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Supplement of

Concentrations and radiative forcing of anthropogenic aerosols from 1750 to 2014 simulated with the Oslo CTM3 and CEDS emission inventory

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Table S1: Total global emissions of main aerosols and precursors in 2010 [Tg] in the three emission inventories CEDS version 2016 (Hoesly et al., 2018), RCP4.5 (Thomson et al., 2011) and ECLIPSEv5 (Klimont et al., 2017) (excluding agricultural waste burning and international aviation).

	BC	OC	NO _x	SO ₂
CEDSv16	7.75	18.3	137.2	113.8
RCP4.5	5.61	13.4	101.5	108.2
ECLIPSEv5	7.25	15.5	120.3	102.5

Table S2: Summary of bias, error and correlation of modeled against measured surface concentrations by region/network.

		N. America (IMPROVE)	Europe (EMEP, ACTRIS)	China (CAWNET) (Zhang et al., 2012)	N. America (CASTNET)	India (Kumar et al., 2015)
EC	RMSE	0.20	0.56	3.1	-	2.8
	NMB	6.1	18	-18	-	-43
	R	0.54	0.29	0.66	-	0.60
OC	RMSE	0.43	1.3	16	-	-
	NMB	-20	-16	-58	-	-
	R	0.68	0.34	0.49	-	-
Nitrate	RMSE	0.43	0.91	8.1	0.75	-
	NMB	4.7	20	-58	-11	-
	R	0.46	0.76	0.74	0.68	-
Sulfate	RMSE	0.42	1.8	21	1.2	-
	NMB	-12	-40	-76	-43	-
	R	0.87	0.20	0.89	0.95	-

Table S3: Atmospheric residence time (ratio of burden to total wet plus dry deposition) [days] of each aerosol species in the sensitivity simulations.

Simulation	BC	OA	Sulfate	NH4	Nitrate (fine)	Nitrate (coarse)	Sea salt	Dust
ECLv5	4.4	5.3	5.2	3.6	4.6	7.2	0.46	3.4
RCP-CMIP5	4.3	5.3	5.3	3.6	4.7	7.5	0.46	3.4
LSIINC	4.0	4.6	4.9	3.3	3.9	4.7	0.43	3.4
LSIDEC	6.2	9.0	6.5	4.2	4.6	6.2	0.54	3.8
SOLDEC	4.9	6.0	6.1	4.1	4.5	6.4	0.59	3.6
1x1	4.6	5.5	5.7	3.9	4.6	5.2	0.44	3.3
MET	4.3	5.0	5.5	3.6	4.0	5.1	0.48	3.5

Table S4: Historical evolution of aerosol burdens [mg m^{-2}]. Note that for consistency with RF calculations, OA is the sum of total secondary OA and primary OA from fossil fuel plus biofuel and biomass is the sum of BC and primary OA from biomass burning sources. Coarse model nitrate and ammonium contributes little to RF and are excluded.

Year	BC	OA	Biomass	Sulfate	Nitrate	NH4
1750	0.012	0.79	1.1	3.4	0.023	0.18
1850	0.018	0.84	1.2	3.4	0.027	0.20
1900	0.031	0.92	1.2	3.7	0.025	0.24
1910	0.036	0.95	1.2	3.9	0.027	0.25
1920	0.038	0.96	1.2	3.9	0.027	0.26
1930	0.039	0.99	1.2	3.9	0.030	0.28
1940	0.043	1.0	1.2	4.1	0.024	0.29
1950	0.046	1.1	1.3	4.2	0.030	0.32
1960	0.066	1.2	1.1	4.7	0.024	0.36
1970	0.075	1.3	1.2	5.4	0.035	0.45
1980	0.10	1.4	1.2	5.7	0.053	0.53
1985	0.10	1.4	1.1	5.6	0.061	0.56
1990	0.12	1.5	1.2	5.7	0.072	0.59
1995	0.12	1.5	1.6	5.5	0.11	0.62
2000	0.12	1.5	1.2	5.3	0.10	0.59
2005	0.14	1.6	1.4	5.5	0.13	0.64
2010	0.16	1.7	1.5	5.4	0.17	0.68
2014	0.17	1.7	1.4	5.4	0.15	0.69

Table S5: Changes in burden, AOD, AAOD, RFari, and normalized RF over the period 1750-2010 for individual aerosol components and the net RFari.

