Supplementary information

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	NOx	SO2
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 [*Zhang et al.*, 2012]	N. America (CASTNET)	India [*Kumar et al.*, 2015]
EC	RMSE	0.20	0.56	3.1	-	2.8
	MNB	6.1	18	-18	-	-43
	R	0.54	0.29	0.66	-	0.60
OC	RMSE	0.43	1.3	16	-	-
	MNB	-20	-16	-58	-	-
	R	0.68	0.34	0.49	-	-
Nitrate	RMSE	0.43	0.91	8.1	0.75	-
	MNB	4.7	20	-58	-11	-
	R	0.46	0.76	0.74	0.68	-
Sulfate	RMSE	0.42	1.8	21	1.2	-
	MNB	-12	-40	-76	-43	-
	R	0.87	0.20	0.89	0.95	-

Table S3: Global, annual mean burdens [mg m^{-2}] of primary organic aerosols (POA) by source and total natural plus anthropogenic secondary organic aerosols (SOA) in the CEDSv16/CMIP6 baseline simulation.

POA	Fossil fuel	0.77
	Biomass burning	1.43
	Marine	0.08
SOA	Total	1.13

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

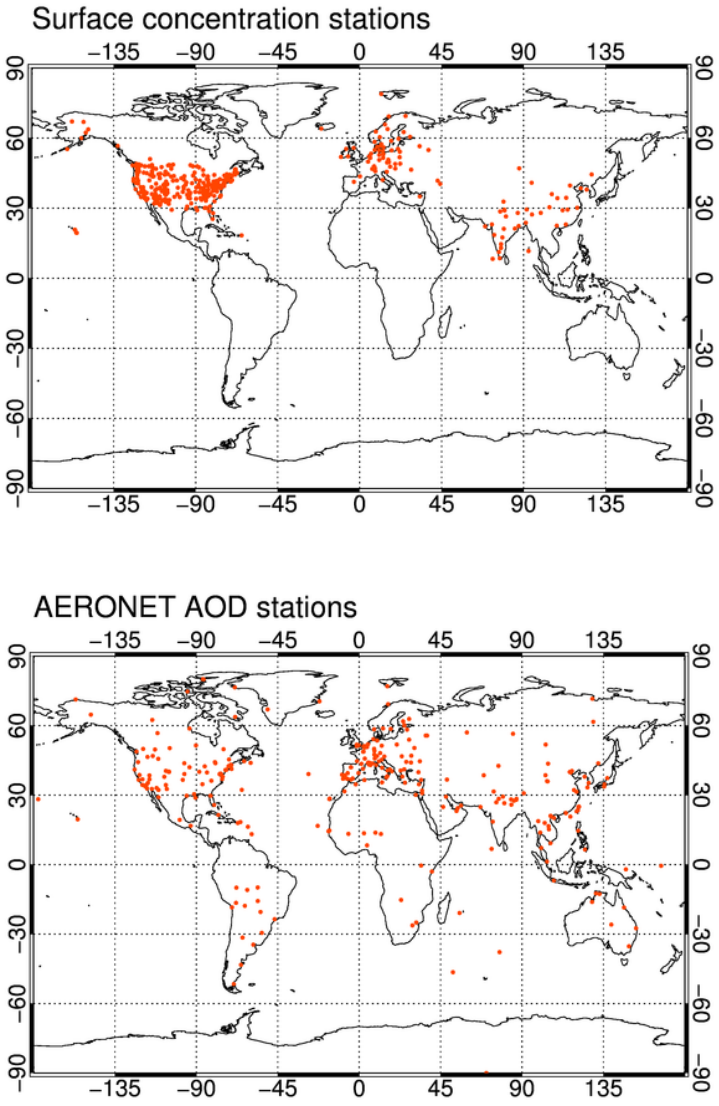

Figure S1: Geographical coverage of surface concentration (top) and AOD (bottom) observations

