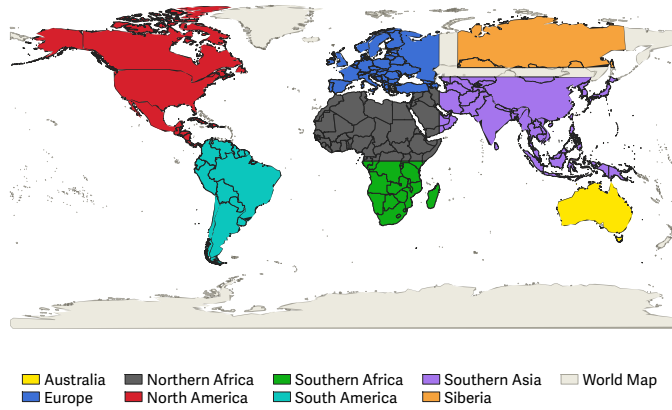


S1: definition of statistical metrics for evaluation of model with observations

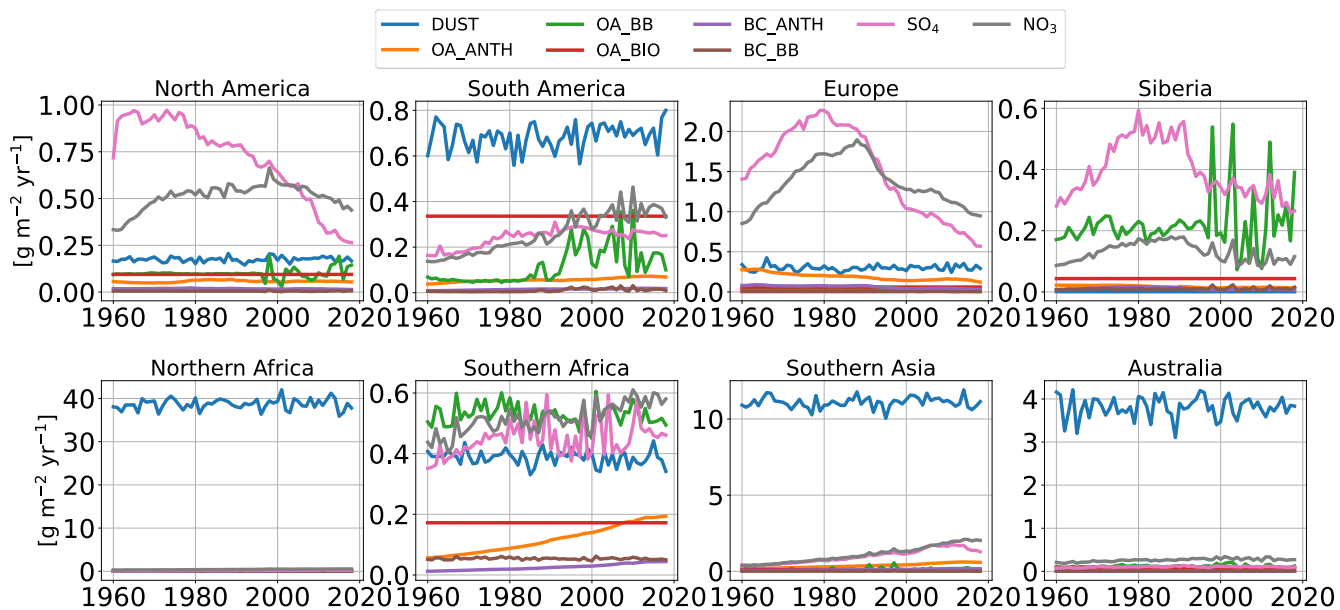
Below, we report the definition of the metrics used for evaluating the model against observations: normalized mean bias (NMB), root mean square error (RMSE) and Pearson's correlation coefficient (r). N represents the total number of model-observation pair values while M_i and O_i represent the i-th model and observed values, respectively

$$NMB = \sum_{i=1}^n \frac{(M_i - O_i)}{\sum_{i=1}^n O_i}$$
$$RMSE = \sum_{i=1}^n \frac{\sqrt{(M_i - O_i)^2}}{N}$$
$$r = \sum_{i=1}^n \frac{(M_i - \bar{M})(O_i - \bar{O})}{\sqrt{\sum_{i=1}^n (M_i - \bar{M})^2 \sum_{i=1}^n (O_i - \bar{O})^2}}$$