Aerosol Component	Burden (mg m ⁻²)	AOD (550 nm)	AOD ant fraction (%)	AAOD (550 nm)	RF (W m ⁻²)	Normalized RF (W g ⁻¹)	Normalized RF (W m ⁻²)
Sulfate	2.05	0.018	40	0	-0.31	-151	-17
BC (FF+BF)	0.15	0.0023	92	0.0018	0.30	2014	130
POA (FF+BF)	0.53	0.0034	79	0.0001	-0.06	-108	-17
SOA	0.42	0.0026	37	0.0001	-0.03	-77	-12
Biomass	0.38	0.0021	24	0.0003	0.02	53	10
Nitrate	0.14	0.0014	89	0	-0.02	-155	-16
Total	3.66	0.029	22	0.0024	-0.16	-45	-6

FF is fossil fuel emissions and BF is biofuel emissions.

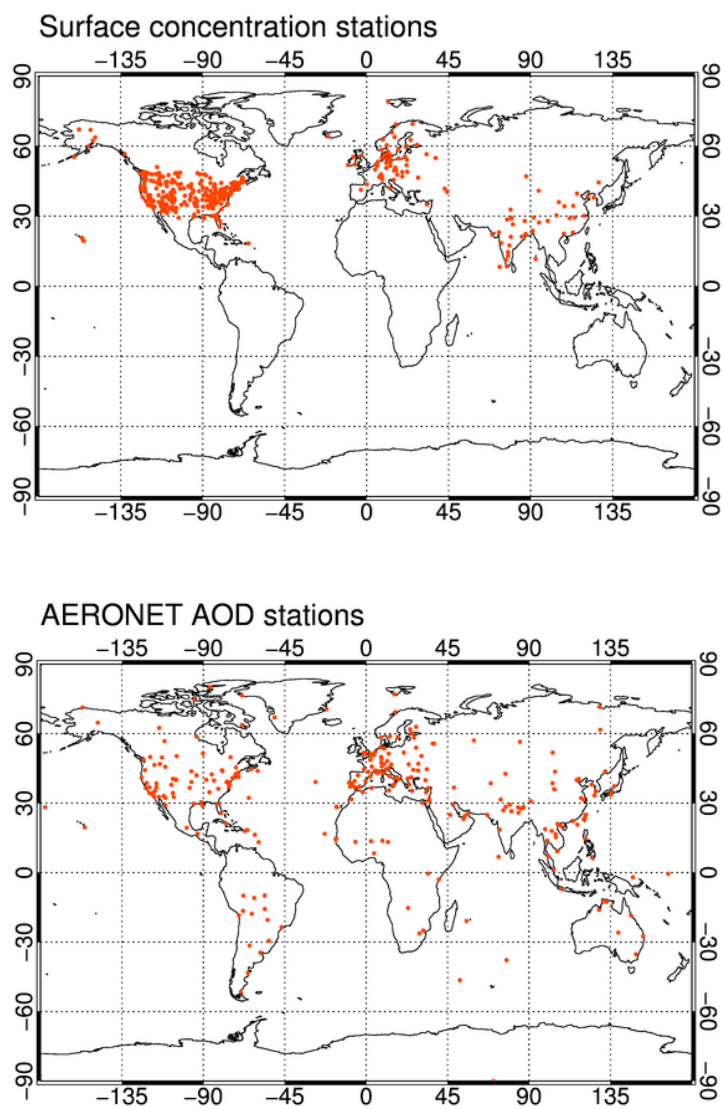


Figure S1: Geographical coverage of surface concentration (top) and AOD (bottom) observations

