Supplementary for "Global modelling of secondary organic aerosol (SOA) with organic nucleation"

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Introduction

This file provides the supporting figures and tables for the main text. The result of sensitivity experiment for the formation of SOA from IEPOX is shown in this file.

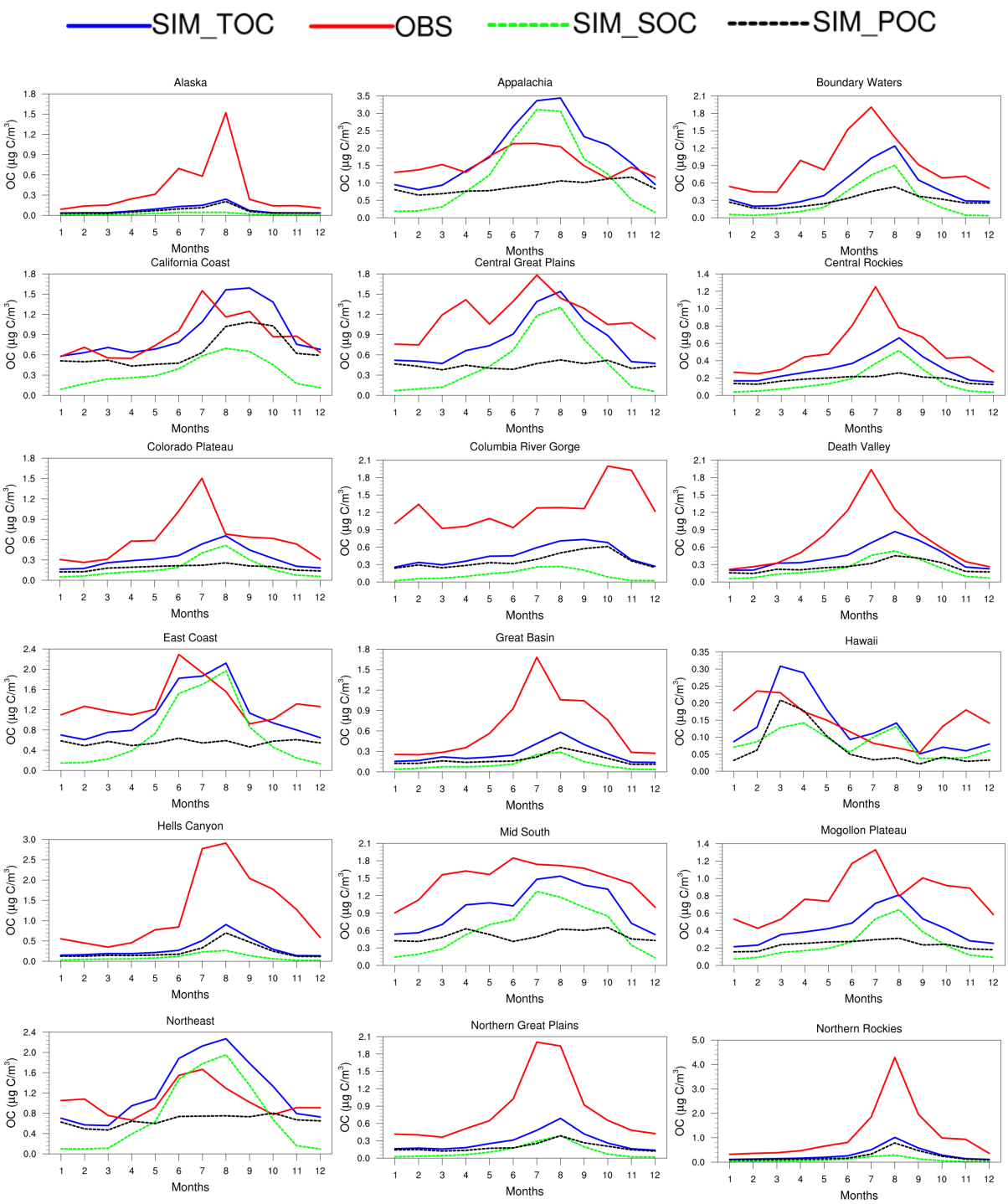
Supplemental section

Text S1. Sensitivity to the formation of SOA from IEPOX

The IEPOX form low-volatility products when it is kinetically uptake by sulfate with the rate that is proportional to the available surface area of these aerosols. However, Gaston et al. (2014) indicated the IEPOX reaction probability may be in proportion to the total particle volume. This sensitivity experiment case was designed to examine the influence of volume-controlled process for IEPOX on the SOA simulation, comparing with the surface-area-controlled process. The number concentration and burden of SOA in this case is shown in following Table S5. Compared with the result shown in Table 4 in the main text, the number concentrations of newSOA in three modes are close to those in the BASE case with change less than 6%. The number concentration of newSOA in nucleation mode decreases by 95×10^{10} m^{-2} in global annual average, while the number concentration of newSOA in Aitken and accumulation mode increase by 251×10^{10} and 0.9×10^{10} m^{-2} . The controlled process for IEPOX have neglectable influence on burden of total burden of SOA as well newSOA with the change of less than 1% from the BASE case. The most significant influence is the distribution of SOA on sulfate, because the low volatility products form from IEPOX are internally mixed with sulfate in the BASE and this sensitivity case. As a result, there are more IEPOX uptake by sulfate in large size with the volume-controlled process than that with surface-area-controlled process. The burden of SOA internally mixed with sulfate in nucleation and Aitken mode decrease by 64.8% and 38.0% in this sensitivity case compared to the BASE case, while the burden of SOA internally mixed with sulfate in accumulation mode increases by 9.8%. However, the total burden of SOA internally mixed with sulfate is changed by 0.5% in the global annual average.

Gaston, C. J., T. P. Riedel, Z. F. Zhang, A. Gold, J. D. Surratt, and J. A. Thornton (2014), Reactive Uptake of an Isoprene-Derived Epoxydiol to Submicron Aerosol Particles, *Environ. Sci. Technol.*, 48(19), 11178-11186, doi:10.1021/es5034266.