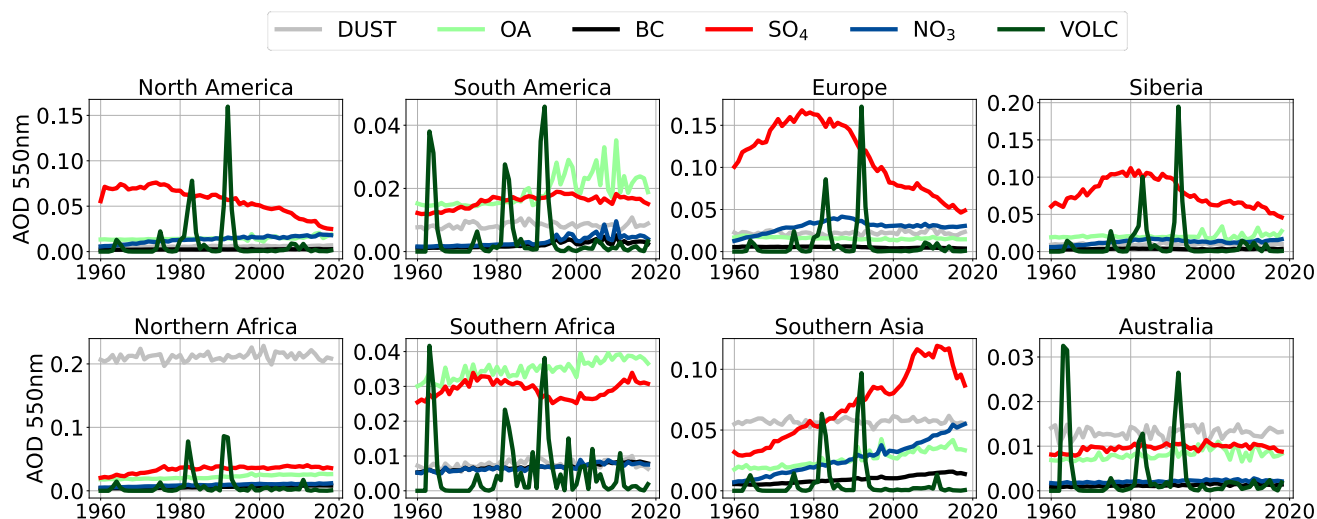
S2: Supplemental Figures and Tables



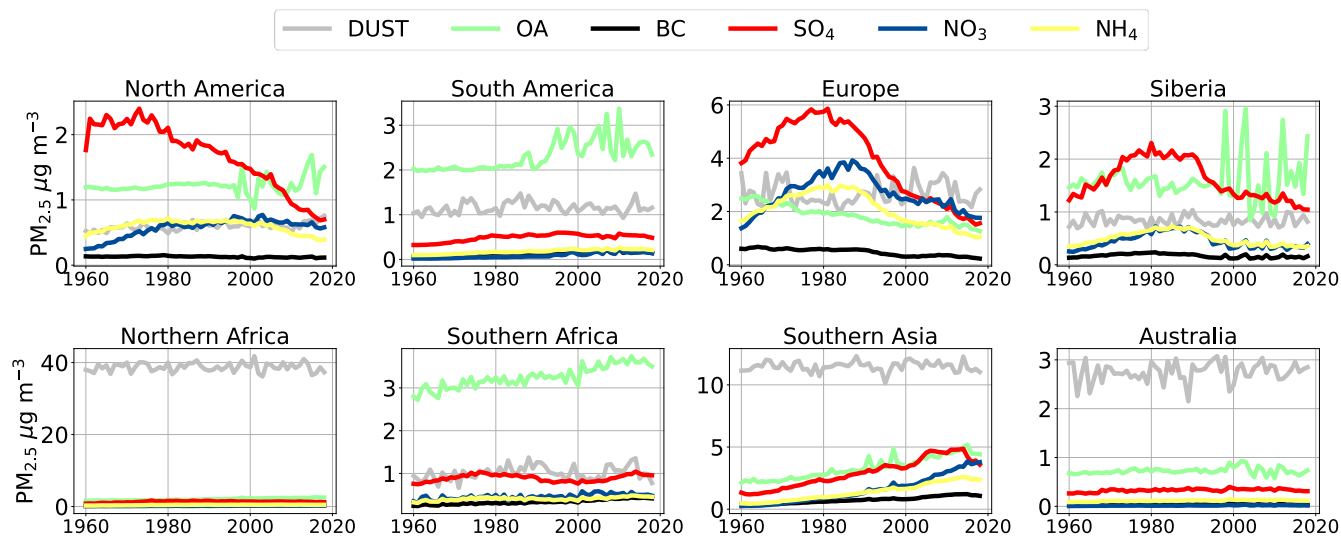
Supplemental Figure 1. Geographical definition of the 8 regions used in this study.



Supplemental Figure 2. Ref-D1 regional annual timeseries of emissions and production of aerosols by region for the period 1960-2018. ANTH=anthropogenic, BB=biomass burning, BIO=biogenic. SO₄ includes direct emissions + production but no emissions from volcanic eruptions, NO₃ refers to production. OA includes all organic mass (emissions+SOA production).

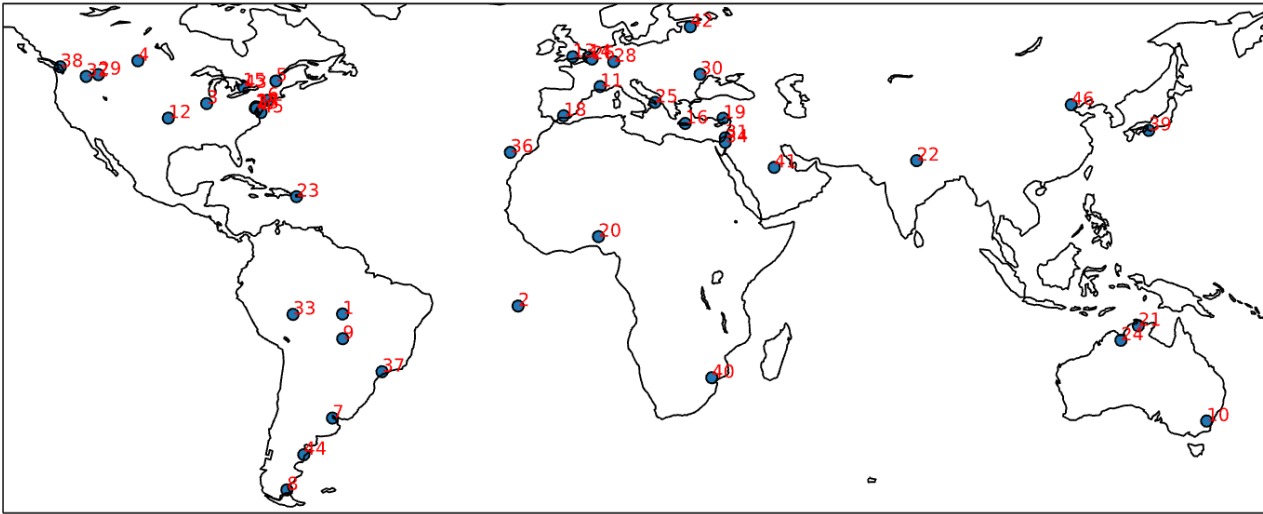


(a)



(b)

Supplemental Figure 3. Ref-D1 regional annual timeseries of individual components of AOD (a) and surface PM_{2.5} (b) for 1960-2018.

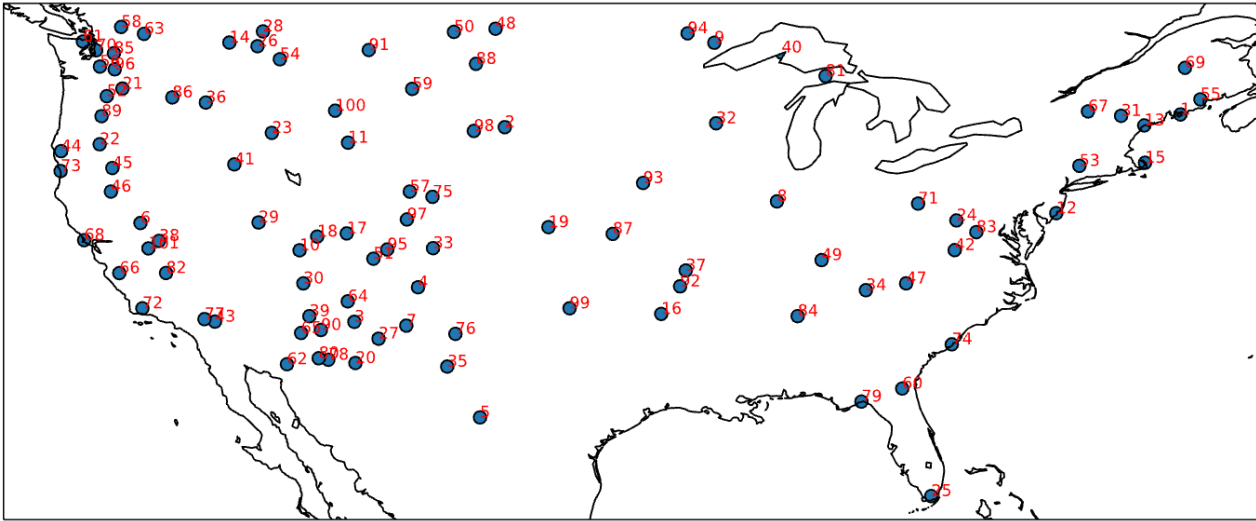


Supplemental Figure 4. Map of selected AERONET stations for AOD and AE observations. Information on each site is reported in Supplemental Table 1.