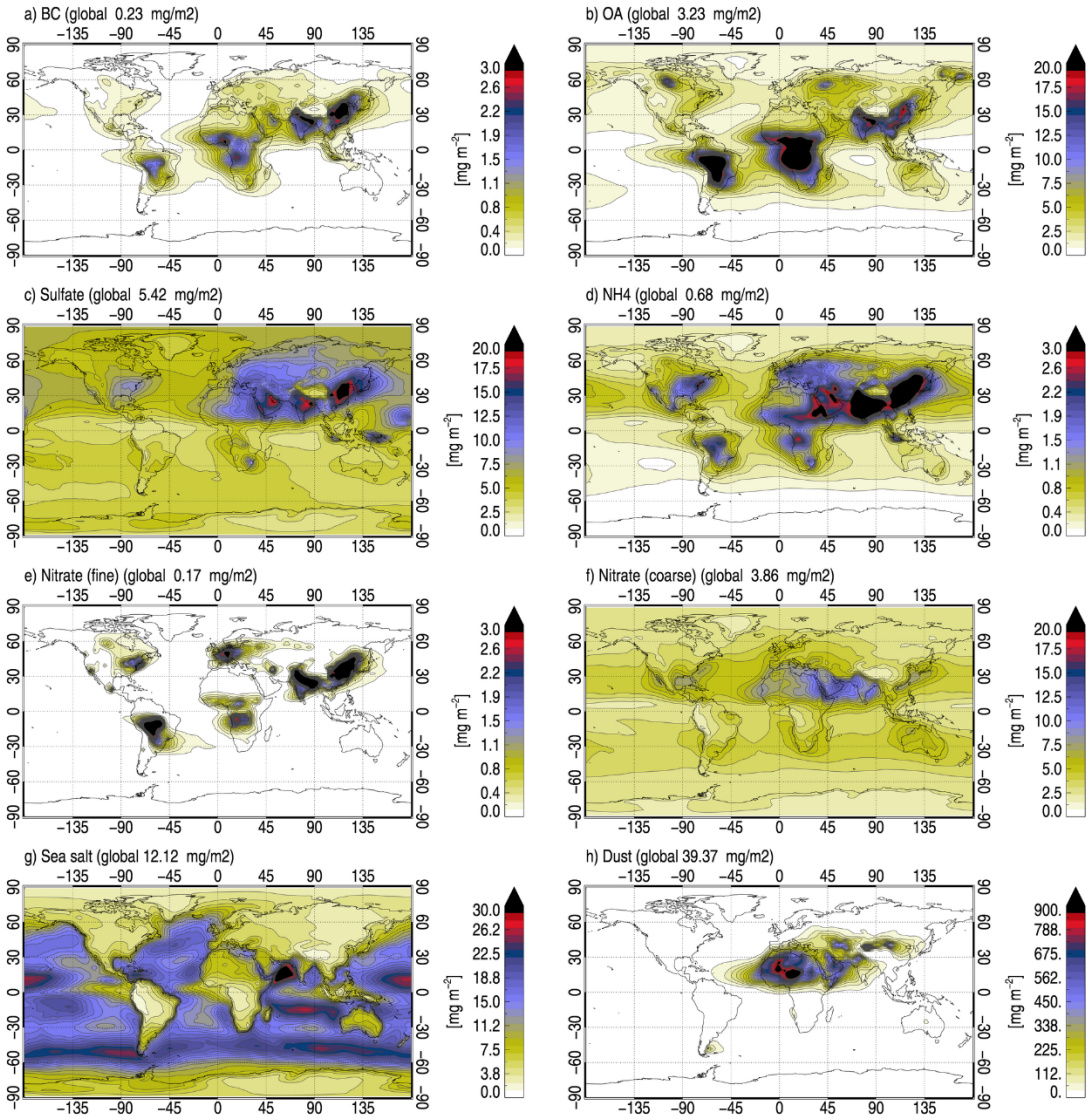

Figure S2: Annual mean (year 2010) aerosol burdens in OsloCTM3 baseline simulation.