Figures


SIM_TOC
OBS
SIM_SOC
SIM_POC
Alaska
Appalachia
Boundary Waters
California Coast
Central Great Plains
Central Rockies
Colorado Plateau
Columbia River Gorge
Death Valley
East Coast
Great Basin
Hawaii
Hells Canyon
Mid South
Mogollon Plateau
Northeast
Northern Great Plains
Northern Rockies
OC (µg C/m3)
Months

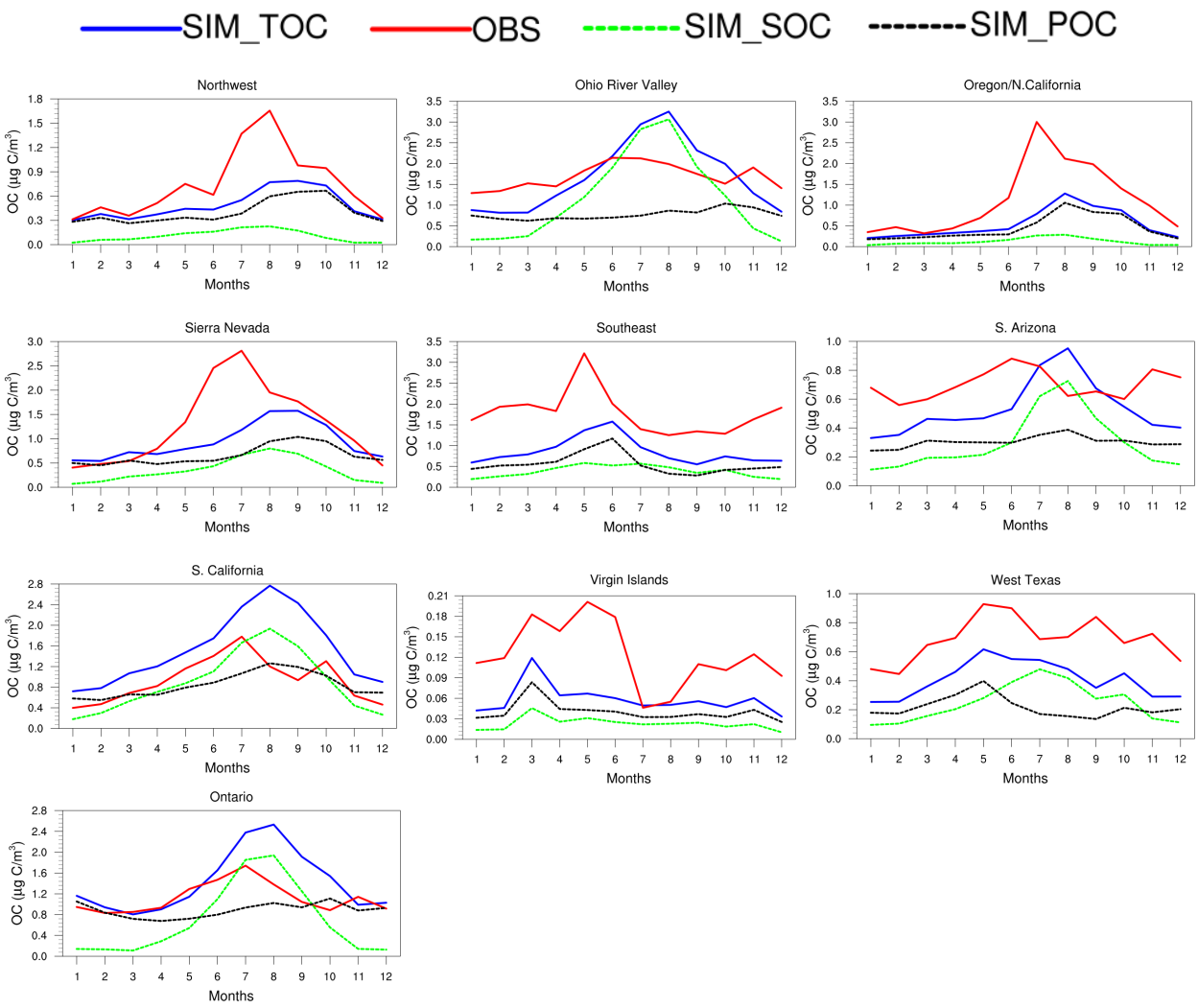
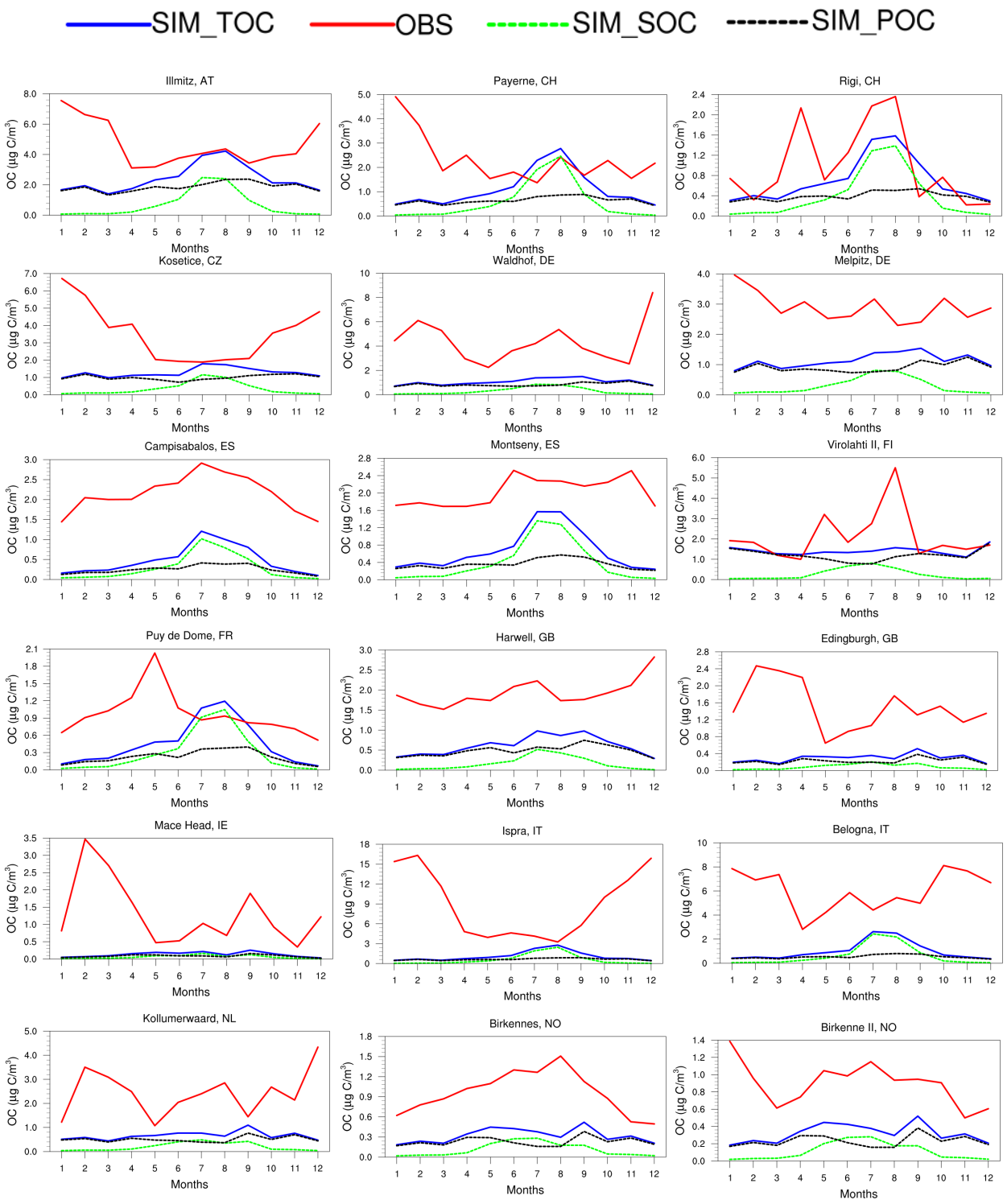


Figure S1. The monthly variation of simulated concentration for the BASE case of total organic carbon (SIM_TOC, blue solid line), secondary organic carbon (SIM_SOC, green dashed line) and primary organic carbon (SIM_POC, black dashed line) as well as the observations (OBS, red solid line) in regions of the IMPROVE network.