Supplemental Table 1. List of selected AERONET sites used in this study.

ID	station	latitude	longitude
1	Alta Floresta	-9.871	-56.104
2	Ascension Island	-7.976	-14.415
3	BONDVILLE	40.053	-88.372
4	Bratts Lake	50.204	-104.713
5	CARTEL	45.380	-71.931
6	CCNY	40.821	-73.949
7	CEILAP-BA	-34.555	-58.506
8	CEILAP-RG	-51.601	-69.319
9	CUIABA-MIRANDA	-15.731	-56.071
10	Canberra	-35.271	149.111
11	Carpentras	44.083	5.058
12	Cart Site	36.607	-97.486
13	Chilbolton	51.144	-1.437
14	Dunkerque	51.035	2.368
15	Egbert	44.232	-79.781
16	FORTH CRETE	35.333	25.282
17	GSFC	38.993	-76.840

18	Granada	37.164	-3.605
19	IMS-METU-ERDEMLI	36.565	34.255
20	Ilorin	8.484	4.675
21	Jabiru	-12.661	132.893
22	Kanpur	26.513	80.232
23	La Parguera	17.970	-67.045
24	Lake Argyle	-16.108	128.749
25	Lecce University	40.335	18.111
26	Lille	50.612	3.142
27	MD Science Center	39.281	-76.612
28	Mainz	49.999	8.300
29	Missoula	46.917	-114.083
30	Moldova	47.001	28.816
31	Nes Ziona	31.923	34.789
32	Rimrock	46.488	-116.992
33	Rio Branco	-9.957	-67.869
34	SEDE BOKER	30.855	34.782
35	SERC	38.889	-76.556
36	Santa Cruz Tenerife	28.473	-16.247
37	Sao Paulo	-23.562	-46.735
38	Saturn Island	48.775	-123.128
39	Shirahama	33.693	135.357
40	Skukuza	-24.992	31.588
41	Solar Village	24.907	46.397
42	Toravere	58.265	26.467
43	Toronto	43.790	-79.470
44	Trelew	-43.250	-65.309
45	Wallops	37.933	-75.472
46	XiangHe	39.754	116.962



Supplemental Figure 5. Map of selected IMPROVE stations for PM_{2.5} observations. Information on each site is reported in Supplemental Table 2.

Supplemental Table 2. List of selected IMPROVE sites used in this study.

ID	Station Code	State	Latitude	Longitude
1	ACAD1	ME	44.38	-68.26
2	BADL1	SD	43.74	-101.94
3	BALD1	AZ	34.06	-109.44
4	BAND1	NM	35.78	-106.27
5	BIBE1	TX	29.30	-103.18
6	BLIS1	CA	38.98	-120.10
7	BOAP1	NM	33.87	-106.85
8	BOND1	IL	40.05	-88.37
9	BOWA1	MN	47.95	-91.50
10	BRCA1	UT	37.62	-112.17
11	BRID1	WY	42.97	-109.76
12	BRIG1	NJ	39.47	-74.45
13	CABA1	ME	43.83	-70.06
14	CABI1	MT	47.95	-115.67
15	CACO1	MA	41.98	-70.02
16	CACR1	AR	34.45	-94.14
17	CANY1	UT	38.46	-109.82
18	CAPI1	UT	38.30	-111.29
19	CEBL1	KS	38.77	-99.76
20	CHIR1	AZ	32.01	-109.39
21	CORI1	WA	45.66	-121.00
22	CRLA1	OR	42.90	-122.14
23	CRMO1	ID	43.46	-113.56
24	DOSO1	WV	39.11	-79.43

25	EVER1	FL	25.39	-80.68
26	FLAT1	MT	47.77	-114.27
27	GICL1	NM	33.22	-108.24
28	GLAC1	MT	48.51	-114.00
29	GRBA1	NV	39.01	-114.22
30	GRCA2	AZ	35.97	-111.98
31	GRGU1	NH	44.31	-71.22
32	GRR11	MN	43.94	-91.41
33	GRSA1	CO	37.72	-105.52
34	GRSM1	TN	35.63	-83.94
35	GUMO1	TX	31.83	-104.81
36	HECA1	OR	44.97	-116.84
37	HEGL1	MO	36.61	-92.92
38	HOOV1	CA	38.09	-119.18
39	IKBA1	AZ	34.34	-111.68
40	ISLE1	MI	47.46	-88.15
41	JARB1	NV	41.89	-115.43
42	JARI1	VA	37.63	-79.51
43	JOSH1	CA	34.07	-116.39
44	KALM1	OR	42.55	-124.06
45	LABE1	CA	41.71	-121.51
46	LAVO1	CA	40.54	-121.58
47	LIGO1	NC	35.97	-81.93
48	LOST1	ND	48.64	-102.40
49	MACA1	KY	37.13	-86.15
50	MELA1	MT	48.49	-104.48
51	MEVE1	CO	37.20	-108.49
52	MOHO1	OR	45.29	-121.78
53	MOMO1	CT	41.82	-73.30
54	MONT1	MT	47.12	-113.15
55	MOOS1	ME	45.13	-67.27
56	MORA1	WA	46.76	-122.12
57	MOZI1	CO	40.54	-106.68
58	NOCA1	WA	48.73	-121.06
59	NOCH1	MT	45.65	-106.56
60	OKEF1	GA	30.74	-82.13
61	OLYM1	WA	48.01	-122.97
62	ORPI1	AZ	31.95	-112.80
63	PASA1	WA	48.39	-119.93
64	PEFO1	AZ	35.08	-109.77
65	PHOE1	AZ	33.50	-112.10
66	PINN1	CA	36.48	-121.16
67	PMRF1	VT	44.53	-72.87
68	PORE1	CA	38.12	-122.91
69	PRIS1	ME	46.70	-68.03
70	PUSO1	WA	47.57	-122.31
71	QUCI1	OH	39.94	-81.34
72	RAFA1	CA	34.73	-120.01
73	REDW1	CA	41.56	-124.08
74	ROMA1	SC	32.94	-79.66
75	ROMO1	CO	40.28	-105.55
76	SACR1	NM	33.46	-104.40

