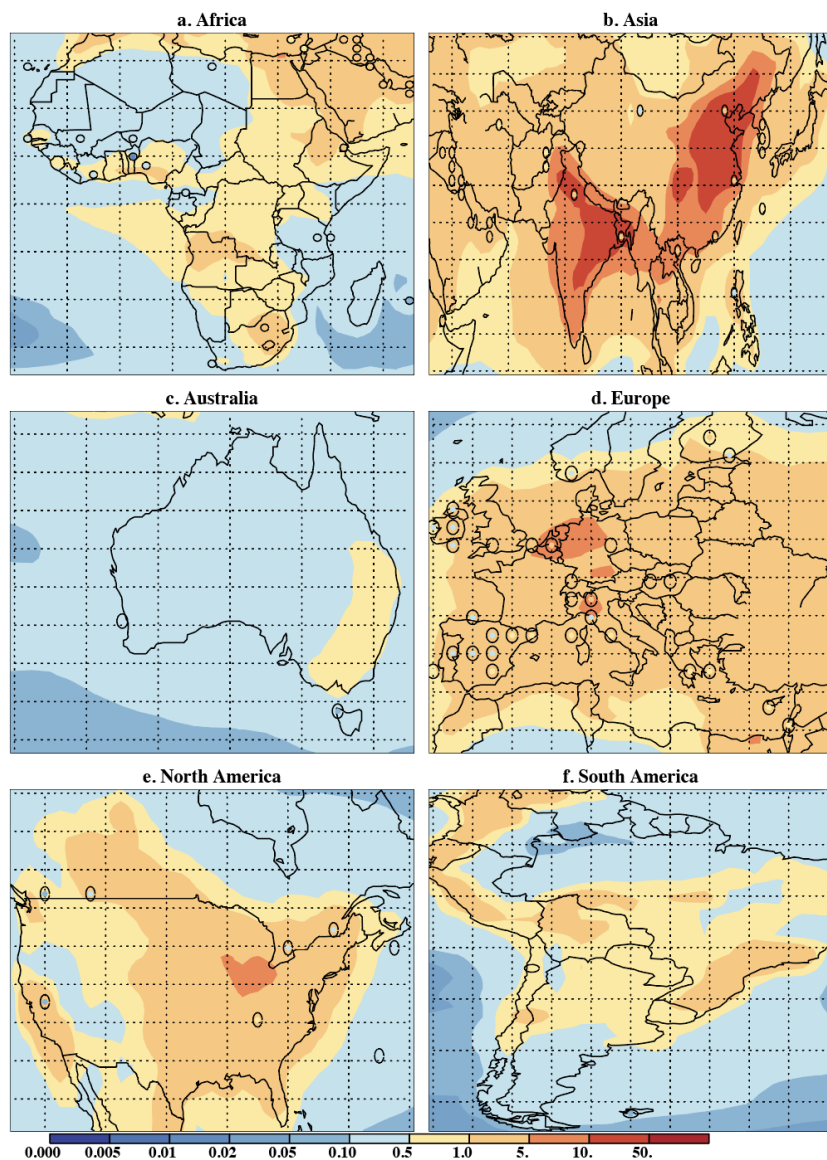


Figure S17: Modeled (background) and observed (circles) NO_3 PM_{10} annual mean concentrations in $\mu\text{g}/\text{m}^3$ for a. Africa, b. Asia, c. Australia, d. Europe, e. N. America and f. S. America. These show the exact same information as in Figure 71 but in subregions.