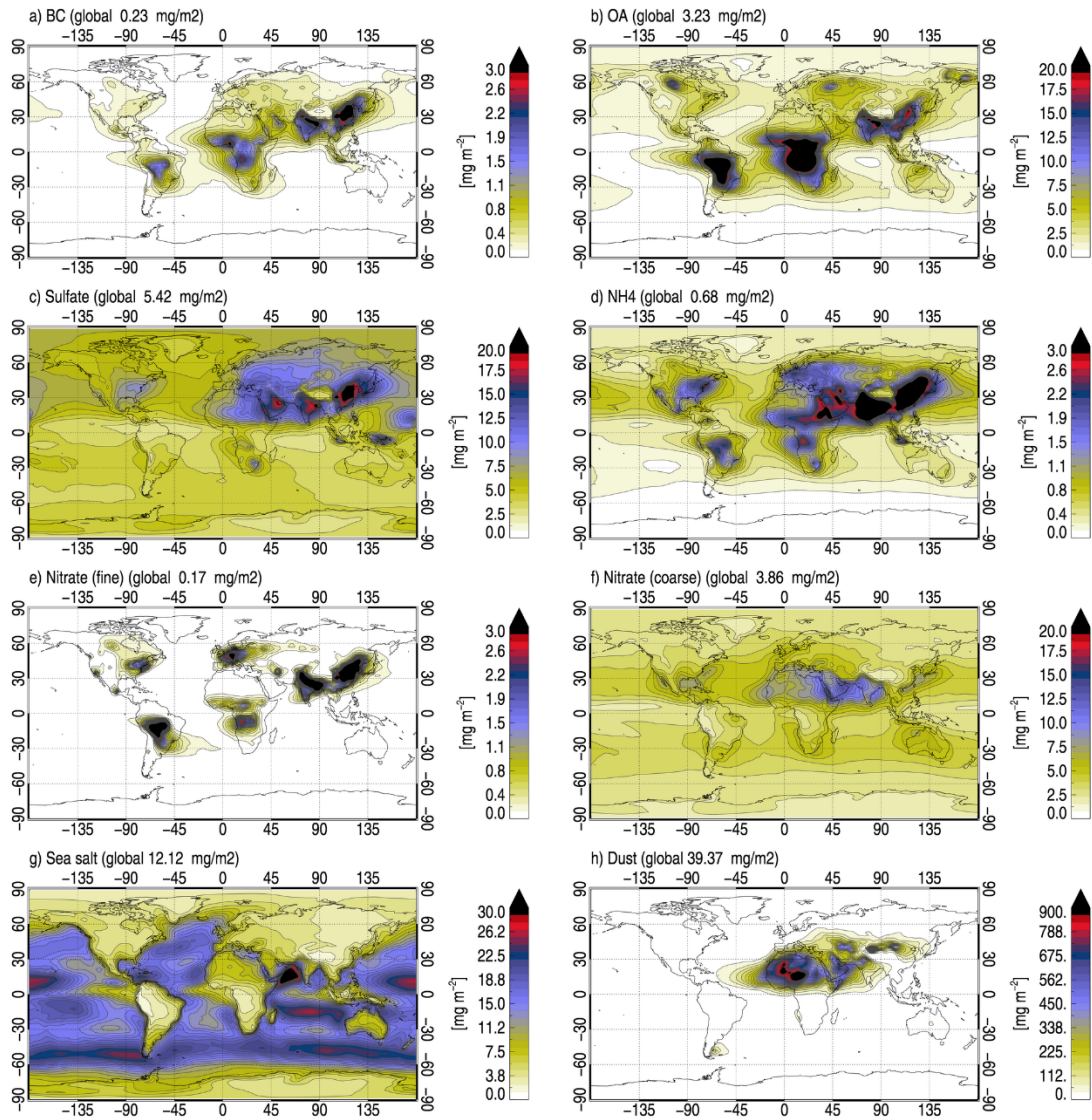


Figure S2: Annual mean (year 2010) aerosol burdens in the OsloCTM3 CEDSv16-CMIP6 baseline simulation.