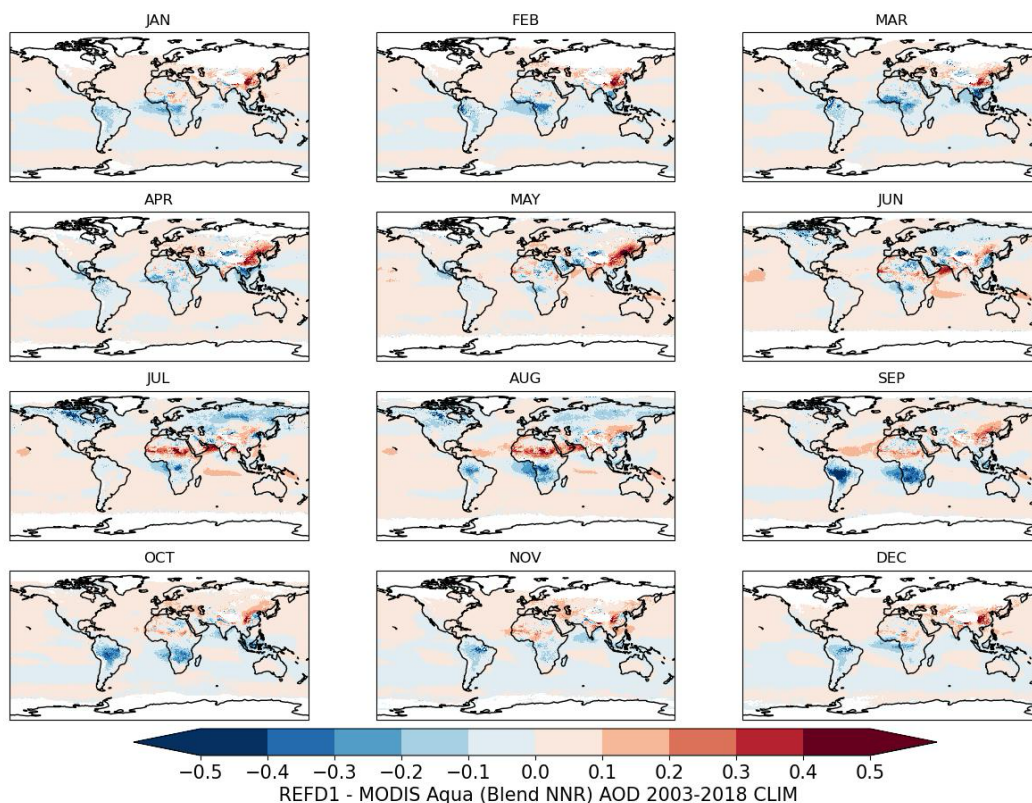
77	SAGO1	CA	34.19	-116.91
78	SAGU1	AZ	32.17	-110.74
79	SAMA1	FL	30.09	-84.16
80	SAWE1	AZ	32.25	-111.22
81	SENE1	MI	46.29	-85.95
82	SEQU1	CA	36.49	-118.83
83	SHEN1	VA	38.52	-78.43
84	SIPS1	AL	34.34	-87.34
85	SNPA1	WA	47.42	-121.43
86	STAR1	OR	45.22	-118.51
87	TALL1	KS	38.43	-96.56
88	THRO1	ND	46.89	-103.38
89	THSI1	OR	44.29	-122.04
90	TONT1	AZ	33.65	-111.11
91	ULBE1	MT	47.58	-108.72
92	UPBU1	AR	35.83	-93.20
93	VILA1	IA	40.97	-95.05
94	VOYA2	MN	48.41	-92.83
95	WEMI1	CO	37.66	-107.80
96	WHPA1	WA	46.62	-121.39
97	WHR11	CO	39.15	-106.82
98	WICA1	SD	43.56	-103.48
99	WIMO1	OK	34.73	-98.71
100	YELL2	WY	44.57	-110.40
101	YOSE1	CA	37.71	-119.71



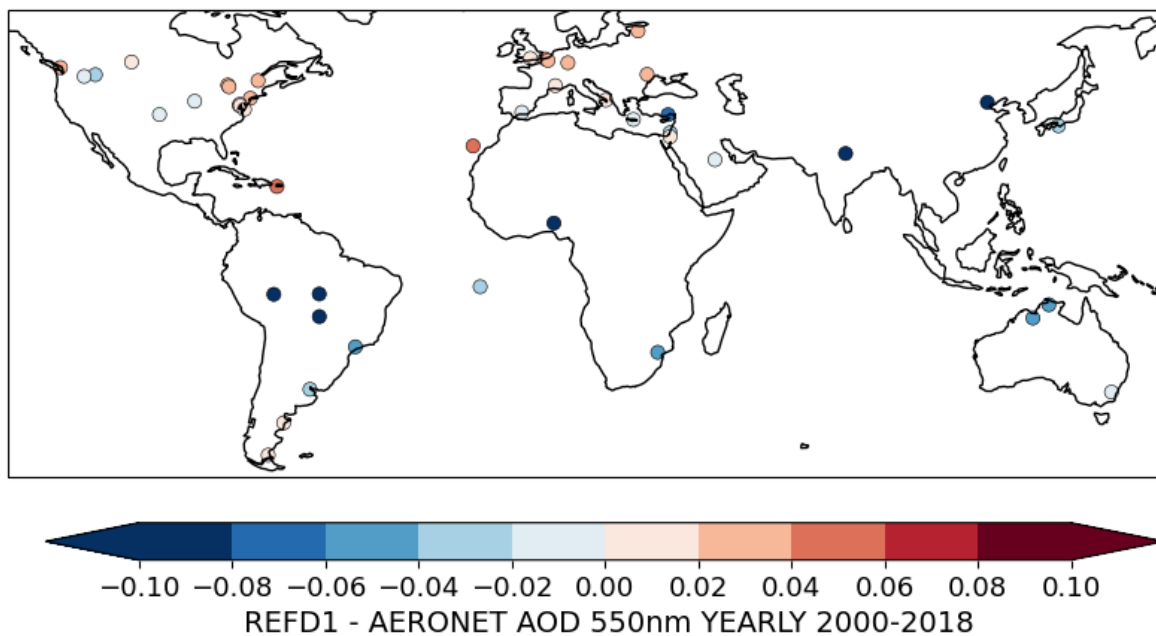
Supplemental Figure 6. Map of IMPROVE sites with long-term co-located measurements of speciated PM, scattering coefficient and RH. Information on each site is reported in Supplemental Table 3.