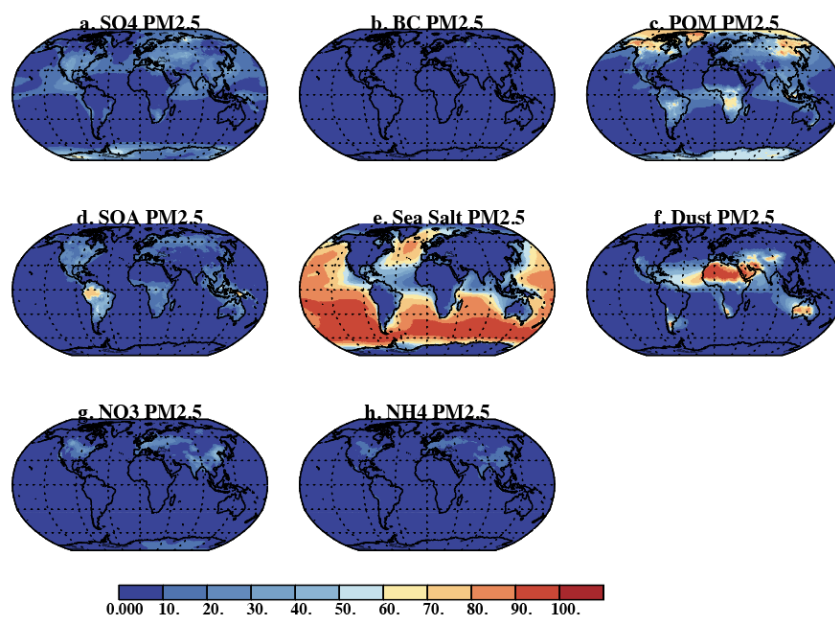


Figure S18: The modeled relative contribution (%) of different aerosol types to the total surface concentration in the $PM_{2.5}$ aerosol for a) SO_4 , b) BC, c) Primary Organic Material (POM), d) Secondary Organic Aerosol (SOA), e) Sea salt, f) dust, g) NO_3 and h) NH_4 .

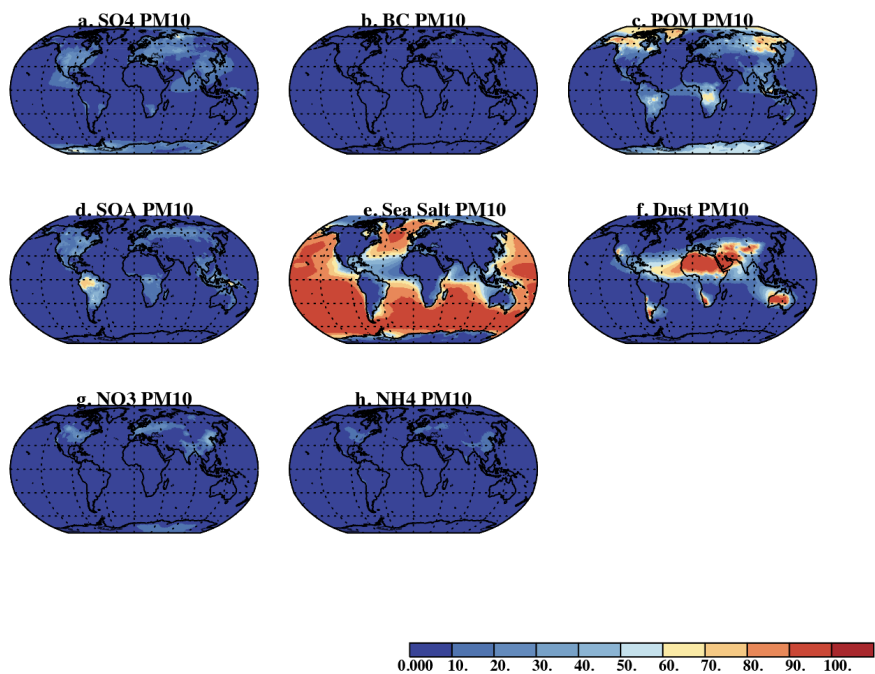


Figure S19: The modeled relative contribution (%) of different aerosol types to the total surface concentration in the PM₁₀ aerosol for a) SO₄, b) BC, c) Primary Organic Material (POM), d) Secondary Organic Aerosol (SOA), e) Sea salt, f) dust, g) NO₃ and h) NH₄.

Table S1: Modelled aerosol composition (rows) and assumed contribution to observed measurements (fractions). Model constituents that are not included in the base CESM are marked with *.

Measured constituent								
Modeled aerosol composition	PM	SO ₄	EC	OM	Na	Al	NO ₃	NH ₄
SO ₄	1.00	1.00	0.00	0.00	0.00	0.00	0.00	0.00
Black carbon	1.00	0.00	1.00	0.00	0.00	0.00	0.00	0.00
Primary Organic Matter	1.00	0.00	0.00	1.00	0.00	0.00	0.00	0.00

Secondary

Organic

Carbon	1.00	0.00	0.00	1.00	0.00	0.00	0.00	0.00
Sea salts	1.00	0.00	0.00	0.00	0.31	0.00	0.00	0.00
Dust	1.00	0.00	0.00	0.00	0.00	0.07	0.00	0.00
NO ₃ *	1.00	0.00	0.00	0.00	0.00	0.00	1.00	0.00
NH ₄ *	1.00	0.00	0.00	0.00	0.00	0.00	0.00	1.00

Table S2: Measurement variables from the supplemental data which are combined in the comparisons and the factor relation. PM=particulate matter; BC=black carbon; EC=elemental carbon; OM=organic matter; OC=organic carbon; Note that the formula from Ash to dust was established by the University of Miami network (Prospero et al., 1996)

Measured reference constituent	Other measured constituents		
PM	PM		
SO ₄ ⁺²	SO ₄	(96/32)*S	
BC	BC	EC	
OM	1.8*OC	OM	
Na	Na		
Al	Al	1.4*0.07*Ash	0.07*Dust
NO ₃ ⁻	NO ₃		
NH ₄ ⁺	NH ₄		

Table S3: Uncertainty estimates for model-observational comparisons for different size and composition fractions for the observations. The averaged normalized standard deviation is used for the spatial, within year, and within month estimates of variability. The resulting total annual and monthly uncertainty are assumed to be the square root of the square of each of these errors added together, along with the Measurement system error which is assumed to be 0.3. For more details see Section 2.3 and 3.s. The number of grid boxes with data for the spatial comparison (and used for the within year, within month and interannual variability statistics) is shown as Num.

