Figure S1: Modeled percentage of PM_{10} concentrations that are within $PM_{6.9}$ for the annual average.
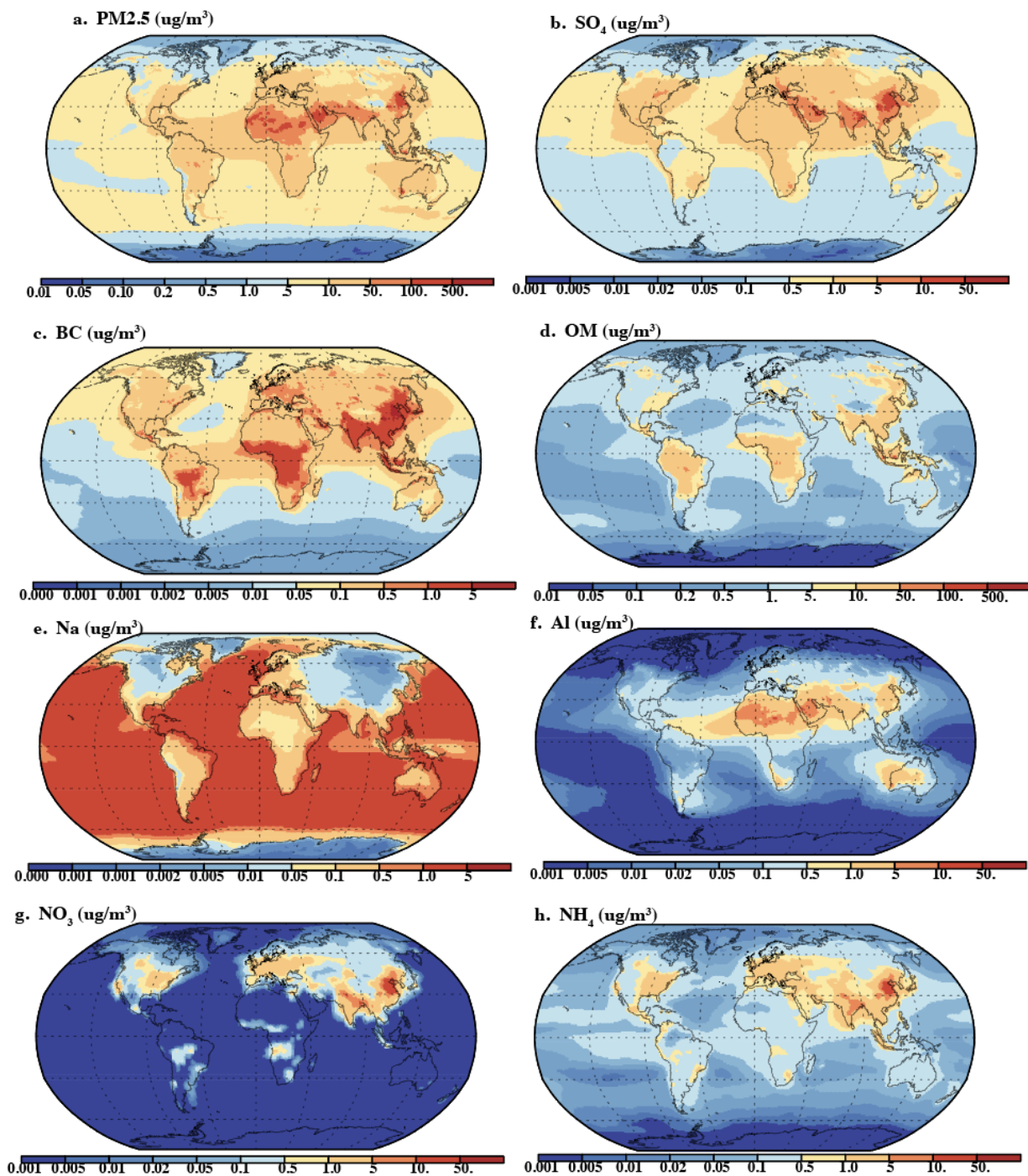
Figure S2: Modeled PM₂.₅ annual mean concentrations $\mu g/m^3$ for a) total PM b) SO₄, c) BC, d) OM, e) Na, f) Al, g) NO₃, h) NH₄. These show the same model simulations as Figure 2 and 4, but show only the model simulations.