Site Name	SiteCode	Latitude	Longitude
Mount Rainier	MORA1	46.758	-122.124
Mammoth Cave NP	MACA1	37.132	-86.148
Great Smoky Mountains NP	GRSM1	35.633	-83.942
Shenandoah NP	SHEN1	38.523	-78.435
Grand Canyon NP	GRCA2	35.973	-111.984
Big Bend NP	BIBE1	29.303	-103.178

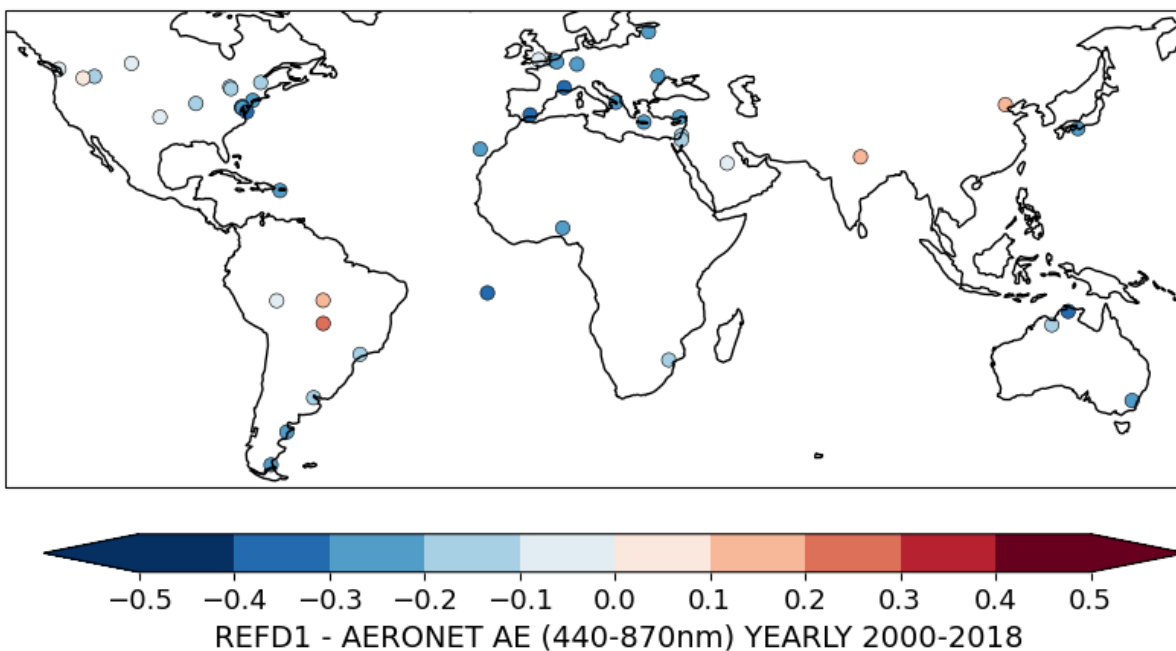
Supplemental Table 3. IMPROVE sites with long-term co-located measurements of speciated PM, scattering coefficient and RH.



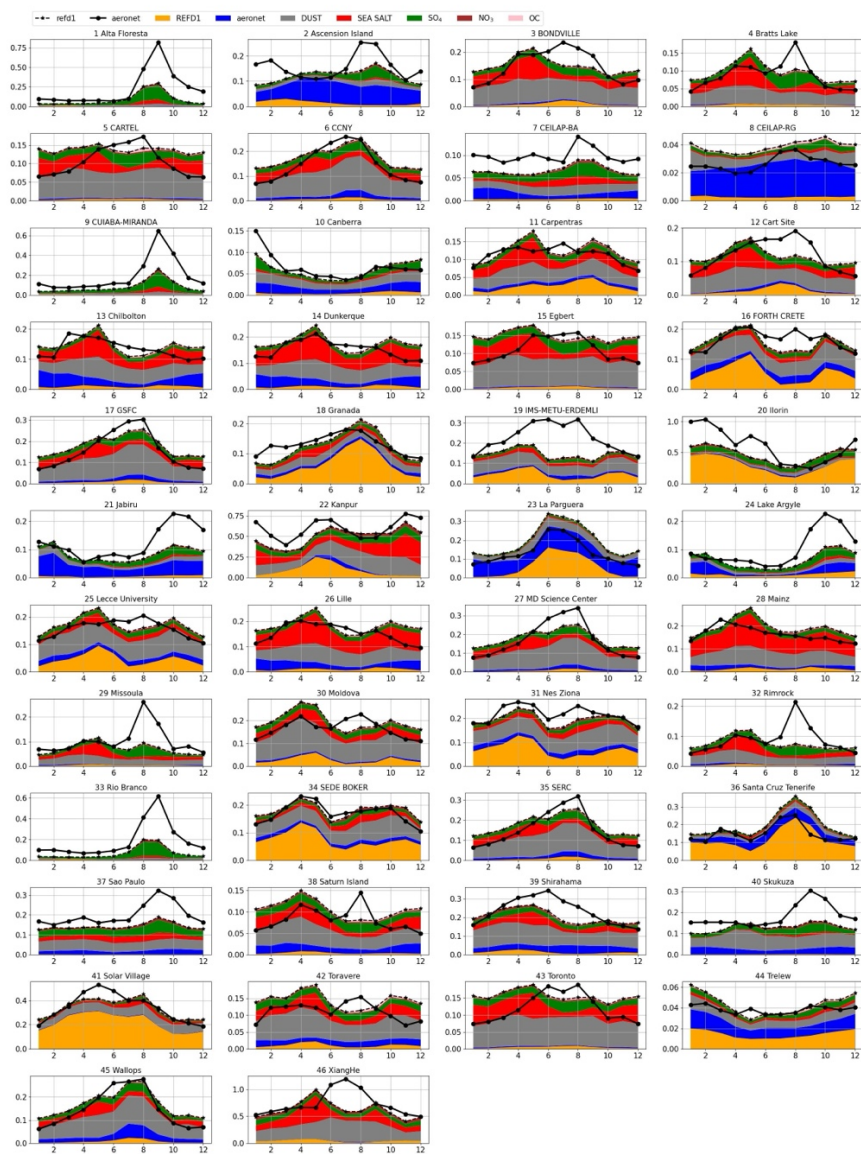
Supplemental Figure 7. Climatological monthly average difference in total AOD between GEOSCCM and MODIS Aqua (2003-2018). In the comparison, we exclude areas where MODIS has no observations in the monthly means (white areas).