SIM_TOC
OBS
SIM_SOC
SIM_POC
Illmitz, AT
Payerne, CH
Rigi, CH
Kosetice, CZ
Waldhof, DE
Melpitz, DE
Campisabalos, ES
Montseny, ES
Virolahti II, FI
Puy de Dome, FR
Harwell, GB
Edingburgh, GB
Mace Head, IE
Ispra, IT
Belogna, IT
Kollumerwaard, NL
Birkennes, NO
Birkenne II, NO
OC (µg C/m3)
Months

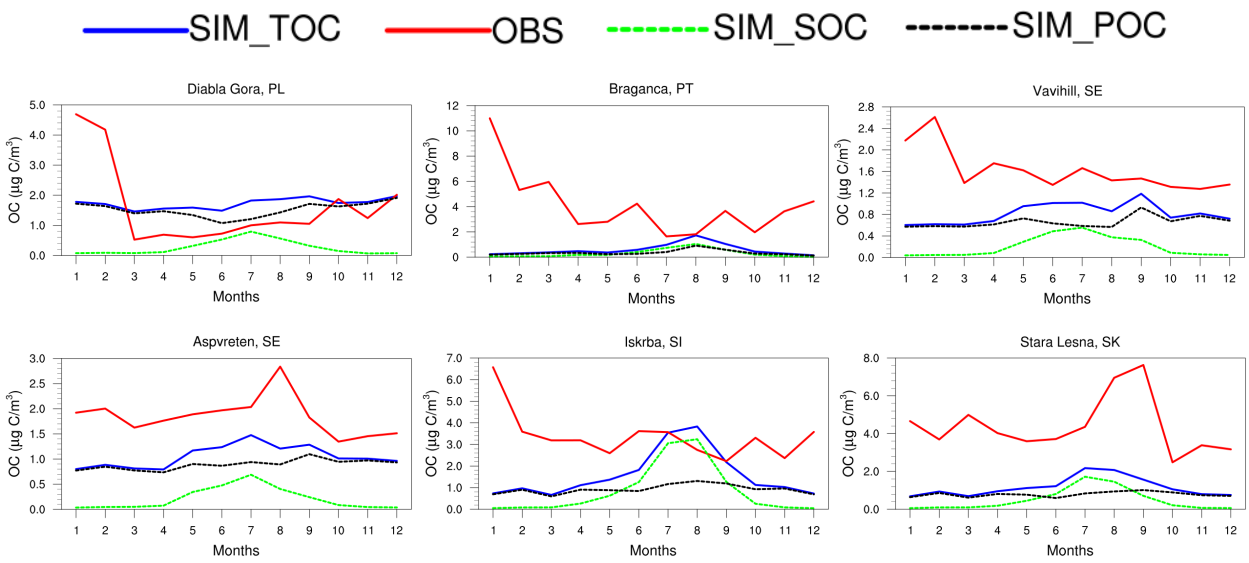
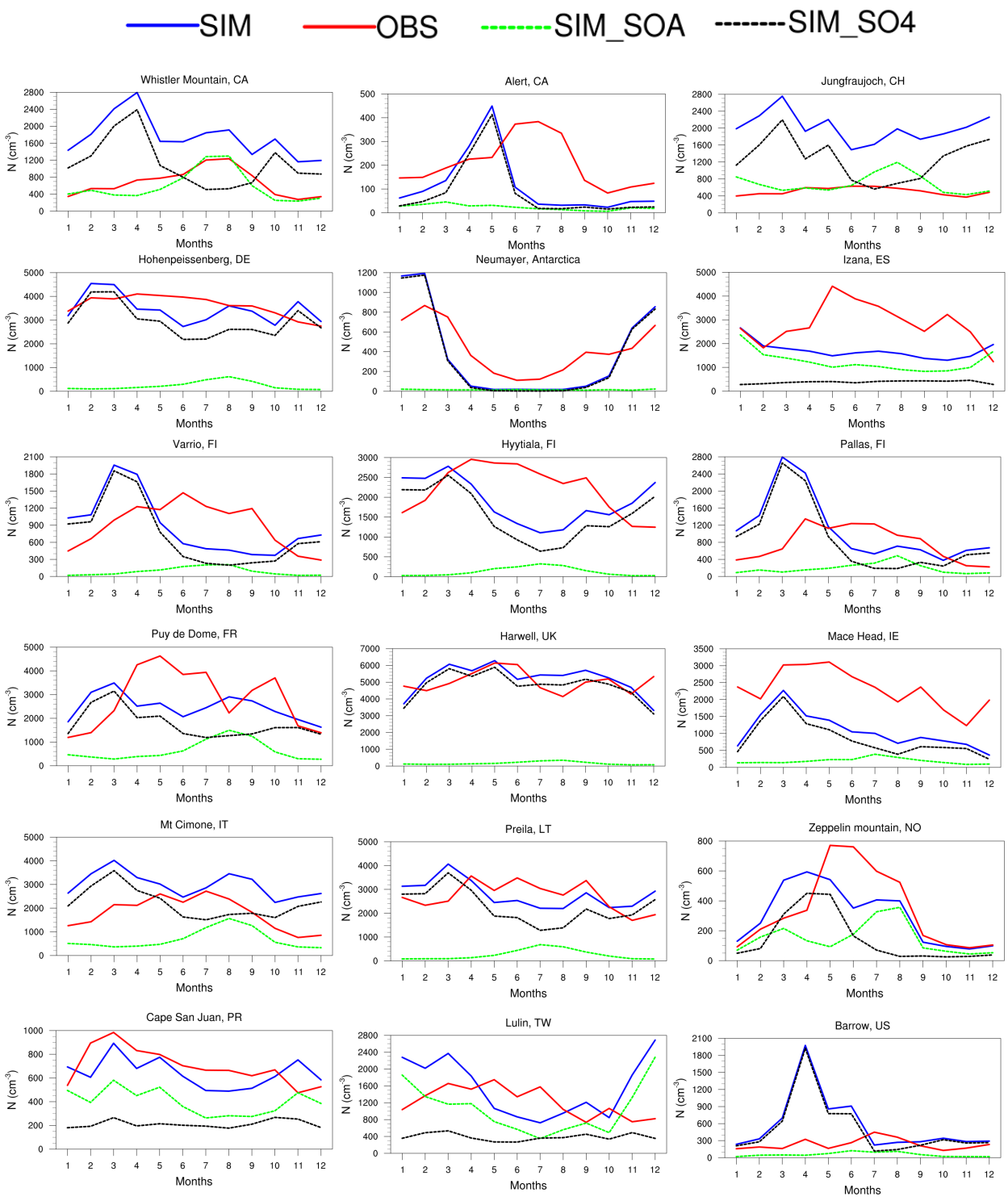