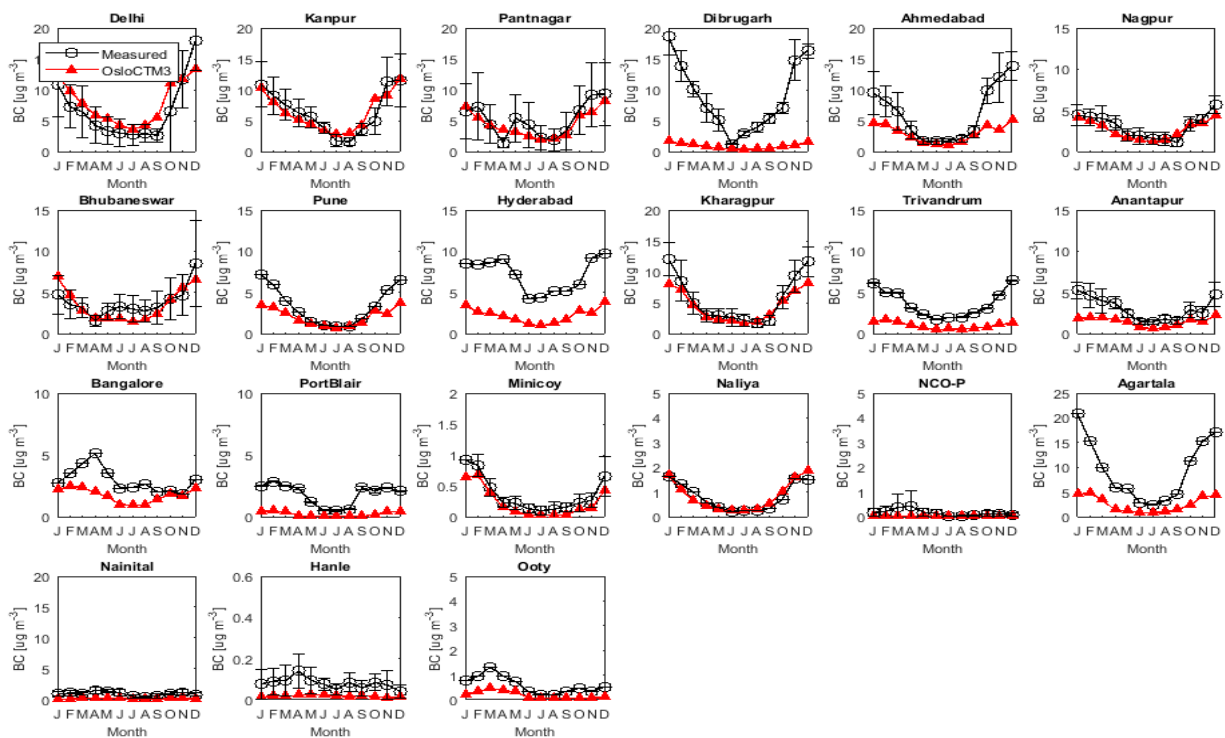


Figure S3: Modeled (year 2010) and measured monthly BC surface concentrations over India (observations from Kumar et al. (2015)).