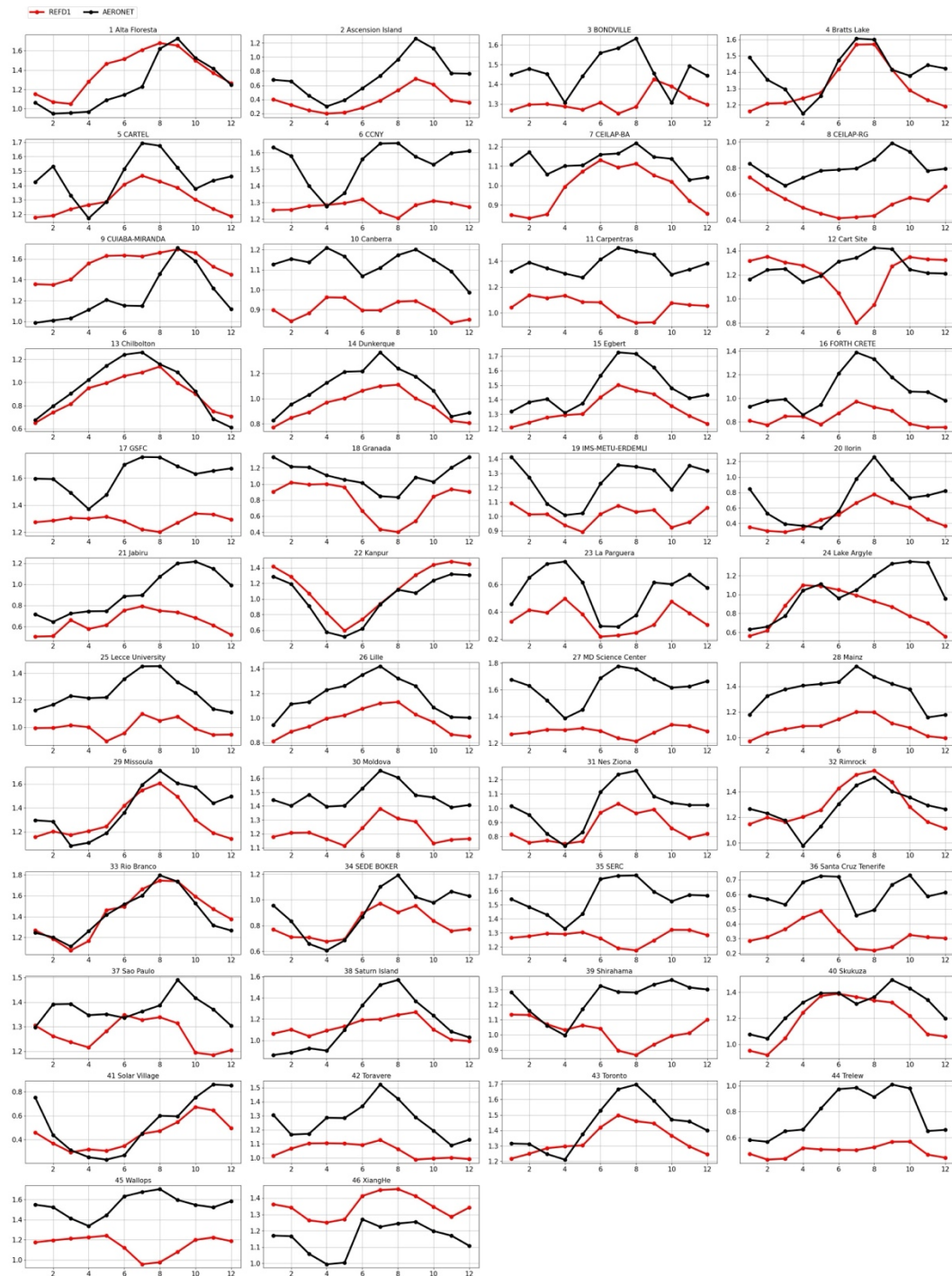
Supplemental Figure 8. Climatological annual average difference in total AOD between GEOSCCM and AOD measured at selected AERONET sites (2003-2018).



Supplemental Figure 9. Climatological monthly average difference in total AE between GEOSCCM and AE measured at selected AERONET sites (2003-2018).



Supplemental Figure 10. Climatological annual cycle difference in AOD between GEOSCCM (speciated) and observations (total) at each selected AERONET sites.

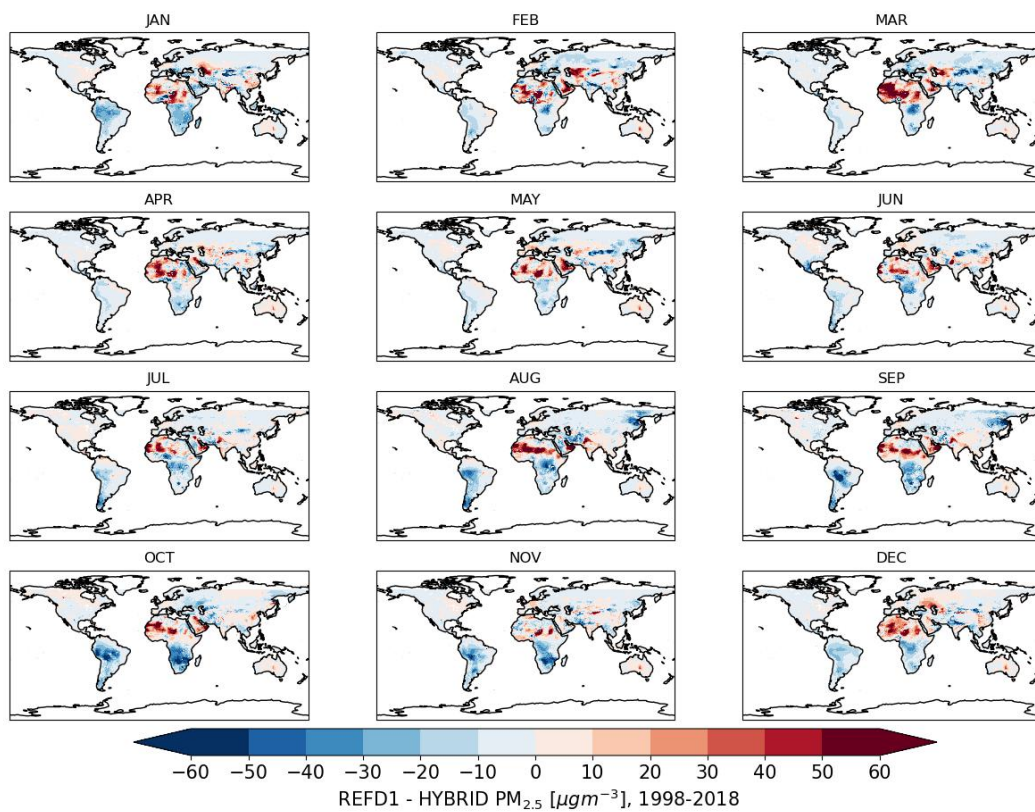


Supplemental Figure 11. Climatological annual cycle difference in total AE between GEOSCCM and observations at each selected AERONET sites.

Supplemental Table 4. Statistics for monthly comparison of AOD and AE between GEOSCCM and selected AERONET sites.