Figure S2. The monthly variation of simulated concentration for the BASE case of total organic carbon (SIM_TOC, blue solid line), secondary organic carbon (SIM_SOC, green dashed line) and primary organic carbon (SIM_POC, black dashed line) as well as the observations (OBS, red solid line) at the EMEP network sites.


SIM
OBS
SIM_SOA
SIM_SO4
Whistler Mountain, CA
Alert, CA
Jungfraujoch, CH
Hohenpeissenberg, DE
Neumayer, Antarctica
Izana, ES
Varrio, FI
Hyytiala, FI
Pallas, FI
Puy de Dome, FR
Harwell, UK
Mace Head, IE
Mt Cimone, IT
Preila, LT
Zeppelin mountain, NO
Cape San Juan, PR
Lulin, TW
Barrow, US
N (cm-3)
Months

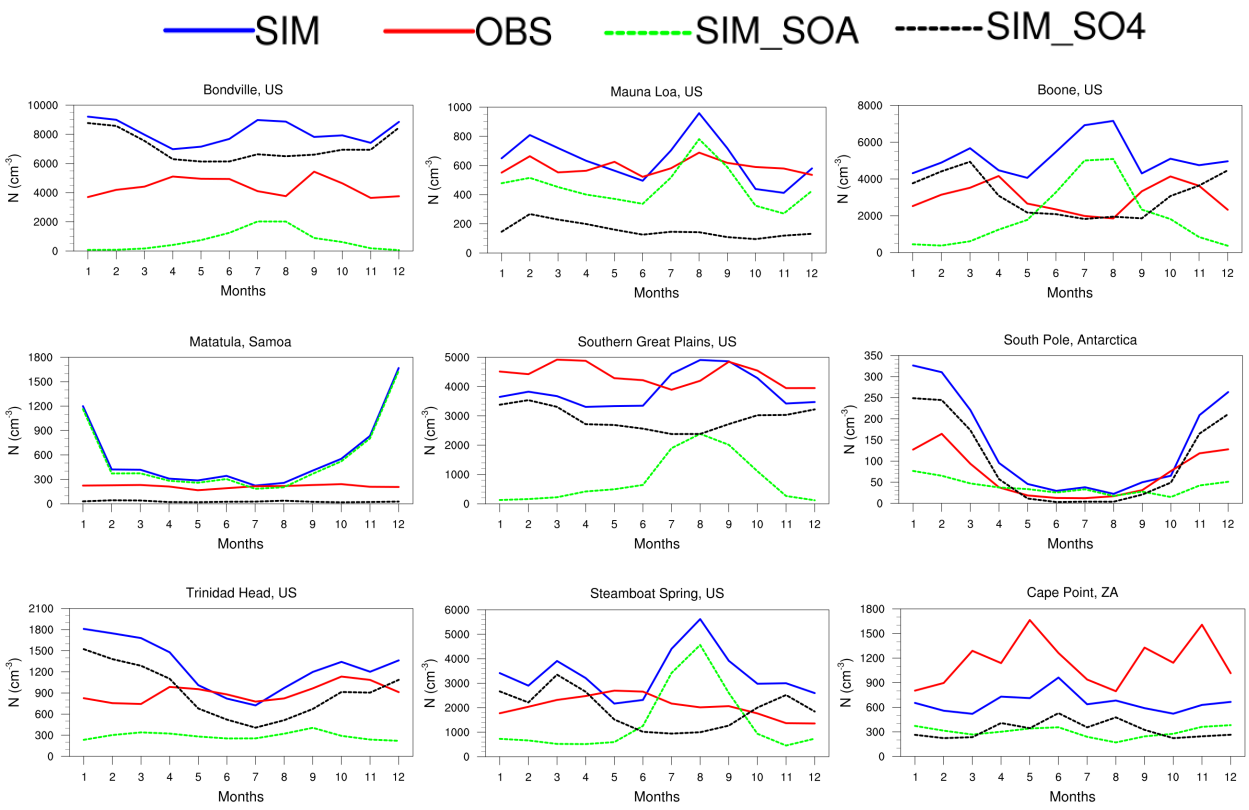


Figure S3. The monthly variation of simulated number concentration for the BASE case of total aerosol (SIM, blue solid line), newSOA (SIM_SOA, green dashed line) and new sulfate particles (SIM_SO4, black dashed line) as well as the observations (OBS, red solid line) for the sites in the EMEP data base (http://ebas.nilu.no).

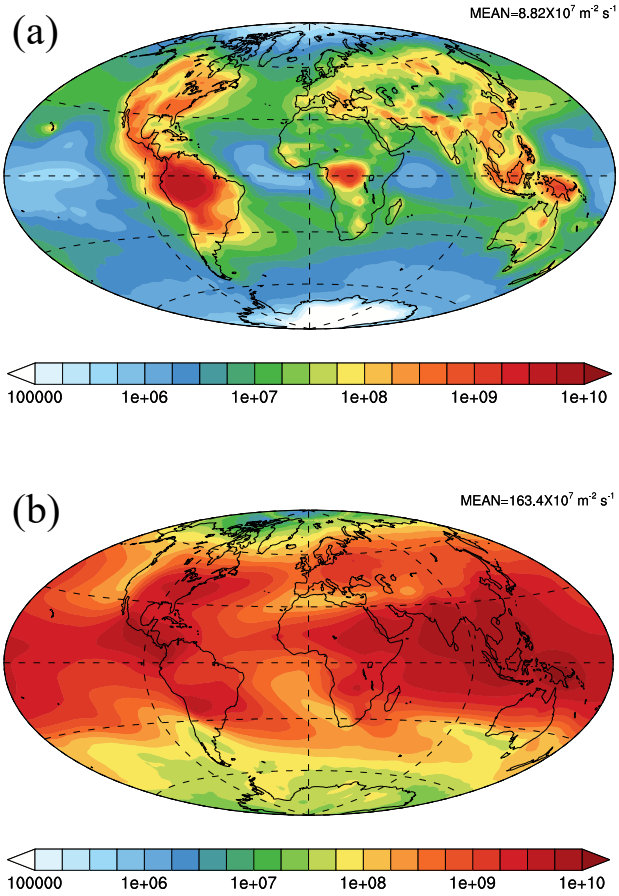


Figure S4. The annual average vertically integrated organic nucleation rate (a) and sulfuric acid nucleation rate (b) for the BASE case.