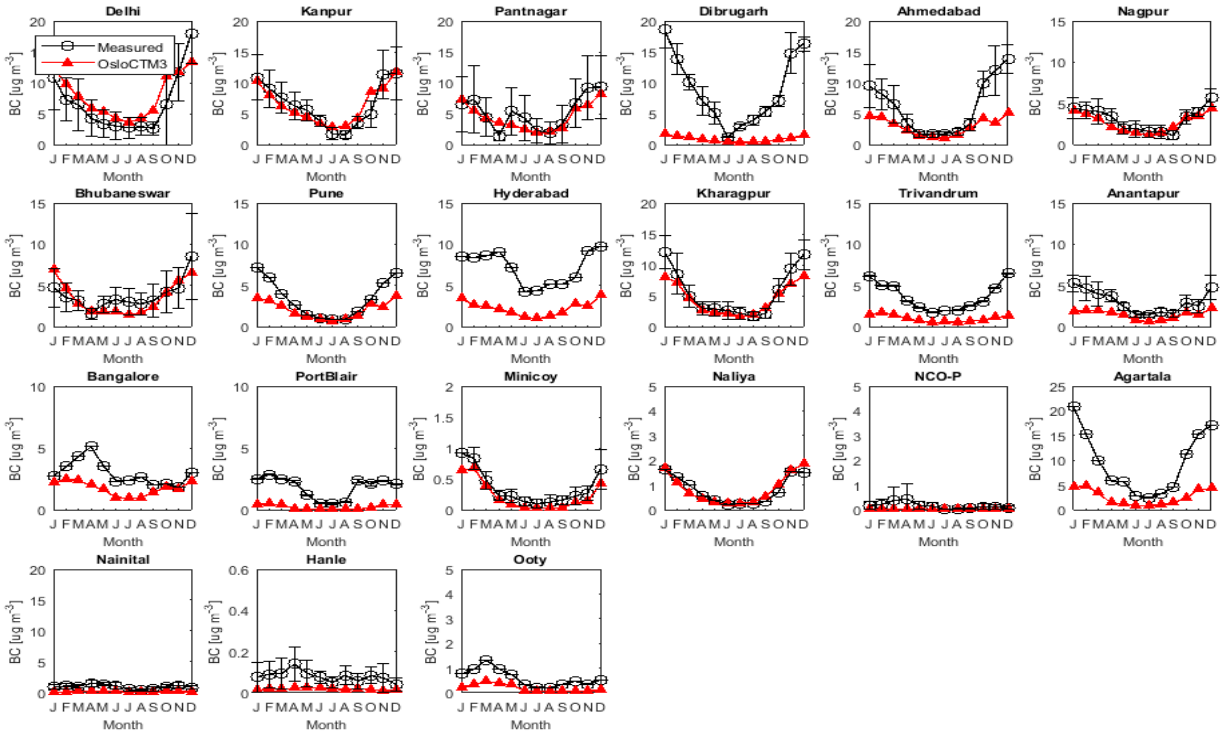

Figure S3: Modeled (year 2010) and measured monthly BC surface concentrations over India (observations from Kumar et al. [2015]).