Size	Composition	Spatial	Within Year	Meas. error	Within Month	Inter-annual	Total Annual Unc.	Total Monthly Unc.	Num.
PM _{2.5}	PM	0.24	0.53	0.08	0.47	0.18	0.68	0.63	548
	SO ₄	0.32	0.54	0.02	0.49	0.39	0.79	0.76	163
	BC	0.42	0.50	0.11	0.47	0.22	0.76	0.74	79
	OC	0.37	0.55	0.14	0.48	0.19	0.76	0.72	141
	Na	0.34	0.74	0.53	0.73	0.36	1.08	1.07	161
	Al	0.26	0.67	0.24	0.55	0.46	0.93	0.85	161
	NH ₄	0.41	0.95	0.04	0.73	0.28	1.11	0.93	143
	NO ₃	0.34	0.64	0.02	0.62	0.26	0.83	0.82	93
PM ₁₀	PM	0.25	0.47	0.13	0.43	0.17	0.65	0.62	536
	SO ₄	0.51	0.42	0.20	0.34	0.19	0.77	0.73	50
	BC	0.33	0.48	0.07	0.40	0.20	0.69	0.64	23
	OC	0.31	0.47	0.19	0.41	0.21	0.70	0.66	21
	Na	0.35	0.62	0.19	0.53	0.37	0.88	0.82	47
	Al	0.61	0.72	0.06	0.55	0.56	1.14	1.04	51
	NO ₃	0.20	0.63	0.01	0.54	0.24	0.77	0.70	29
	NH ₄	0.30	0.74	0.01	0.59	0.30	0.90	0.78	36

Commented [1]: check this

Table S4. Comparison of climatological averages versus decadal averages (2010-2019) for different size and composition in the observations. The Kendal (ranked) correlation (r), the slope and uncertainty in the slope, root mean squared (rms), climatological average, and decadal average, number of climatological observation station datasets, and the number of decadal observational datasets, the percentage of the values which are more than 20% different, and the % of values which are outside the 3x uncertainty when decadal averages are compared to climatological averages.

Size	Comp.	r	Slope	Unc. Slope	rms	Clim. Avg	Dec. Avg.	Clim. Num	Dec. Num	% > 20%	% > 3x
PM _{2.5}	PM	0.92	1.04	0.0007	3.41	19.5	20.3	7080	5486	3.9	0.04
	SO ₄	0.87	0.90	0.0033	0.37	1.61	1.48	817	580	27.2	0.00
	BC	0.92	1.01	0.0129	0.44	2.65	2.71	62	20	10.0	0.00
	OM	0.92	0.97	0.0030	0.25	2.51	2.39	602	423	7.8	0.00
	Al	0.88	0.99	0.0032	0.04	0.07	0.07	829	564	11.5	0.18
	Na	0.90	1.05	0.0034	0.16	0.10	0.11	836	579	20.0	0.35
	NO ₃	0.93	0.94	0.0022	0.28	0.77	0.74	805	575	11.5	0.00
	NH ₄	0.80	0.88	0.0026	0.19	0.81	0.73	569	365	40.0	0.00
PM ₁₀	PM	0.93	1.03	0.0007	8.82	47.8	49.8	6142	3511	6.8	0.14
	SO ₄	0.89	1.00	0.0058	0.66	2.12	2.19	185	78	10.3	1.28
	BC	0.95	1.20	0.0194	3.29	3.07	4.13	30	13	23.1	0.00
	OM							11	0		
	Na	0.96	0.99	0.0065	0.11	0.62	0.62	162	61	9.8	0.00
	Al	0.97	1.00	0.0050	0.52	0.56	0.62	165	76	9.2	0.00
	NO ₃	0.93	0.98	0.0072	0.47	1.74	1.66	179	63	11.1	0.00
	NH ₄	0.94	1.02	0.0094	0.40	0.74	0.76	138	46	8.7	2.17

Table S5: PM_{2.5} comparison statistics for Figures 3 and 5 model data comparisons including Kendall rank correlation coefficient (r), slope (x=observations, y=model) for linear regression with uncertainties, root mean square differences between modeled and observations (RMS), average model at the location of the observations ($\mu\text{g}/\text{m}^3$), fraction of the modeled values which are outside the 3x uncertainty bound and the number of observations (Num. Obs). If there is no data for a region, the region is not included in the table.