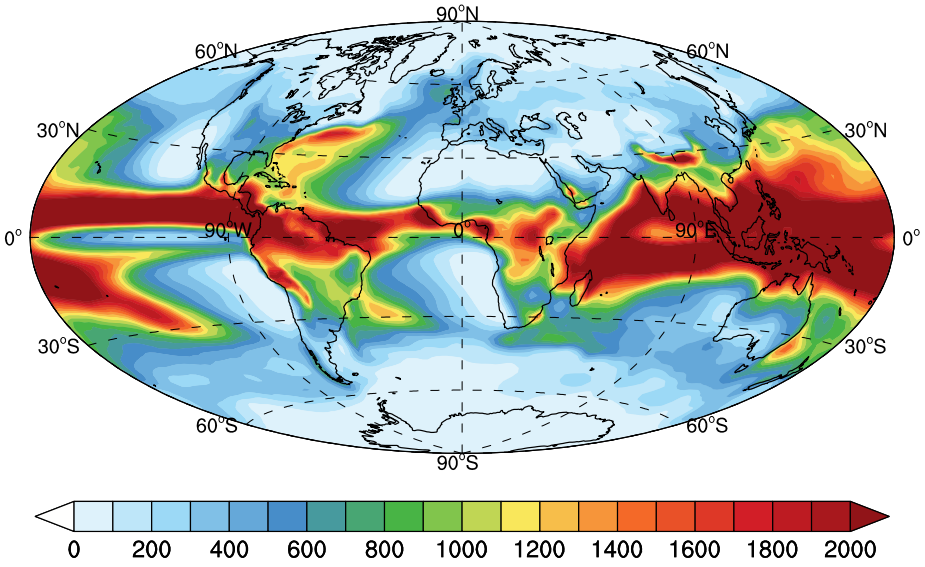


Figure S5. The annual average convective precipitation rate (mm