Figure S3: Modeled PM$_{10}$ annual mean concentrations   µg/m^3 for a) total PM b) SO$_4$, c) BC, d) OM, e) Na, f) Al, g) NO$_3$, h) NH$_4$. These show the same model simulations as Figure 2 and 4, but show only the model simulations.
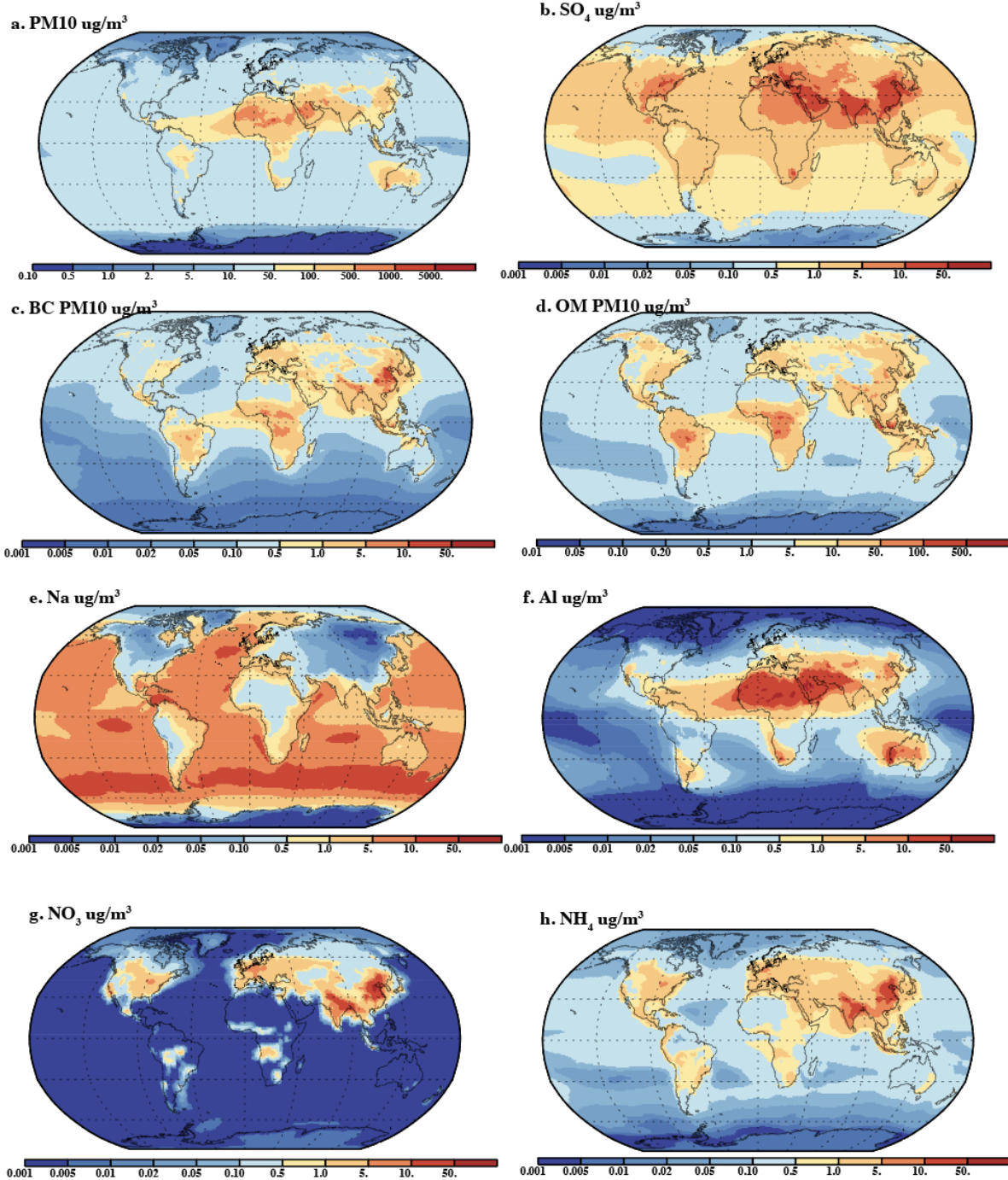
Table S1: Anthropogenic Percentage of Dust [a] (Table reproduced from Brodsky et al. (2023)

	North America	South America	North Africa	South Africa	Western Asia	Central Asia	Eastern Asia	Australia
% Anthropogenic Ginoux et al. (2012)	54	41	8.2	54	30	45	40	76[b]
% Anthropogenic - Tuned Run	49	40	8.4	59	32	48	38	15

[a] Average emissions over the regions specified in Ginoux et al. (2012) were calculated and crop area effectiveness as a dust source was modified to create the tuned anthropogenic crop dust source.

[b] Other studies have found a lower percent agricultural dust for Australia (Bullard et al., 2008; Mahowald et al., 2009; Webb and Pierre, 2018)

Table S2: Modelled aerosol composition (rows) and assumed contribution to observed measurements (fractions). Model constituents that are not included in the base CESM are marked with *.