Compositi on	Region	Kendall Rank Correl. (r)	($\mu\text{g}/\text{m}^3$ / $\mu\text{g}/\text{m}^3$) Slope	($\mu\text{g}/\text{m}^3$ / $\mu\text{g}/\text{m}^3$) Slope Unc.	(μg) RMS	(μg) Avg. Mod el	(μg) Avg. Obs	Frac. >3x	Num. Obs.
PM	Globe	0.60	1.2	0.00	14	20	17	0.06	749
PM	Africa	0.40	0.8	0.01	13	23	22	0.04	45
PM	Asia	0.47	1.3	0.01	21	36	31	0.08	271
PM	Australia	0.09	1.2	0.03	5.2	9.1	6.0	0.08	51
PM	Europe	0.37	1.0	0.01	6.4	13	12	0.00	99
PM	N. America	0.55	1.7	0.01	4.6	9.8	6.9	0.02	259
PM	S. America	0.00	0.58	0.02	14	11	17	0.39	23
SO ₄	Globe	0.64	1.8	0.01	1.8	2.9	1.6	0.18	318
SO ₄	Africa	0.41	1.6	0.04	2.3	2.8	1.2	0.28	18
SO ₄	Asia	0.58	1.4	0.01	2.6	4.9	3.2	0.13	55
SO ₄	Australia	0.73	3.9	0.34	0.60	0.90	0.45	0.50	6
SO ₄	Europe	0.36	1.6	0.03	1.54	2.68	1.66	0.10	42
SO ₄	High Latitudes	-1.00	-5.9	1.25	0.31	0.43	0.43	0.00	2
SO ₄	N. America	0.68	2.1	0.01	1.51	2.60	1.33	0.18	187
SO ₄	S. America	0.21	0.16	0.04	0.57	0.90	0.53	0.50	8
BC	Globe	0.63	0.80	0.01	1.1	0.58	0.86	0.12	220
BC	Africa	0.14	0.51	0.01	2.0	0.97	1.9	0.22	18
BC	Asia	0.36	1.0	0.02	1.7	1.4	1.8	0.20	45
BC	Australia	1.00	0.39	0.02	1.4	0.90	1.8	0.00	3
BC	Europe	0.24	0.41	0.01	1.9	0.59	2.1	0.36	11
BC	N. America	0.54	0.95	0.01	0.21	0.24	0.2	0.05	135
BC	S. America	0.43	0.48	0.02	1.5	0.92	1.9	0.29	7
OM	Globe	0.43	1.1	0.01	2.9	4.5	3.9	0.09	264
OM	Africa	0.52	3.3	0.15	7.4	10.1	6.5	0.43	7
OM	Asia	0.21	1.3	0.02	4.3	5.97	5.6	0.19	32
OM	Europe	0.47	0.9	0.01	2.5	3.7	4.0	0.00	41
OM	N. America	0.45	1.1	0.01	2.2	4.2	3.5	0.08	183
Na	Globe	0.53	2.7	0.02	0.35	0.27	0.13	0.30	308
Na	Africa	0.18	3.2	0.14	0.54	0.53	0.18	0.40	15
Na	Asia	-0.01	3.3	0.06	0.43	0.41	0.27	0.46	54