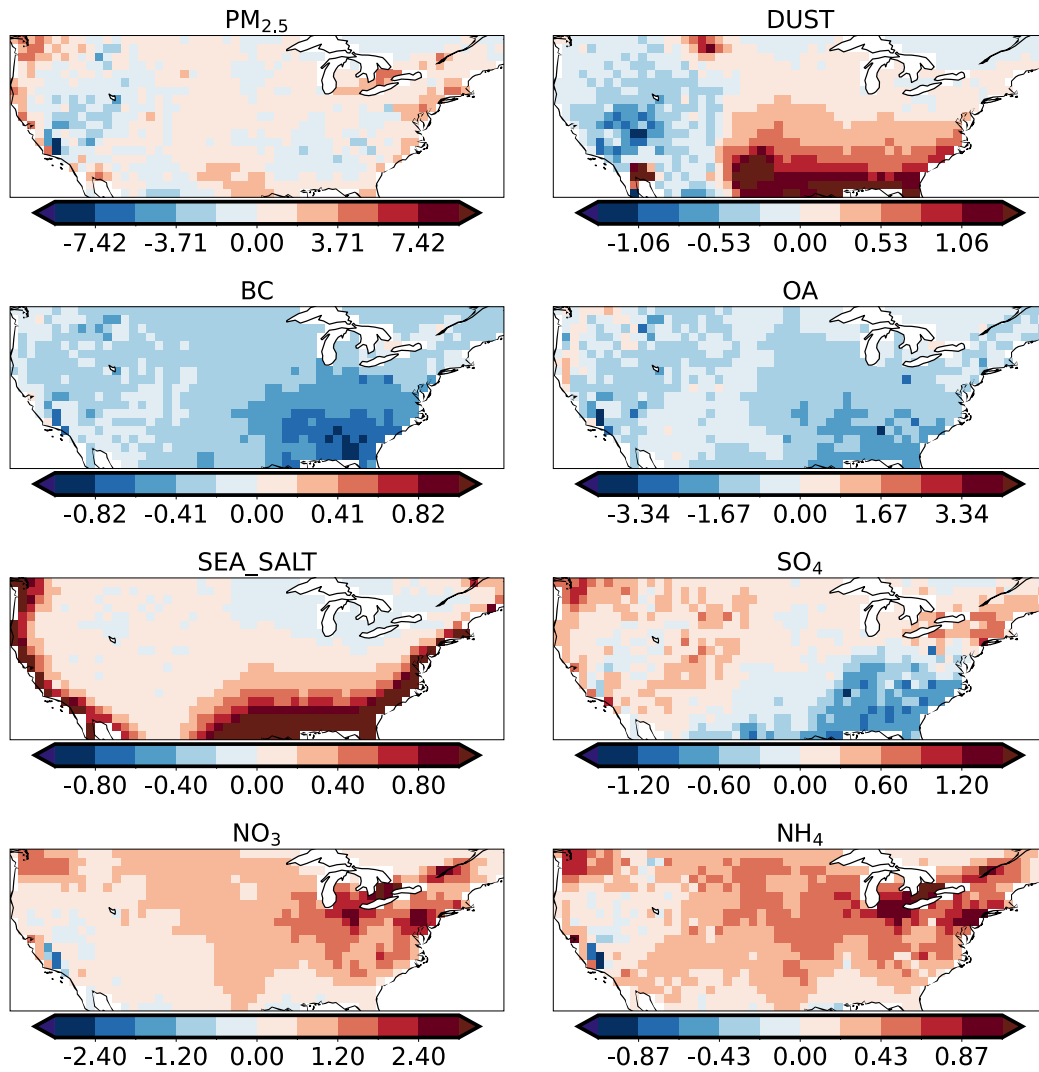
ID	station	MB_AOD	NMB_AOD	RMSE_AOD	r_AOD	MB_AE	NMB_AE	RMSE_AE	r_AE
1	Alta Floresta	-0.15	-0.61	0.26	0.83	0.13	0.10	0.31	0.55
2	Ascension Island	-0.04	-0.24	0.07	0.31	-0.34	-0.46	0.39	0.83
3	BONDVILLE	0.00	0.00	0.09	0.46	-0.16	-0.11	0.32	-0.08
4	Bratts Lake	0.01	0.13	0.05	0.37	-0.11	-0.08	0.28	0.29
5	CARTEL	0.03	0.30	0.06	0.29	-0.16	-0.11	0.30	0.32
6	CCNY	0.02	0.14	0.06	0.79	-0.28	-0.18	0.37	-0.11
7	CEILAP-BA	-0.03	-0.32	0.05	0.32	-0.14	-0.12	0.28	0.09
8	CEILAP-RG	0.01	0.47	0.02	0.12	-0.28	-0.35	0.39	-0.10
9	CUIABA-MIRANDA	-0.12	-0.57	0.21	0.85	0.30	0.24	0.41	0.42
10	Canberra	-0.01	-0.12	0.06	0.92	-0.25	-0.22	0.37	0.14
11	Carpentras	0.00	0.00	0.05	0.25	-0.34	-0.25	0.41	-0.13
12	Cart Site	0.00	-0.01	0.05	0.44	-0.04	-0.03	0.31	-0.29
13	Chilbolton	0.00	0.03	0.06	0.33	-0.09	-0.09	0.27	0.57
14	Dunkerque	0.02	0.11	0.06	0.29	-0.15	-0.14	0.29	0.50
15	Egbert	0.03	0.30	0.06	0.26	-0.15	-0.10	0.29	0.30
16	FORTH CRETE	0.00	-0.03	0.05	0.32	-0.24	-0.22	0.37	0.23
17	GSFC	0.02	0.13	0.07	0.82	-0.33	-0.20	0.38	-0.03
18	Granada	-0.01	-0.09	0.06	0.34	-0.33	-0.30	0.44	0.36
19	IMS-METU-ERDEMLI	-0.08	-0.35	0.13	0.04	-0.24	-0.20	0.36	0.13
20	Ilorin	-0.14	-0.22	0.50	0.33	-0.24	-0.33	0.33	0.58
21	Jabiru	-0.04	-0.34	0.07	0.59	-0.30	-0.31	0.41	0.41
22	Kanpur	-0.10	-0.16	0.20	0.41	0.14	0.14	0.25	0.81
23	La Parguera	0.06	0.47	0.08	0.80	-0.21	-0.38	0.31	0.36
24	Lake Argyle	-0.04	-0.37	0.07	0.75	-0.22	-0.21	0.41	0.21
25	Lecce University	0.01	0.04	0.06	0.22	-0.29	-0.23	0.39	0.03
26	Lille	0.02	0.11	0.06	0.21	-0.21	-0.18	0.30	0.49
27	MD Science Center	0.01	0.04	0.08	0.77	-0.34	-0.21	0.42	0.04
28	Mainz	0.02	0.14	0.06	0.41	-0.29	-0.21	0.33	0.50
29	Missoula	-0.03	-0.32	0.10	0.46	-0.10	-0.07	0.31	0.42
30	Moldova	0.03	0.18	0.08	0.28	-0.26	-0.18	0.31	0.30
31	Nes Ziona	-0.02	-0.10	0.07	0.28	-0.15	-0.14	0.28	0.41
32	Rimrock	-0.01	-0.15	0.09	0.15	0.02	0.02	0.26	0.34