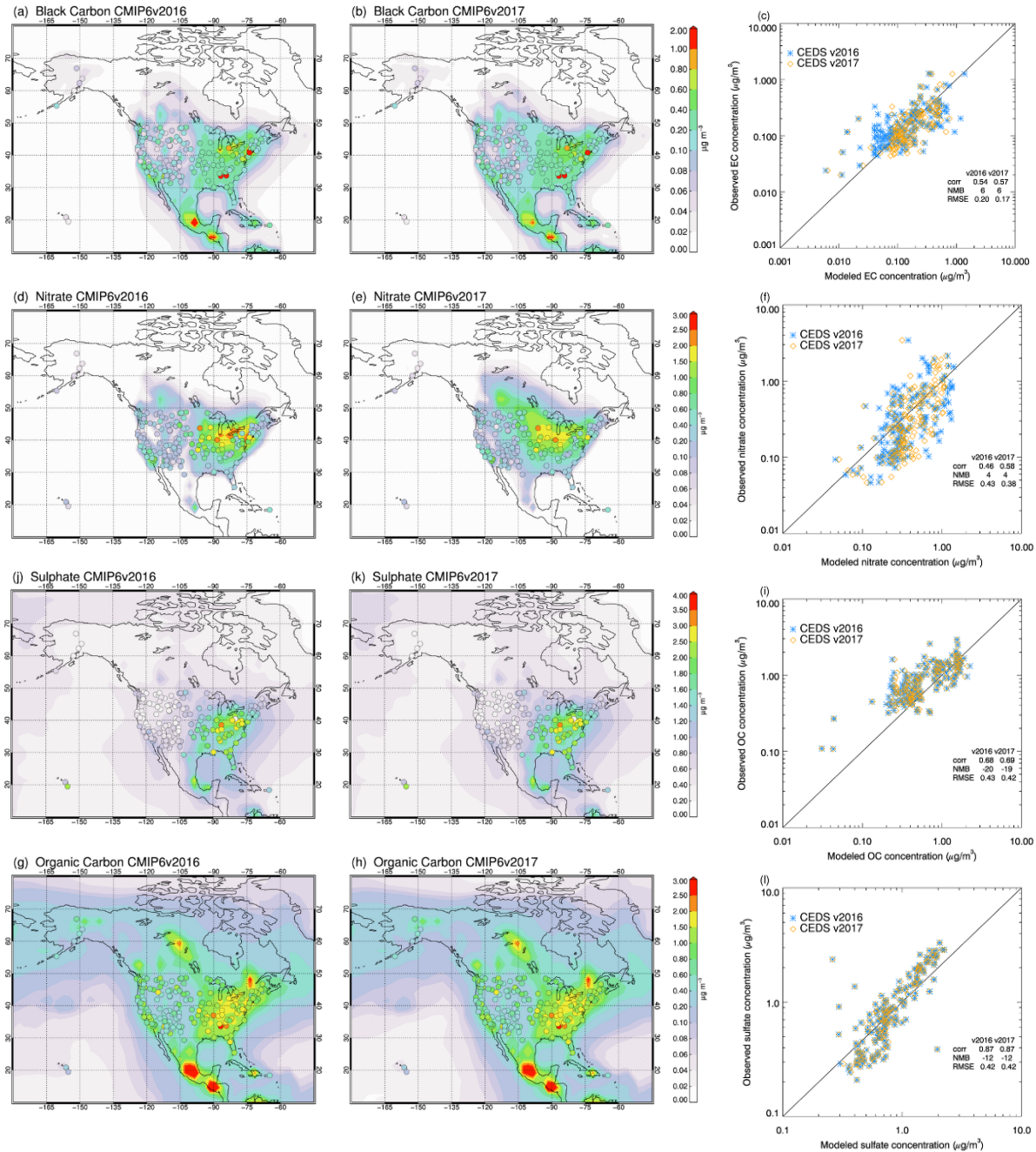


Figure S4: Modeled annual mean surface concentration of BC (a), nitrate (d), OC (g) and sulfate (j) over North America for year 2010 using the 2017 version of the CEDES emissions with observed concentrations from the IMPROVE network indicated with filled circles (left column). Middle column same as left column but using the 2016 version of the CEDES emissions. Modeled annual mean surface concentration of BC (c), nitrate (f), OC (i) and sulfate (l) for the year 2010 versus measured concentrations from the IMPROVE network with old (blue) and new (orange) emissions (right column).