Na	Australia	0.60	1.7	0.12	0.60	0.83	0.27	0.83	6
Na	Europe	0.41	4.9	0.10	0.41	0.41	0.15	0.56	39
Na	High Latitudes	1.00	1.8	2.0	0.07	0.21	0.13	0.00	2
Na	N. America	0.53	2.2	0.02	0.28	0.17	0.08	0.17	187
Na	S. America	0.40	1.8	0.07	0.16	0.22	0.14	0.40	5
Al	Globe	0.53	1.4	0.01	0.60	0.18	0.14	0.44	290
Al	Africa	0.60	3.1	0.07	0.45	0.43	0.25	0.33	15
Al	Asia	0.60	0.84	0.02	1.34	0.64	0.48	0.69	55
Al	Australia	0.20	8.3	0.96	0.05	0.06	0.05	0.33	6
Al	Europe	0.44	1.1	0.02	0.06	0.08	0.08	0.24	17
Al	HighLatitudes	-1.00	-0.09	0.00	0.02	0.00	0.02	1.00	2
Al	N. America	0.60	1.4	0.01	0.05	0.04	0.04	0.38	187
Al	S. America	-0.50	-5.3	0.34	0.16	0.07	0.11	0.63	8
NO ₃	Globe	0.55	2.9	0.01	3.11	2.0	0.78	0.46	283
NO ₃	Africa	-0.07	11	0.49	0.54	0.29	0.20	0.73	11
NO ₃	Asia	0.38	2.1	0.03	6.82	3.0	0.98	0.50	44
NO ₃	Australia	1.00	21	3.4	0.75	0.66	0.22	1.00	2
NO ₃	Europe	0.62	2.9	0.03	2.58	3.2	1.55	0.53	40
NO ₃	N. America	0.60	2.9	0.02	1.52	1.6	0.62	0.41	183
NO ₃	S. America	-1.00	-9.0	10.5	0.39	0.09	0.47	0.50	2
NH ₄	Globe	0.52	1.8	0.01	1.16	1.3	0.79	0.16	222
NH ₄	Africa	0.75	1.3	0.03	0.28	0.57	0.39	0.18	11
NH ₄	Asia	0.36	1.8	0.03	2.17	1.9	1.3	0.12	41
NH ₄	Australia	1.00	2.7	0.13	1.56	1.9	0.63	0.50	2
NH ₄	Europe	0.57	2.2	0.03	0.91	1.5	0.79	0.23	39
NH ₄	N. America	0.56	1.8	0.01	0.73	1.2	0.68	0.13	126
NH ₄	S. America	-1.00	-0.37	0.04	0.33	0.40	0.50	0.00	2

Table S6: The % difference between the PM_{2.5} comparison statistics for Figures 3 and 5 model data comparisons using decadal averages for 2010-2019 versus those used in Table S5. Columns where the amount of datasets included change less than 25% are highlighted in grey. These values including Kendall rank correlation coefficient (r), slope (x=observations, y=model) for linear regression with uncertainties, root mean square differences between modeled and observations (RMS), average model at the location of the observations ($\mu\text{g}/\text{m}^3$), fraction of the modeled values which are outside the 3x uncertainty bound and the number of observations (Num. Obs). If there is no data in the region for that composition for the climatological data, the line in the table is skipped. If there is not data for the decadal comparison, the line in the table is left blank.

Compositi on	Region	Kendall Rank Correl. (r)	($\mu\text{g}/\text{m}^3$ / $\mu\text{g}/\text{m}^3$) Slope	($\mu\text{g}/\text{m}^3$ / $\mu\text{g}/\text{m}^3$) Slope Unc.	(μg) R MS	(μg) Avg. Model	(μg) Avg. Obs	Frac. >3x	Num. Obs.
	PM Globe	-3	-4	-5	-4	-3	2	-6	-12
	PM Africa	6	-26	10	3	-8	4	-13	-42
	PM Asia	9	-7	7	-4	0	8	10	-13
	PM Australia	120	27	45	-30	-5	25	-72	-12
	PM Europe N.	-4	2	16	1	-1	-5	NaN	-12
	PM America S.	-13	18	39	16	1	-6	21	-3
	PM America	-930	-30	-19	9	-44	3	-15	-22
	Al Globe	5	7	14	-11	-58	-26	-1	-24
SO ₄	Globe	-7	26	43	11	-3	-17	62	-21
SO ₄	Africa	50	-10	11	-5	31	50	-49	-61
SO ₄	Asia	-15	-10	36	-5	-10	-9	-10	-36
SO ₄	Australia	-9	-80	-87	1	5	31	0	-33
SO ₄	Europe	-22	2	14	16	-1	1	24	-19
SO ₄	High Latitudes N.								
SO ₄	America S.	0	41	28	26	0	-26	100	-10
SO ₄	America	-570	-1400	280	40	31	36	-33	-63
BC	Globe	3	41	38	-22	-14	-34	-1	-26
BC	Africa	420	94	150	-60	33	-41	-100	-56
BC	Asia	62	0	33	9	8	2	-42	-42
BC	Australia								
BC	Europe	-100	-98	-69	7	-15	-3	37	-64
BC	N.								
BC	America	2	23	35	-31	-2	-25	90	-10
BC	S.								
BC	America	130	-62	-11	-29	-77	-38	250	-71
OM	Globe	17	11	8	-15	-3	-16	11	-14
OM	Africa	15	-31	-40	-19	-13	3	-22	-14
OM	Asia	180	7	22	-41	-13	-41	-41	-44