33	Rio Branco	-0.13	-0.64	0.20	0.89	0.01	0.01	0.23	0.64
34	SEDE BOKER	0.01	0.05	0.06	0.35	-0.11	-0.12	0.27	0.39
35	SERC	0.01	0.08	0.07	0.80	-0.30	-0.20	0.39	-0.40
36	Santa Cruz Tenerife	0.05	0.31	0.10	0.43	-0.31	-0.50	0.35	0.32
37	Sao Paulo	-0.06	-0.28	0.09	0.55	-0.11	-0.08	0.19	0.01
38	Saturn Island	0.02	0.24	0.07	0.15	-0.05	-0.04	0.33	0.40
39	Shirahama	-0.03	-0.13	0.10	0.41	-0.20	-0.16	0.30	-0.14
40	Skukuza	-0.06	-0.30	0.09	0.37	-0.11	-0.08	0.24	0.47
41	Solar Village	-0.01	-0.04	0.12	0.60	-0.09	-0.17	0.25	0.51
42	Toravere	0.03	0.26	0.07	0.01	-0.24	-0.18	0.34	0.20
43	Toronto	0.02	0.14	0.07	0.11	-0.12	-0.08	0.21	0.52
44	Trelew	0.01	0.16	0.02	0.21	-0.29	-0.37	0.42	0.19
45	Wallops	0.01	0.08	0.06	0.81	-0.41	-0.26	0.49	-0.25
46	XiangHe	-0.07	-0.10	0.34	0.19	0.20	0.17	0.23	0.51

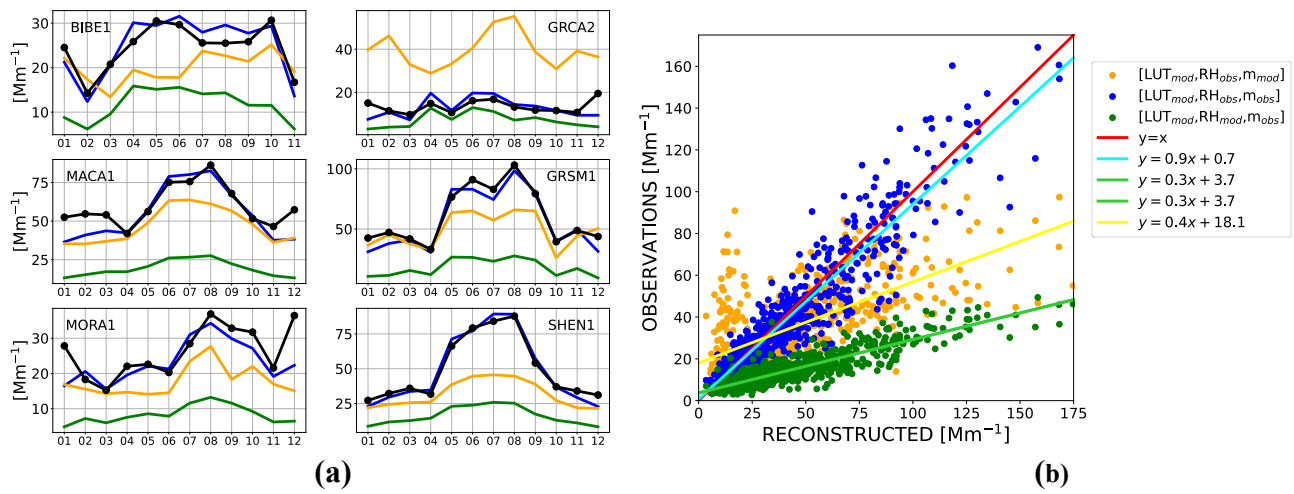


Supplemental Figure 12. Climatological monthly average difference in total PM_{2.5} between GEOSCCM and the model-observation hybrid dataset (1998-2018).

REFD1 - SAT DERIVED mean 2000-2016 [$\mu\text{g m}^{-3}$]



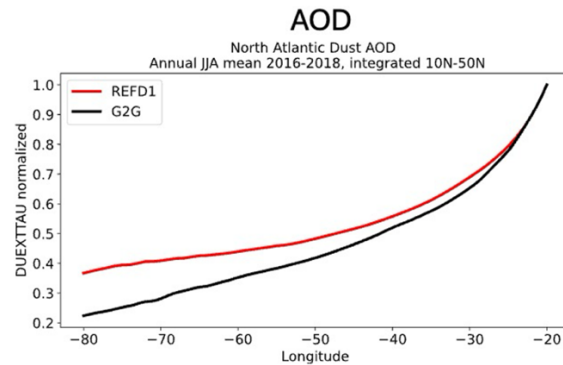
Supplemental Figure 13: mean annual difference in speciated $\text{PM}_{2.5}$ between GEOSCCM Ref-D1 and satellite-derived speciated $\text{PM}_{2.5}$ (2000-2016).



Supplemental Figure 14. Same as Figure 9 but with the nitrate component removed in the scattering coefficient reconstruction.

station	NMB 1	NMB 2	NMB 3	RMSE 1 [Mm-1]	RMSE 2 [Mm-1]	RMSE 3 [Mm-1]	r 1	r 2	r 3
	0.05	-0.5	-0.19	4.48	14.16	9.04	0.83	0.65	0.19
GRCA2	0.03	-0.42	2.08	5.66	7.23	31.24	0.63	0.56	0.32
GRSM1	-0.05	-0.7	-0.23	14.07	55.71	27.9	0.93	0.88	0.79
MACA1	-0.1	-0.68	-0.22	15.08	44.72	22.57	0.85	0.81	0.68
MORA1	-0.06	-0.65	-0.3	9.9	20.58	15.75	0.71	0.63	0.28
SHEN1	-0.02	-0.68	-0.36	9.48	40.03	29.18	0.96	0.94	0.79

Supplemental Table 5. Same as Table 3, but with the nitrate component removed in the scattering coefficient reconstruction.



Supplemental Figure 15. Comparison of summertime dust AOD and dust mass transport over the North Atlantic between Ref-D1 run (free running meteorology), and the GOCART-2G run (replayed meteorology) averaged for JJA 2016-2018.