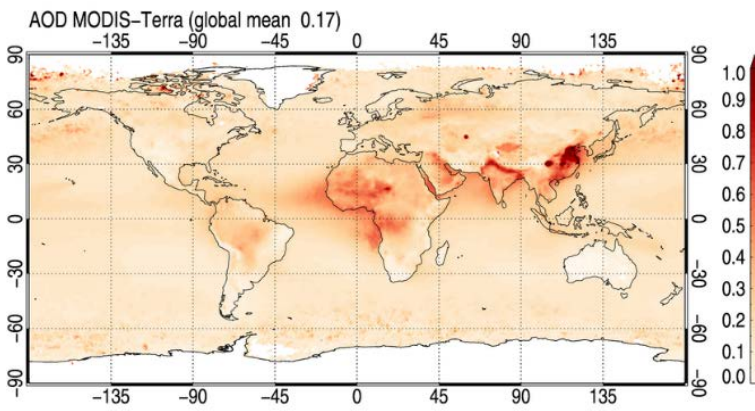
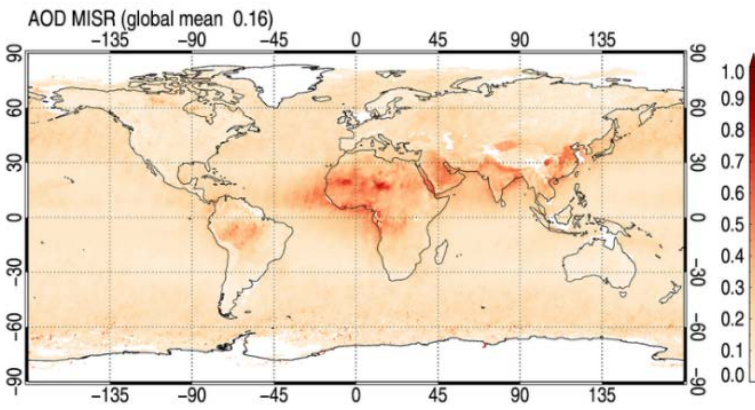


Figure S5: AOD from MISR (top) and MODIS-Terra (bottom)