year^{-1}) for the BASE case.

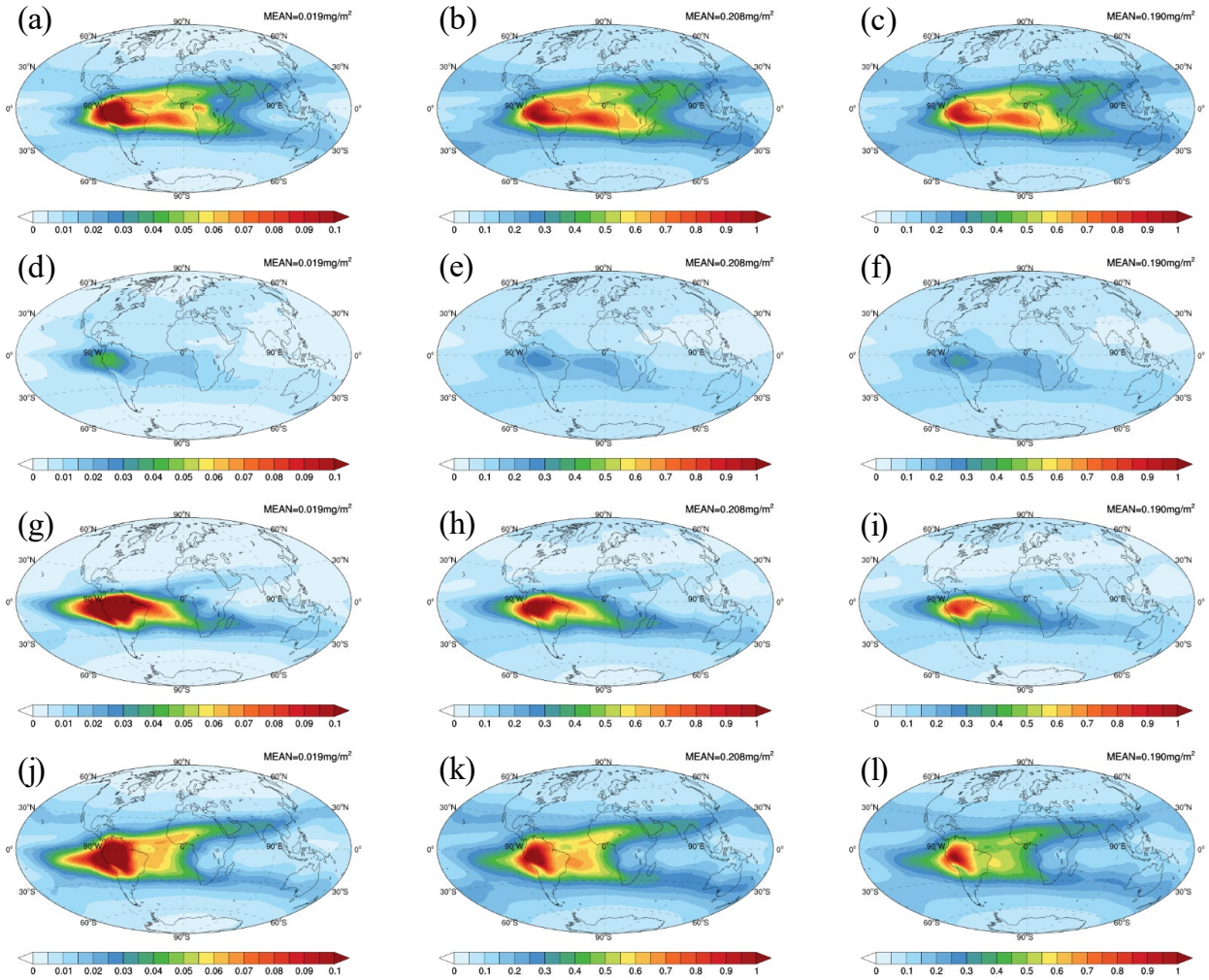
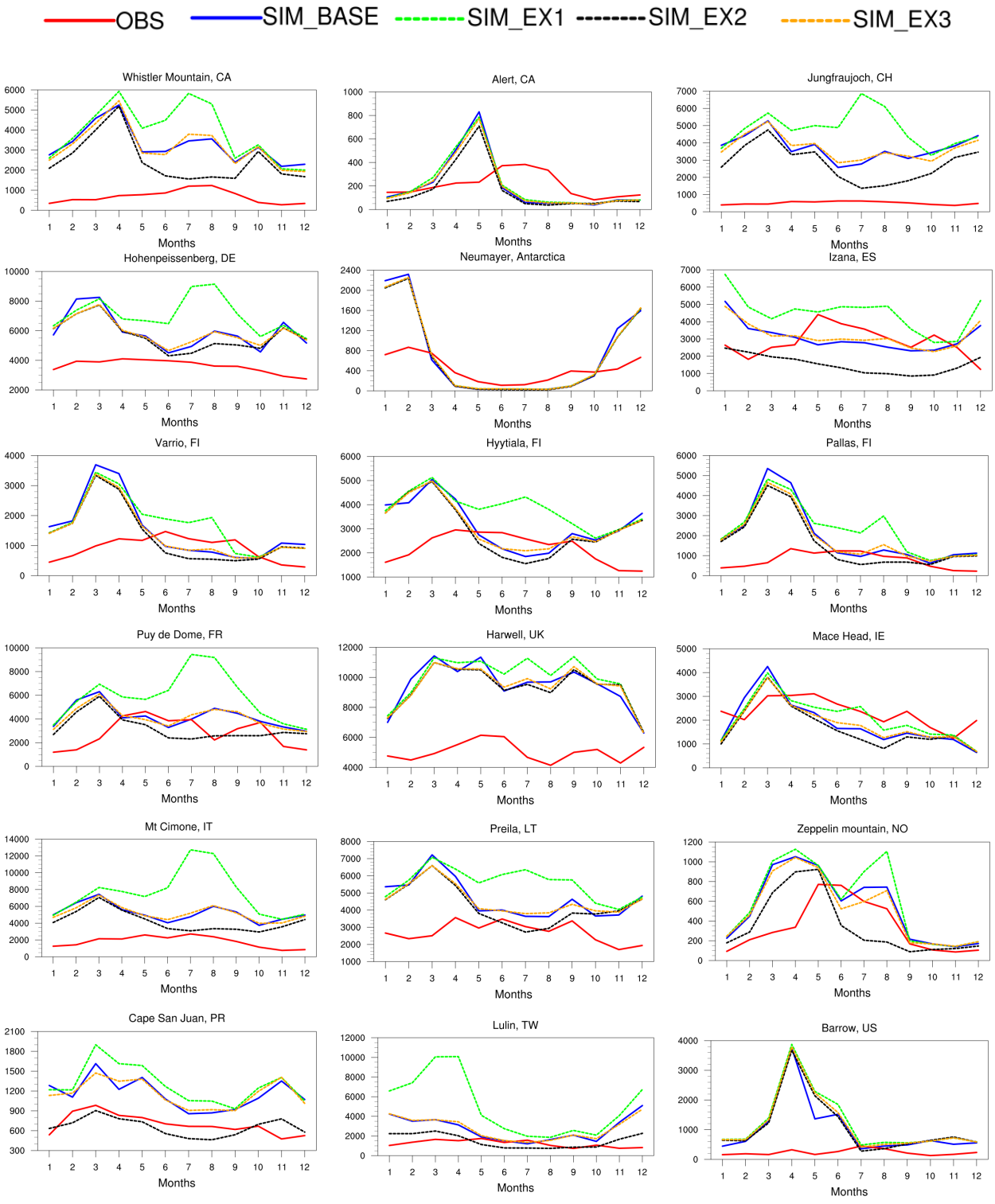