OM	Europe	5	10	2	-25	1	-10	NaN	-10
OM	N. America	14	15	15	2	1	-12	40	-10
Na	Globe	10	-100	-100	32	-3	10	-6	-20
Na	Africa	-100	2500	19000	18	12	13	0	-67
Na	Asia	1400	-100	-96	12	0	16	80	5
Na	Australia	-44	-11	16	-5	4	35	-40	-33
Na	Europe	11	-11	-10	-1	4	-20	2	-15
Na	HighLatitudes								
Na	N. America	1	-1	-7	1	-1	-6	1	-10
Na	S. America	150	250	1400	24	34	90	25	-60
Al	Africa	-44	9	170	15	15	40	50	-73
Al	Asia	-14	-81	-81	-1	-71	-21	6	-38
Al	Australia	-270	-69	-81	-24	-31	14	-25	-33
Al	Europe	-150	-151	-10	26	14	30	21	-59
Al	HighLatitudes								
Al	N. America	8	5	28	-1	-2	0	0	-10
Al	S. America	100	-86	-86	-49	52	-14	-47	-63
NO ₃	Globe	8	22	32	7	2	-8	17	-16
NO ₃	Africa	-550	-65	-72	7	53	12	-21	-36
NO ₃	Asia	79	7	10	20	19	-28	-21	-36
NO ₃	Australia								
NO ₃	Europe	-4	-10	-14	11	-5	20	-13	-18
NO ₃	N. America	-1	34	28	10	2	-16	40	-9
NO ₃	S. America	0	0	0	0	0	0	0	0
NH ₄	Globe	-3	34	48	16	8	-19	110	-27
NH ₄	Africa	9	2	14	28	28	32	-21	-36
NH ₄	Asia	51	0	19	18	16	-6	-2	-39
NH ₄	Australia								
NH ₄	Europe	12	7	20	1	-3	-9	24	-28

NH ₄	N.								
	America	-8	51	61	38	9	-30	210	-22
NH ₄	S. America								

Table S7: PM₁₀ comparison statistics for Figures 6 and 7 model data comparisons including Kendall rank correlation coefficient (r), slope (x=observations, y=model) for linear regression with uncertainties, root mean square differences between modeled and observations (RMS), average model at the location of the observations ($\mu\text{g}/\text{m}^3$), fraction of the modeled values which are outside the 3x uncertainty bound and the number of observations (Num. Obs). If there is no data for a region, the region is not included in the table.

Comp osition	Region	Kendall Rank Correl. (r)	($\mu\text{g}/\text{m}^3$ / $\mu\text{g}/\text{m}^3$) Slope	($\mu\text{g}/\text{m}^3$ / $\mu\text{g}/\text{m}^3$) Slope Unc.	(μg) RMS	(μg) Avg. Model	(μg) Avg. Obs	Frac. >3x	Num. Obs.
PM	Globe	0.55	1.5	0.00	57	58	38	0.08	702
PM	Africa	0.36	1.9	0.04	130	120	53	0.23	43
PM	Asia	0.48	1.9	0.01	79	100	70	0.07	231
PM	Australia	0.08	-160.0	93	22	30	16	0.07	57
PM	Europe	0.52	1.6	0.01	18	35	22	0.14	116
PM	High Latitudes	0.33	2.2	0.08	2.6	4.6	2.8	0.33	3
PM	N. America	0.37	2.8	0.03	13	24	17	0.02	230
PM	S. America	-0.06	0.81	0.04	24	31	34	0.14	22
SO ₄	Globe	0.43	2.8	0.02	5.3	6.0	3.4	0.48	164
SO ₄	Africa	0.32	2.3	0.04	5.0	5.1	1.6	0.50	26
SO ₄	Asia	0.44	1.1	0.02	7.2	9.8	7.6	0.36	50
SO ₄	Australia	-0.15	-0.06	0.02	1.1	1.4	0.82	0.25	12
SO ₄	Europe	0.21	2.6	0.05	4.6	5.6	1.61	0.63	43
SO ₄	High Latitudes	0.67	0.68	0.03	0.50	0.53	0.91	0.25	4
SO ₄	N. America	0.26	3.6	0.10	3.9	4.4	1.7	0.59	22
SO ₄	S. America	0.33	1.2	0.06	0.93	1.6	1.0	0.43	7
BC	Globe	0.47	0.86	0.01	2.2	1.9	2.0	0.16	85
BC	Africa	0.35	0.00	0.00	2.1	2.0	2.0	0.20	20