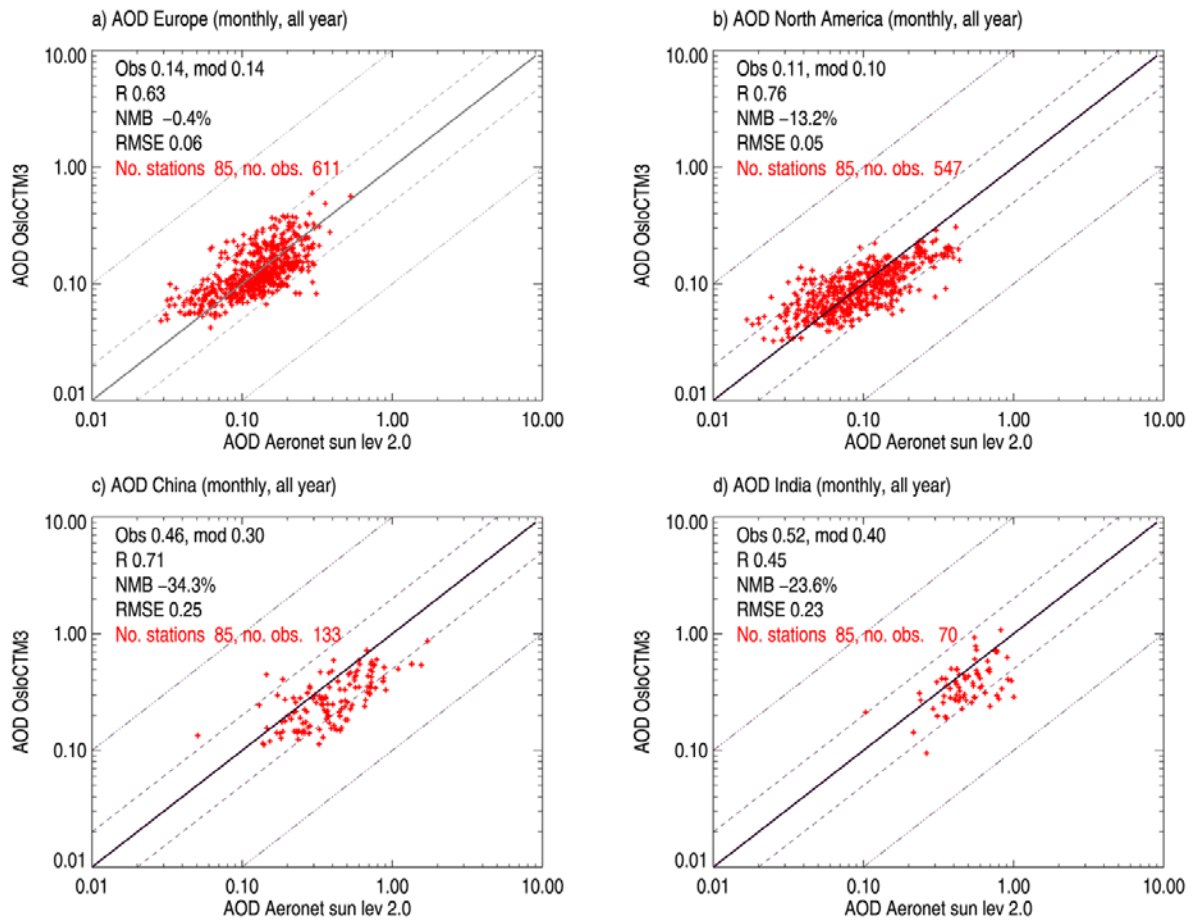


Figure S6: Modeled AOD against observations from AERONET in a) Europe, b) North America, c) China and d) India.

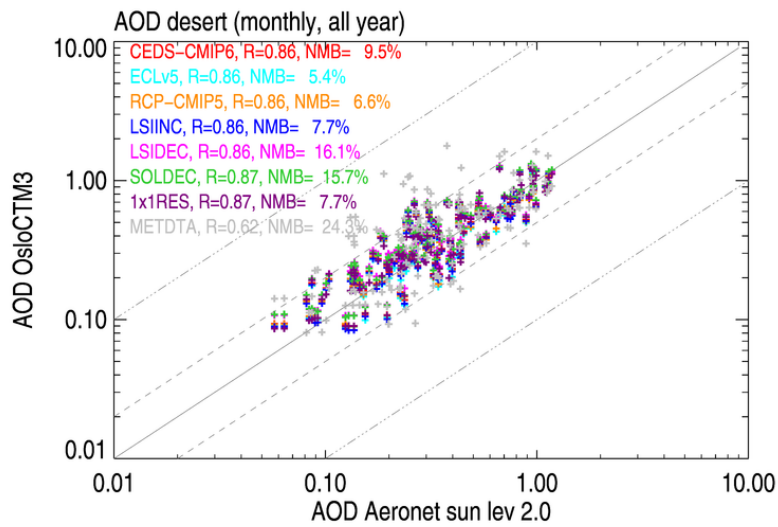
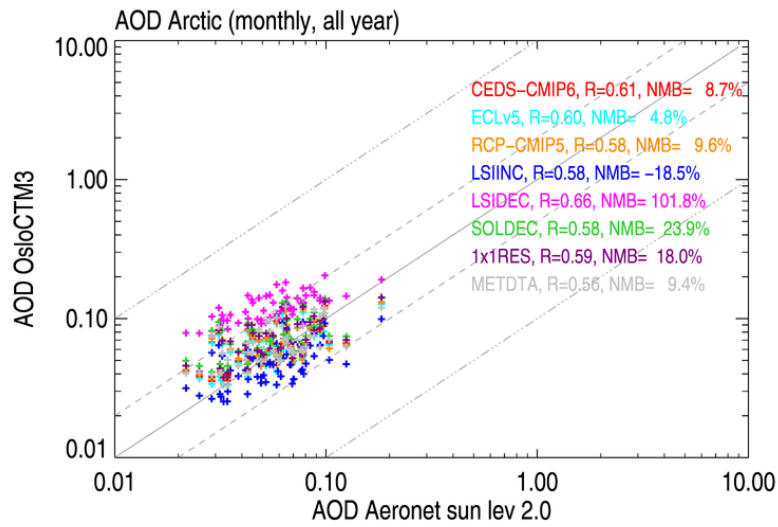