Figure S6. The average mass burden of newSOA in nucleation mode (a, d, g, j), Aitken mode (b, e, h, k) and accumulation mode (c, f, i, l) in the boreal spring (a, b, c), summer (d, e, f), fall (g, h, i) and winter (j, k, l) for BASE case


OBS
SIM_BASE
SIM_EX1
SIM_EX2
SIM_EX3
Whistler Mountain, CA
Alert, CA
Jungfraujoch, CH
Hohenpeissenberg, DE
Neumayer, Antarctica
Izana, ES
Varrio, FI
Hyytiala, FI
Pallas, FI
Puy de Dome, FR
Harwell, UK
Mace Head, IE
Mt Cimone, IT
Preila, LT
Zeppelin mountain, NO
Cape San Juan, PR
Lulin, TW
Barrow, US
Months

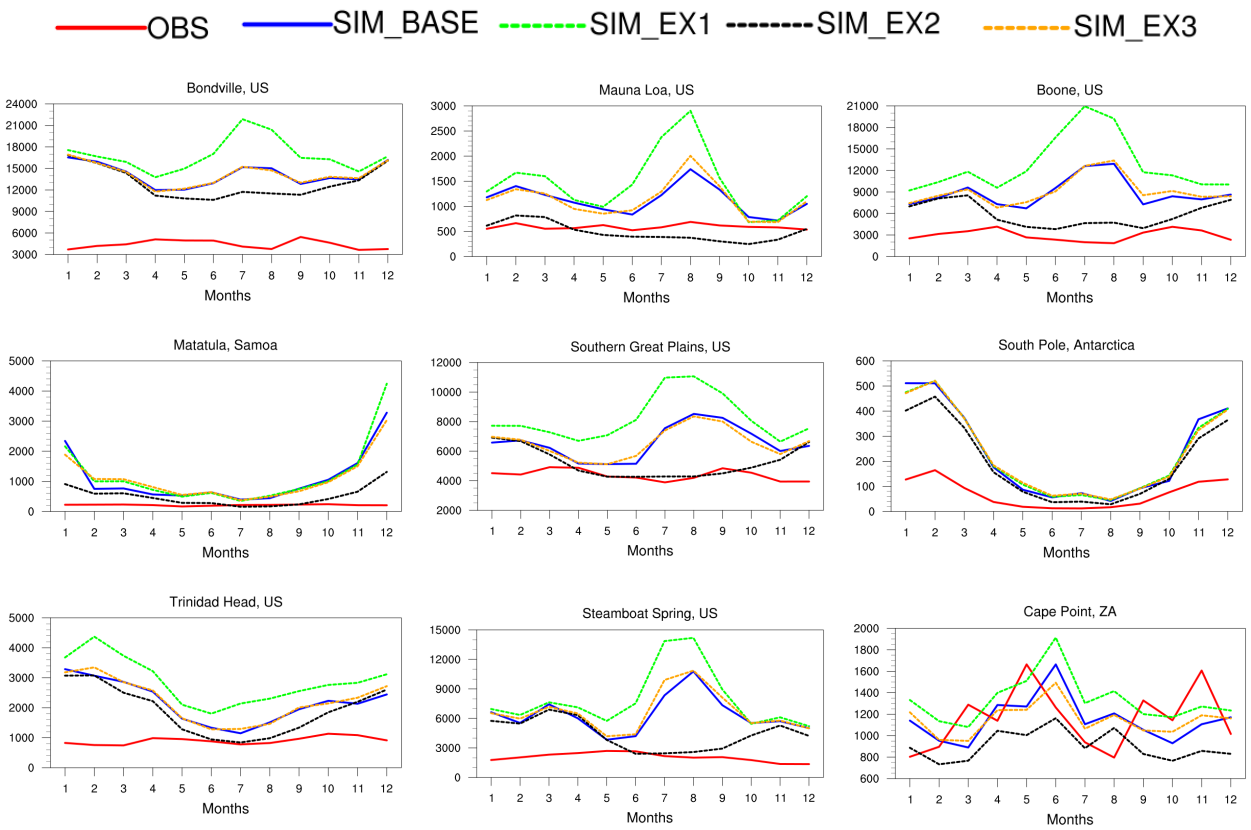


Figure S7 The monthly variation of simulated total aerosol number concentration in the scheme of BASE (SIM_BASE, blue solid line), EX1 (SIM_EX1, green dashed line), EX2 (SIM_EX2, black dashed line) and EX3 (SIM_EX3, yellow dashed line) as well as the observations (OBS, red solid line) at all available sites.

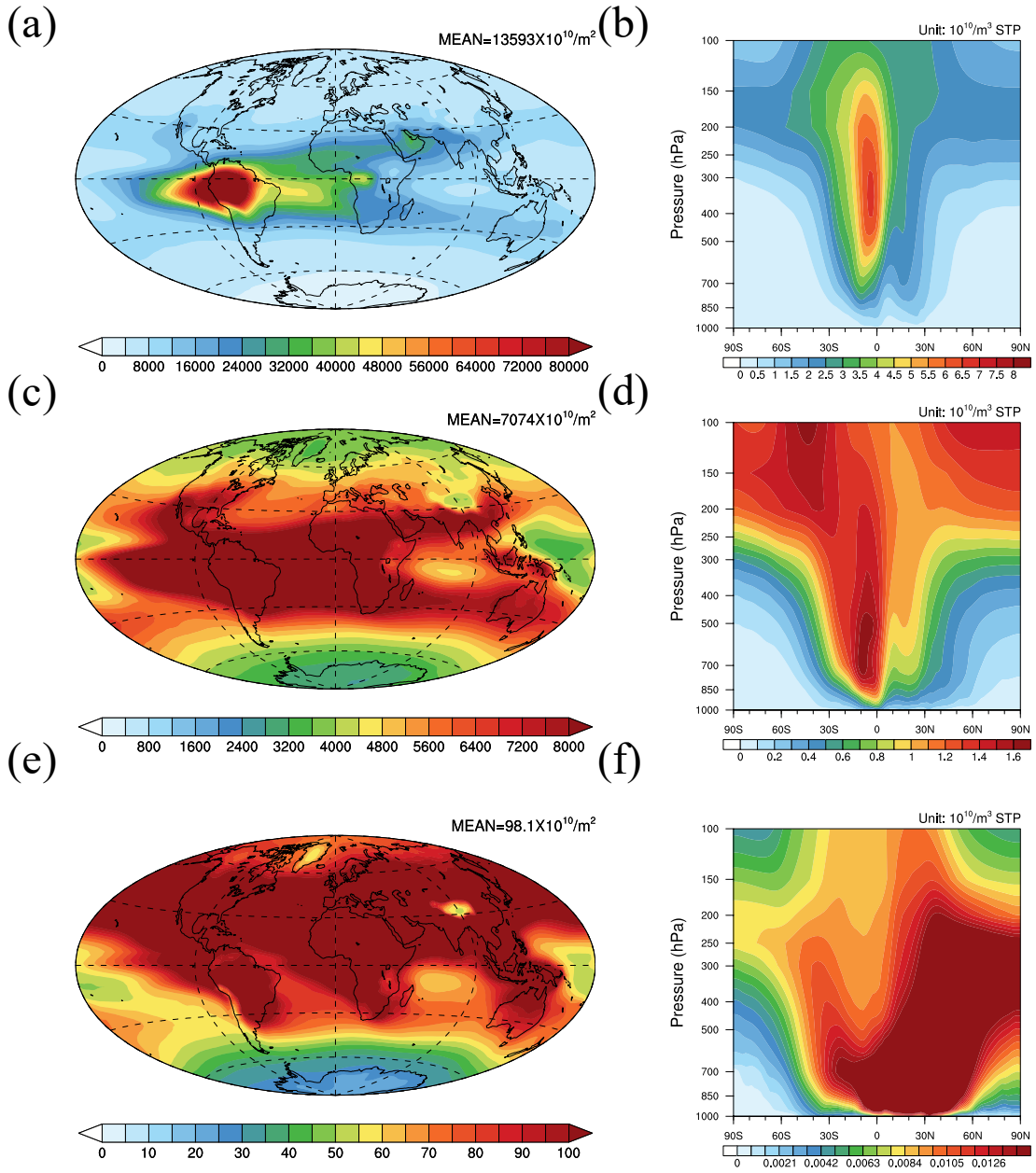


Figure S8 The annual average column number concentration (a,c,e) and zonal average number concentration (b,d,f) of newSOA in the nucleation mode (a,b), Aitken mode (b,d) and accumulation mode (e,f) for EX1 case.