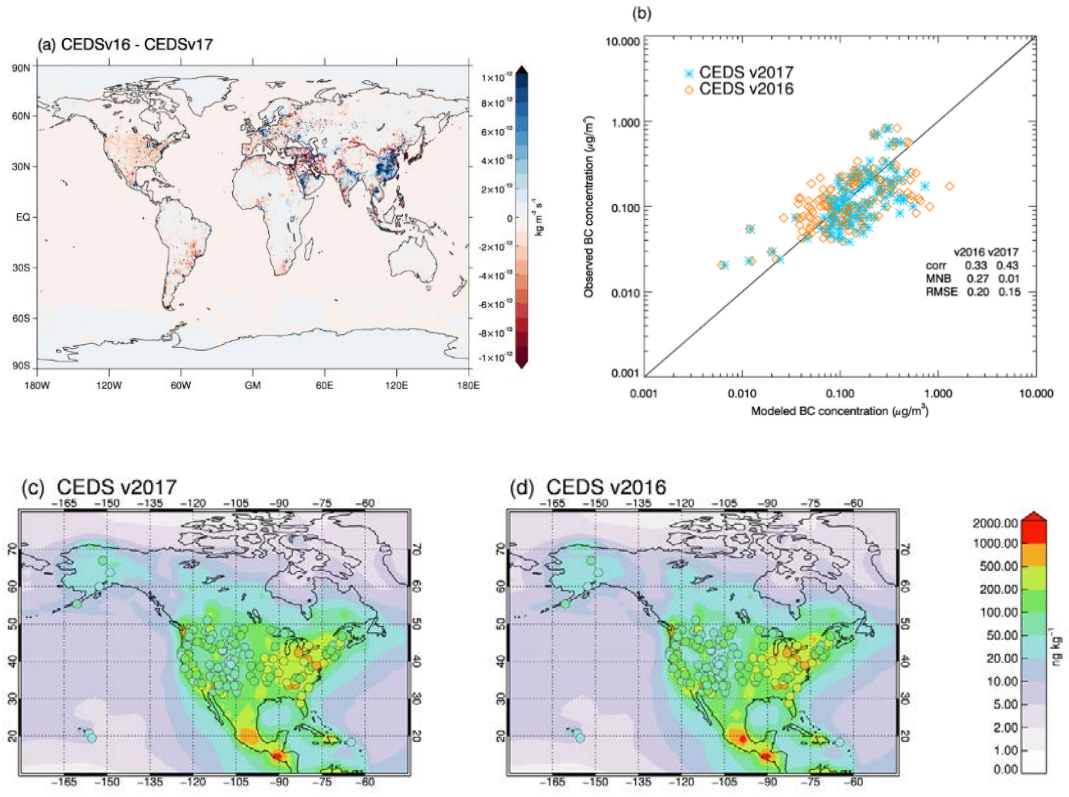

Figure S4: a) Absolute change in annual mean emissions for the year 2000 between CEDS versions released in 2016 and 2017. b) Modeled annual mean BC concentration for the year 2013 versus measured EC from the IMPROVE network with old (orange) and new (blue) emissions. c) Modeled annual mean surface concentration of BC over North America for year 2013 using the 2017 version of the CEDS emissions with observed concentrations from the IMPROVE network indicated with filled circles. d) Same as c) but using the 2016 version of the CEDS emissions.

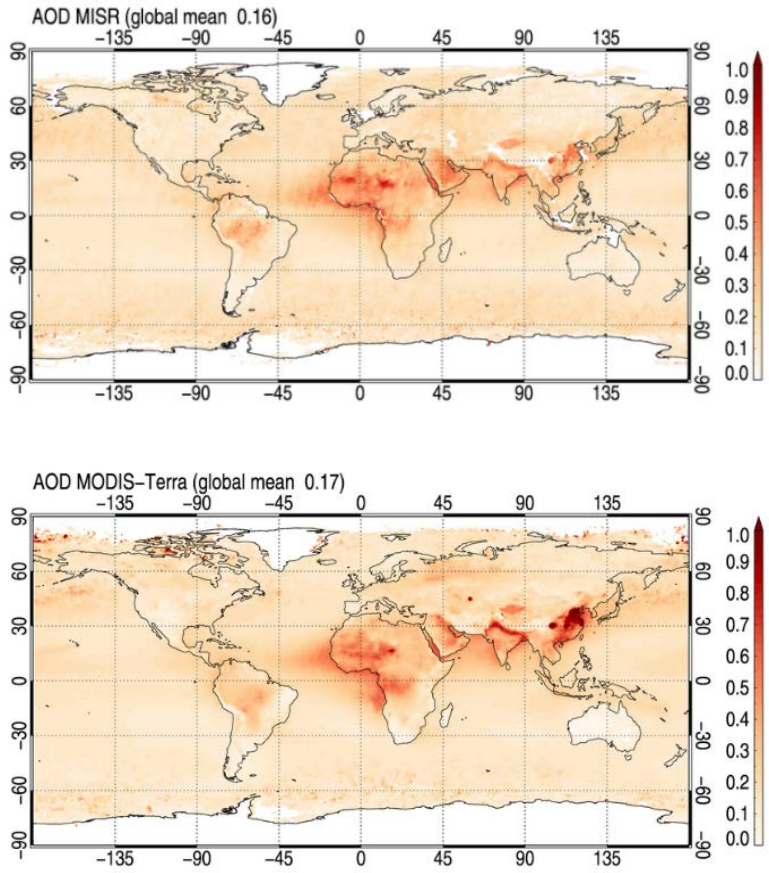

Figure S5: AOD from MISR (top) and MODIS-Terra (bottom)