Modeled aerosol composition	Measured constituent							
	PM	SO₄	EC	OM	Na	Al	NO₃	NH₄
SO₄	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	0.50	0.00	0.00	0.50	0.00	0.00	0.00	0.00
Secondary Organic Carbon	0.50	0.00	0.00	0.50	0.00	0.00	0.00	0.00
Seasalts	1.00	0.00	0.00	0.00	0.39	0.00	0.00	0.00
Dust	1.00	0.00	0.00	0.00	0.00	0.07	0.00	0.00
NO₃*	0.50	0.00	0.00	0.00	0.00	0.00	0.50	0.00
NH4*	1.00	0.00	0.00	0.00	0.00	0.00	0.00	1.00
Agricultural dust*	1.00	0.00	0.00	0.00	0.00	0.07	0.00	0.00
Road dust*	1.00	0.00	0.00	0.00	0.00	0.07	0.00	0.00
Coarse organic carbon*	1.00	0.00	0.00	1.00	0.00	0.00	0.00	0.00
Coarse black carbon*	1.00	0.00	1.00	0.00	0.00	0.00	0.00	0.00
Fine and coarse inorganic industrial matter*	1.00	0.00	0.00	0.00	0.00	0.07	0.00	0.00
Bacteria and Fungi*	1.00	0.00	0.00	1.00	0.00	0.00	0.00	0.00
Other Primary Biogenic Particles from land*	1.00	0.00	0.00	1.00	0.00	0.00	0.00	0.00

| Marine organic aerosols* | 1.00 | 0.00 | 0.00 | 1.00 | 0.00 | 0.00 | 0.00 | 0.00 |

Table S3: Measurement variables from the supplemental data which are combined in the comparisons and the factor relation. PM=particulate matter; BC=black carbon; BC=elemental 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}	SO4	(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^-	NO3		
NH_4^+	NH4		

Table S4: PM$_{2.5}$ comparison statistics for Figures 2 and 4 model data comparisons including correlation coefficient (r), intercept (A) and slope (B) for linear regression with uncertainties (sigma A and sigma B), root mean square differences (rms), average model during time of the observations ($\mu g/m^3$) and the number of observations (N).

	r	A	B	sigma A	sigma B	rms ($\mu g/m^3$)	avg model ($\mu g/m^3$)	avg obs ($\mu g/m^3$)	N obs
PM	0.73	-6.23E-01	1.55	1.19E-01	5.06E-03	20	25	17	752
PM (not gridded)	0.78	-2.31E+00	2.15	6.67E-02	2.16E-03	33	38	19	7091
SO$_4$	0.73	2.00E-01	1.83	1.76E-02	7.21E-03	2.0	3.0	1.7	318
BC	0.26	3.27E-02	0.76	8.63E-03	4.57E-03	3.1	0.6	1.1	221
OM	0.38	6.46E-01	1.09	3.46E-02	6.46E-03	4.2	4.8	4.2	264
Na	0.47	-5.44E-02	1.39	6.71E-03	5.62E-03	1.6	1.0	0.87	283
Al	0.32	-4.79E-02	1.74	1.02E-02	8.73E-03	1.4	1.3	0.86	226
NO$_3$	0.42	-5.29E-02	3.39	3.28E-03	1.99E-02	0.45	0.34	0.13	308
NH$_4$	0.50	-3.16E-02	4.00	4.13E-03	2.05E-02	0.87	0.42	0.14	290

Table S5: PM$_{10}$ comparison statistics for Figures 5 and 6 model data comparisons including correlation coefficient (r), intercept (A) and slope (B) for linear regression with uncertainties (sigma A and sigma B), root mean square differences (rms), average model during time of the observations ($\mu g/m^3$) and the number of observations (N).

	r	A	B	sigma A	sigma B	rms ($\mu g/m^3$)	avg model ($\mu g/m^3$)	avg obs ($\mu g/m^3$)	N obs
PM	0.73	-1.76E+00	1.86	3.34E-01	4.49E-03	71	69	37	706
PM (not gridded)	0.76	-7.59E+00	2.47	2.15E-01	2.47E-03	86	90	40	6219
SO$_4$	0.61	8.28E-01	2.46	6.74E-02	1.59E-02	5.6	6.5	3.0	167
BC	-0.04	-7.26E-02	1.23	2.51E-01	1.77E-02	34	1.94	5.0	87
OM	0.43	1.51E+00	1.69	1.48E-01	2.18E-02	11	12	5.3	72
Na	0.34	-4.20E-02	1.48	2.71E-02	1.45E-02	4.5	2.0	1.3	132
Al	0.50	-4.26E-02	3.18	2.17E-02	2.23E-02	3.7	2.6	0.72	111
NO$_3$	0.44	8.17E-02	-0.01	5.13E-04	0.00E+00	3.3	2.7	1.7	138
NH$_4$	0.59	-8.34E-03	4.62	2.72E-02	3.52E-02	8.4	3.7	0.55	142