BC	Asia	0.19	0.61	0.01	3.6	3.5	3.6	0.14	22
BC	Australia	1.00	1.08	0.12	0.29	1.1	0.77	0.00	2
BC	Europe	0.16	0.4	0.02	1.4	1.2	1.6	0.22	18
BC	N. America	0.52	1.4	0.05	0.58	0.76	0.62	0.13	16
BC	S. America	0.73	0.93	0.03	0.99	2.0	2.3	0.17	6
OM	Globe	0.26	1.5	0.02	10	11	5.6	0.29	72
OM	Africa	0.14	2.6	0.11	18	18	10	0.50	8
OM	Asia	0.10	0.68	0.04	15	15	7.6	0.36	14
OM	Europe	0.40	1.2	0.02	4.2	7.6	4.9	0.08	39
OM	N. America	0.11	7.2	0.26	11	11	2.4	0.89	9
Na	Globe	0.49	2.2	0.02	3.9	2.4	1.90	0.50	135
Na	Africa	0.40	2.2	0.04	3.3	3.2	1.08	0.45	20
Na	Asia	0.43	0.81	0.01	2.7	1.6	2.3	0.35	40
Na	Australia	0.55	0.03	0.00	10	4.5	7.0	0.58	12
Na	Europe	0.63	3.1	0.04	1.8	2.0	0.83	0.56	39
Na	High Latitudes	1.00	1.7	0.06	0.6	1.4	0.87	0.50	4
Na	N. America	0.62	3.0	0.06	2.8	3.2	1.3	0.60	15
Na	S. America	0.80	2.7	0.15	2.2	1.8	0.57	0.80	5
Al	Globe	0.46	1.9	0.02	7.3	3.0	0.69	0.54	140
Al	Africa	0.24	3.8	0.09	6.4	4.5	1.2	0.55	22
Al	Asia	0.47	4.4	0.09	13	7.7	1.3	0.74	38
Al	Australia	0.29	-0.12	0.00	0.21	0.22	0.21	0.25	8
Al	Europe	0.43	1.9	0.03	0.37	0.31	0.21	0.35	43
Al	High Latitudes	0.00	1.1	0.09	0.02	0.00	0.02	0.80	5
Al	N. America	0.42	0.82	0.02	0.89	0.53	0.24	0.56	16
Al	S. America	-0.29	-0.23	0.01	1.0	0.33	0.65	0.75	8
NO ₃	Globe	0.39	3.0	0.03	9.1	3.8	1.5	0.69	129
NO ₃	Africa	0.01	3.8	0.14	0.67	0.37	0.64	0.88	17

NO ₃									
NO ₃	Asia	0.31	1.9	0.03	15	6.3	2.3	0.55	40
NO ₃	Australia	0.31	7.1	0.36	0.70	0.26	0.21	1.00	10
NO ₃	Europe	0.38	3.7	0.05	4.4	4.3	1.8	0.54	35
NO ₃	High Latitudes	-0.33	-2.7	0.77	0.01	0.01	0.02	0.67	3
NO ₃	N. America	0.29	9.3	0.18	4.6	3.6	0.75	0.86	22
NO ₃	S. America	-1.00	-0.01	0.00	1.0	0.00	0.75	1.00	2
NH ₄	Globe	0.57	2.9	0.02	3.6	2.5	0.81	0.49	98
NH ₄	Africa	0.55	2.5	0.05	0.88	0.81	0.31	0.35	17
NH ₄	Asia	0.37	2.1	0.04	6.3	4.7	1.6	0.33	27
NH ₄	Australia	0.73	6.8	0.25	2.4	1.4	0.22	0.83	6
NH ₄	Europe	0.45	3.6	0.06	2.1	2.6	0.85	0.58	31
NH ₄	High Latitudes	0.33	1.7	0.27	0.02	0.01	0.03	0.67	3
NH ₄	N. America	0.09	4.2	0.23	1.7	1.4	0.27	0.67	12
NH ₄	S. America	1.00	0.76	0.04	0.09	0.37	0.41	0.00	2

Table S8: The % difference between the PM₁₀ comparison statistics for Figures 6 and 7 model data comparisons using decadal averages for 2010-2019 versus those used in Table S7. Columns where the amount of datasets included change less than 25% are highlighted in grey. These values including Kendall rank correlation coefficient (r), slope (x=observations, y=model) for linear regression with uncertainties, root mean square differences between modeled and observations (RMS), average model at the location of the observations (μg/m³), fraction of the modeled values which are outside the 3x uncertainty bound and the number of observations (Num. Obs). If there is no data in the region for that composition for the climatological data, the line in the table is skipped. If there is not data for the decadal comparison, the line in the table is left blank.