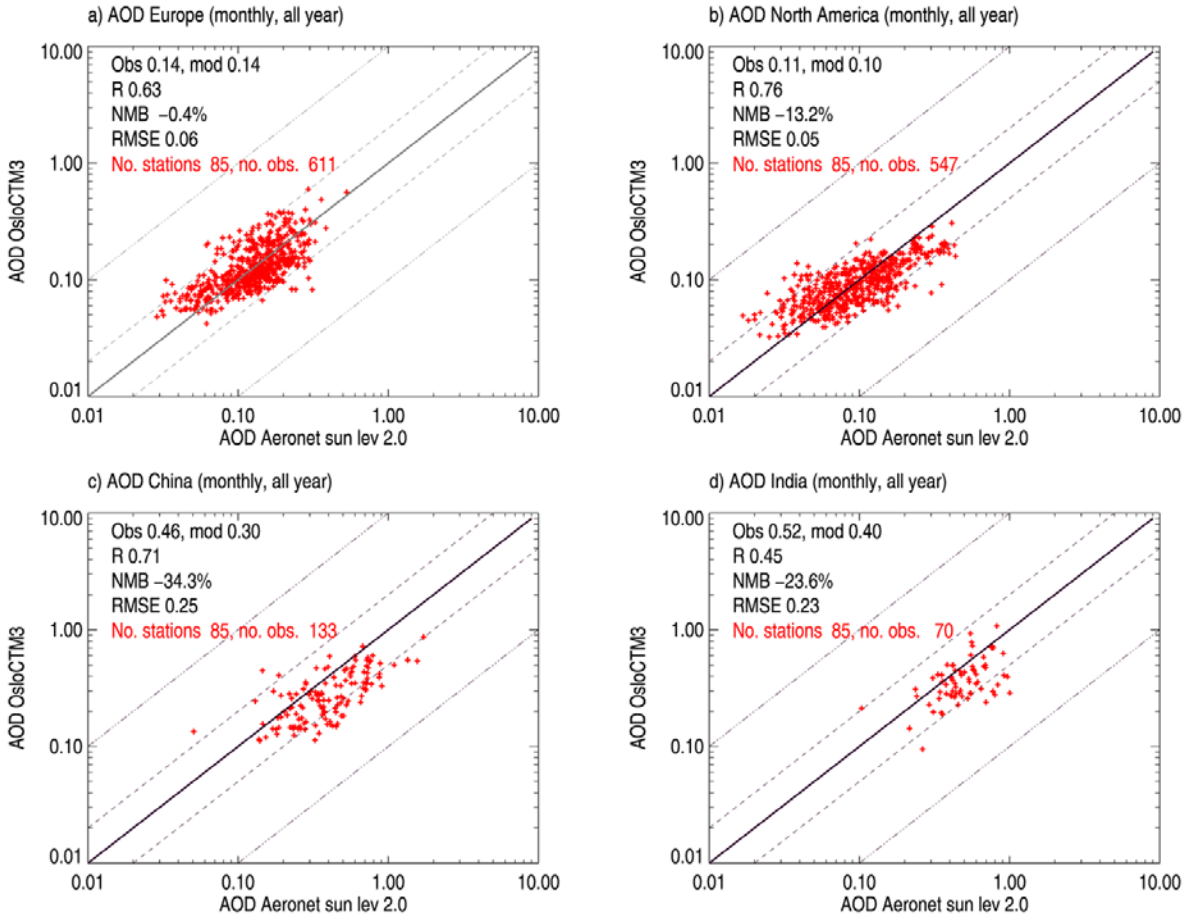

Figure S6: Modeled AOD against observations from AERONET in a) Europe, b) North America, c) China and d) India.