Figure S7: Modeled AOD in each of the sensitivity tests against AERONET stations in the Arctic, i.e., north of 65°N , (top) and North Africa/Middle East (bottom).

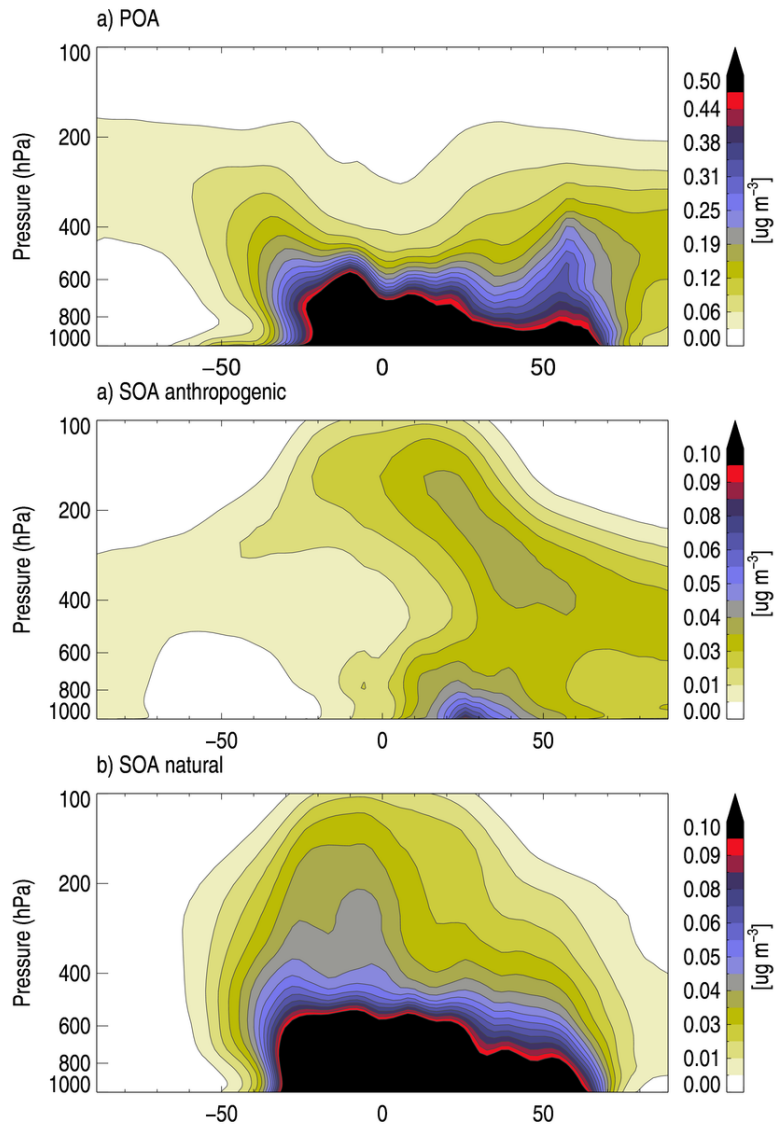


Figure S8: Zonal, annual mean concentration of primary organic aerosol (POA) and secondary organic aerosol (SOA) from anthropogenic and natural emissions simulated by OsloCTM3.

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