Size	Composit ion	Region	Kendall Rank Correl. (r)	(ug/ ug) Slop e	(ug/ug) Slope Unc.	(ug) R M S	(ug) Avg. Model	(ug) Avg. Obs	Frac. >3x	Num. Obs.
PM10	PM	Globe	-3	-6	70	-12	-3	3	20	-17
PM10	PM	Africa	-9	-32	-9	7	11	11	-2	-19
PM10	PM	Asia	-4	-7	-8	-27	-12	-1	0	-12

PM10	PM	Australia	89	-102	-100	-27	-6	25	-72	-11
PM10	PM	Europe	0	1	6	7	0	-1	46	-6
						-10				
PM10	PM	High Latitudes N.	-100	-100	-100	0	-100	-100	-100	-100
PM10	PM	America S.	-7	10	31	15	6	-8	91	-27
PM10	PM	America	311	-35	-40	-5	-28	3	38	-27
PM10	SO4	Globe	-24	14	46	11	11	-32	41	-54
PM10	SO4	Africa	123	33	156	28	37	4	43	-73
PM10	SO4	Asia	-31	92	199	4	-3	-40	9	-54
				-900						
PM10	SO4	Australia	-540	0	1600	43	39	-44	200	-67
PM10	SO4	Europe	-35	-9	19	13	8	-20	37	-19
						-10				
PM10	SO4	High Latitudes N.	-100	-100	-100	0	-100	-100	-100	-100
PM10	SO4	America	-75	79	541	-11	-19	-44	13	-73
						-10				
PM10	SO4	S. America	-100	-100	-100	0	-100	-100	-100	-100
PM10	BC	Globe	37	6	103	8	8	35	-7	-69
				-400						
PM10	BC	Africa	147	00	2300	-29	17	-28	-38	-60
PM10	BC	Asia	-271	-93	-27	8	86	211	83	-82
						-10				
PM10	BC	Australia	-100	-100	-100	0	-100	-100	NaN	-100
PM10	BC	Europe N.	-71	-98	-63	11	-24	10	29	-61
PM10	BC	America	16	6	58	-62	-18	-14	-100	-69
						-10				
PM10	BC	S. America	-100	-100	-100	0	-100	-100	-100	-100
PM10	OM	Globe	-271	-93	-27	8	86	211	83	-82
						-10				
PM10	OM	Africa	-100	-100	-100	0	-100	-100	NaN	-100
PM10	OM	Asia	-71	-98	-63	11	-24	10	29	-61
PM10	OM	Europe	16	6	58	-62	-18	-14	-100	-69

						-10				
PM10	OM	NorthAmerica	-100	-100	-100	0	-100	-100	-100	-100
PM10	Na	Globe	-31	-101	-100	-37	-20	-51	21	-56
PM10	Na	Africa	-100	-325	411	2	-1	-43	33	-75
PM10	Na	Asia	-53	80230	218	15	-24	-11	10	-68
PM10	Na	Australia	-39	0	2300	-79	-32	-84	29	-67
PM10	Na	Europe	-7	14	35	-9	-16	-42	16	-26
PM10	Na	High Latitudes N. America	-100	-100	-100	0	-100	-100	-100	-100
PM10	Na		-8	76	185	10	-16	-42	25	-47
PM10	Na	SouthAmerica	-100	-100	-100	0	-100	-100	-100	-100
PM10	Al	Globe	-9	10	29	-66	-67	-11	-15	-52
PM10	Al	Africa	236	-80	-47	-78	-39	37	-63	-77
PM10	Al	Asia	-49	321	1041	-56	-63	48	2	-68
PM10	Al	Australia	17	-946	705	-14	-22	40	0	-50
PM10	Al	Europe	34	16	1	-9	-4	-22	-4	-16
PM10	Al	High Latitudes N. America	NaN	-100	-100	0	-100	-100	-100	-100
PM10	Al		37	83210	212	24	12	7	33	-50
PM10	Al	S. America	-450	0	2500	-41	77	-83	33	-75
PM10	NO3	Globe	-30	23	160	29	23	6	-23	-60
PM10	NO3	Africa	4200	-93	-88	-15	83	53	-24	-65
PM10	NO3	Asia	38	-20	39	39	36	-9	-48	-65
PM10	NO3	Australia	-100	-100	-100	0	-100	-100	-100	-100
PM10	NO3	Europe	-64	38	153	1	-4	-9	-4	-34
PM10	NO3	High Latitudes N. America	-100	-100	-100	0	-100	-100	-100	-100
PM10	NO3		-30	171	655	-15	-15	-39	-4	-73

						- 10				
PM10	NO3	S. America	-100	-100	-100	0	-100	-100	-100	-100
PM10	NH4	Globe	-19	60	236	40	39	-4	36	-60
PM10	NH4	Africa	57	62	217	50	57	39	-6	-65
PM10	NH4	Asia	-165	-105	-31	73	86	14	114	-74
						- 10				
PM10	NH4	Australia	-100	-100	-100	0	-100	-100	-100	-100
PM10	NH4	Europe	-14	-24	30	16	9	-18	24	-42
						- 10				
PM10	NH4	High Latitudes	-100	-100	-100	0	-100	-100	-100	-100
				107						
PM10	NH4	N. America	267	0	1564	13	16	-34	25	-50
						- 10				
PM10	NH4	S. America	-100	-100	-100	0	-100	-100	NaN	-100