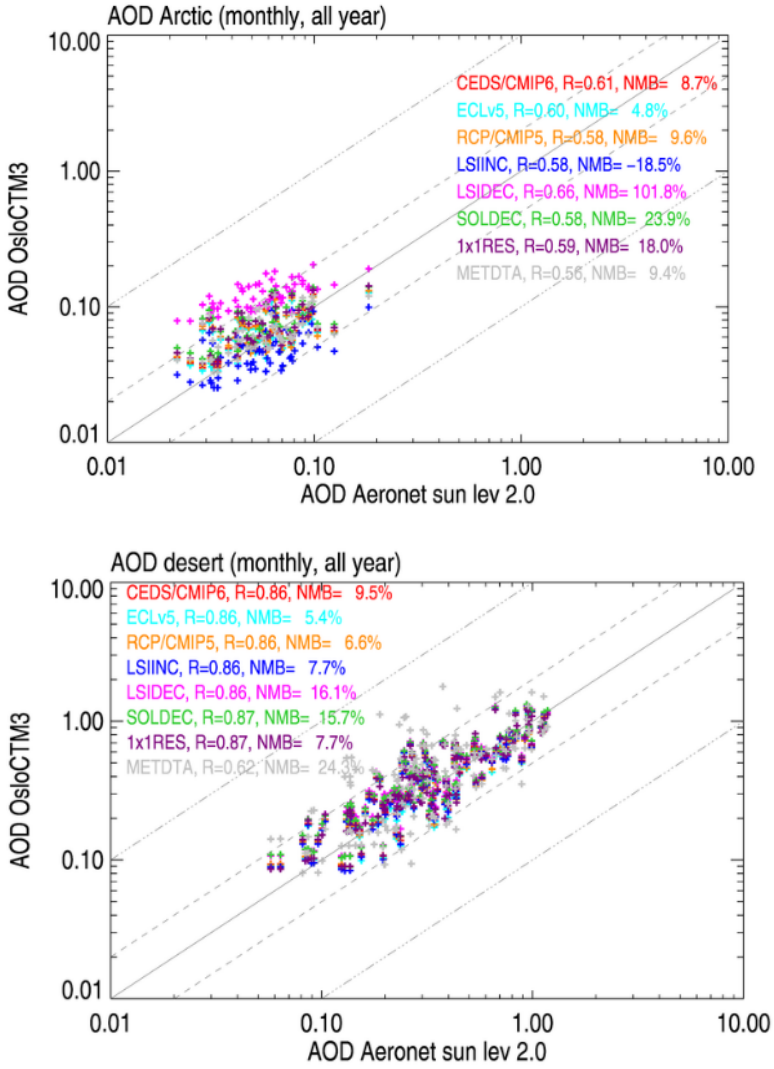

Figure S7: Modeled AOD in each of the sensitivity tests against AERONET stations in the Arctic, i.e., north of 65°N, (top) and North Africa/Middle East (bottom).

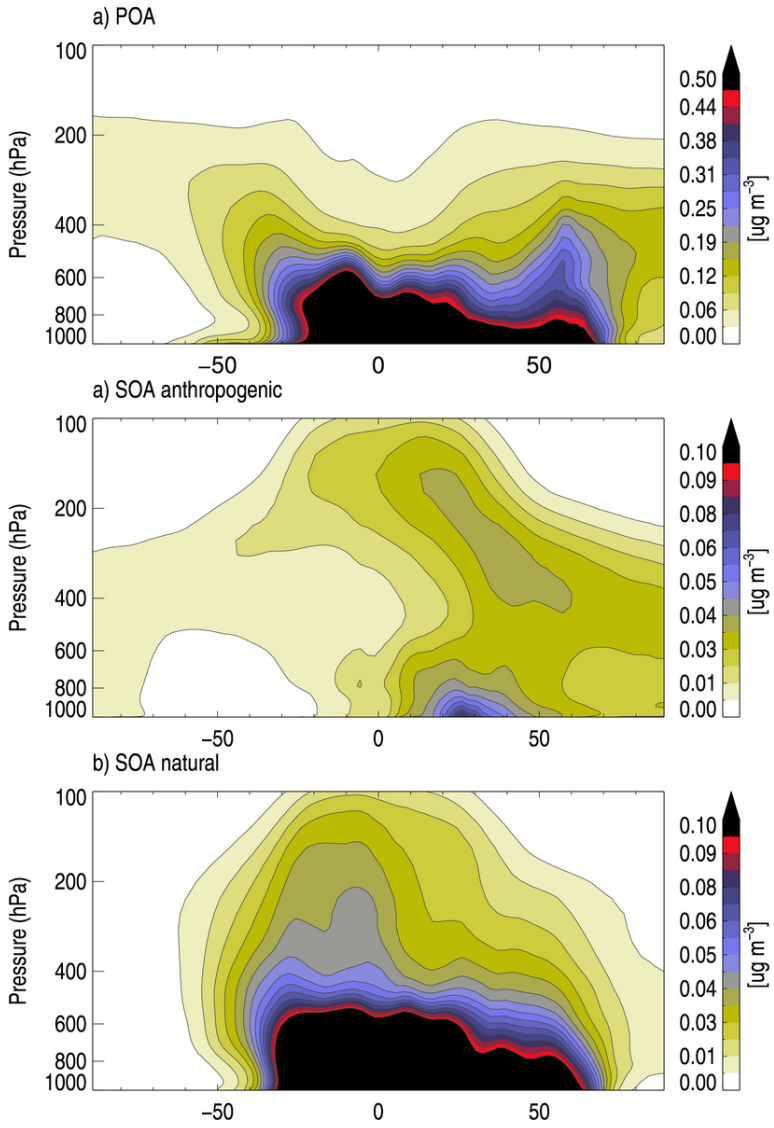

Figure S8: Zonal, annual mean concentration of primary organic aerosol (POA) and secondary organic aerosol (SOA) from anthropogenic and natural emissions simulated by OsloCTM3.

References:

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