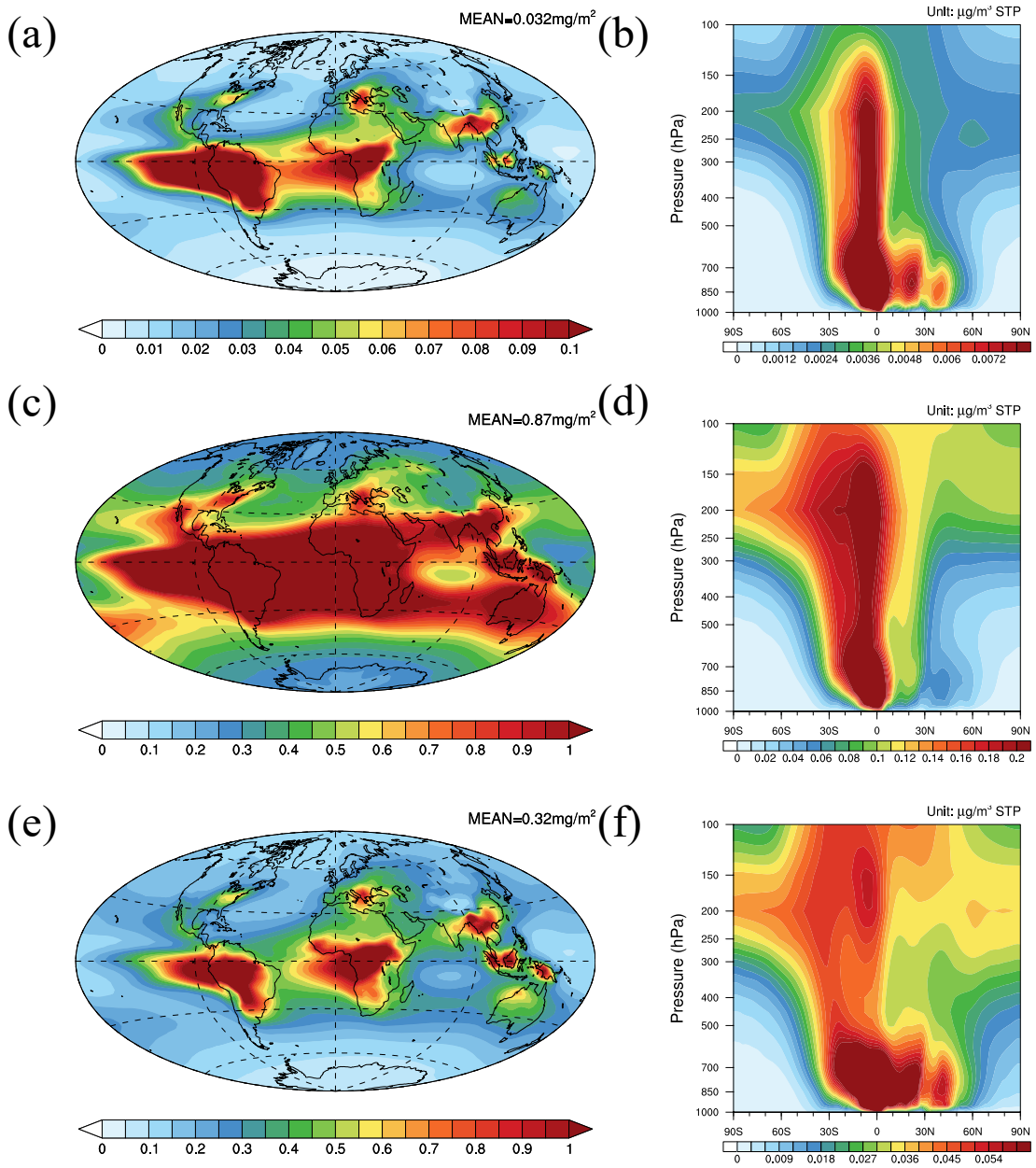


Figure S9 The annual average burden (a,c,e) and zonal average mass concentration (b,d,f) of newSOA in the nucleation mode (a,b), Aitken mode (b,d) and accumulation mode (e,f) for EX1 case.

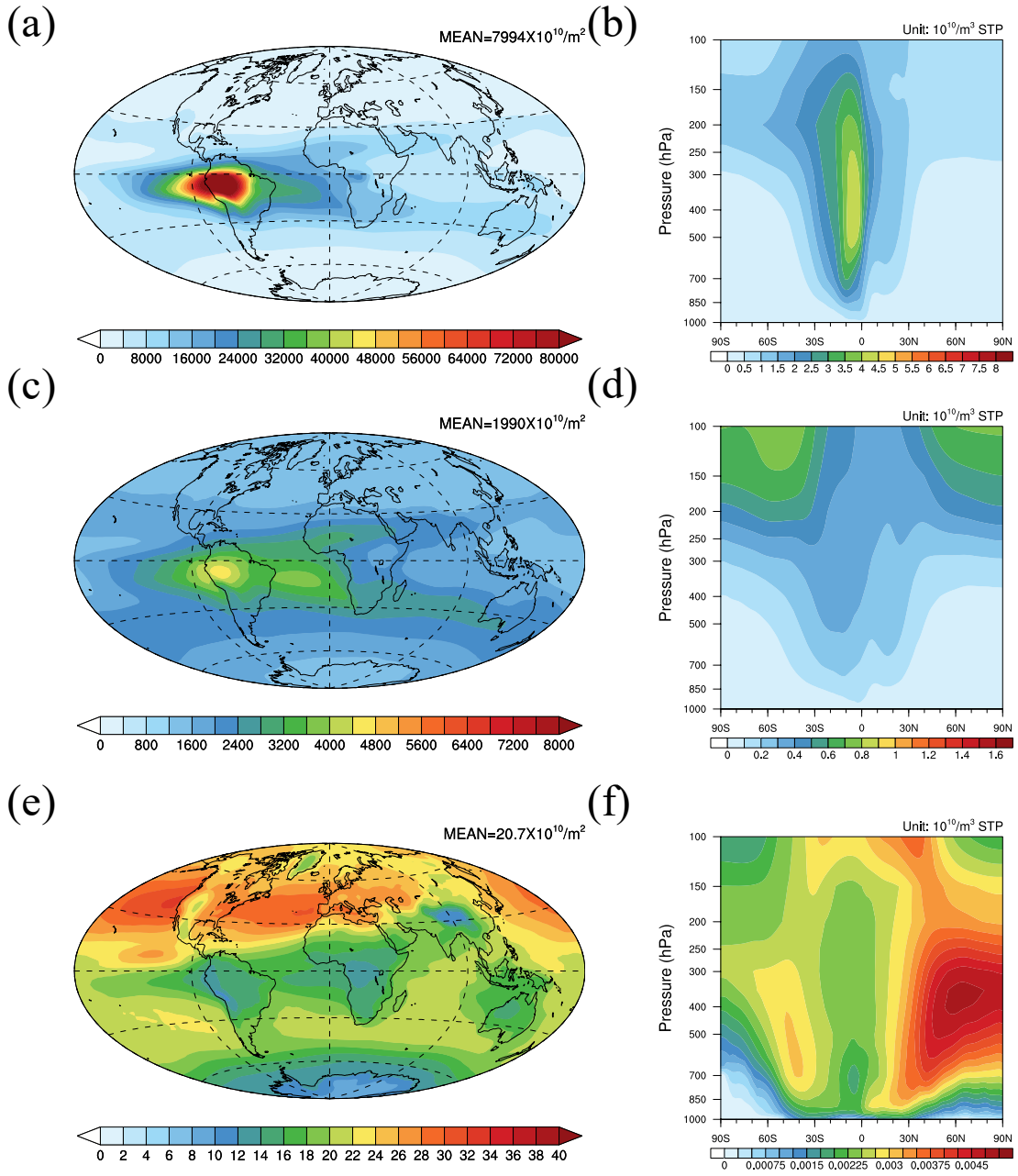


Figure S10. The annual average column number concentration (a,c,e) and zonal averagenumber concentration (b,d,f) of newSOA in the nucleation mode (a,b), Aitken mode (b,d) and accumulation mode (e,f) for EX2 case.

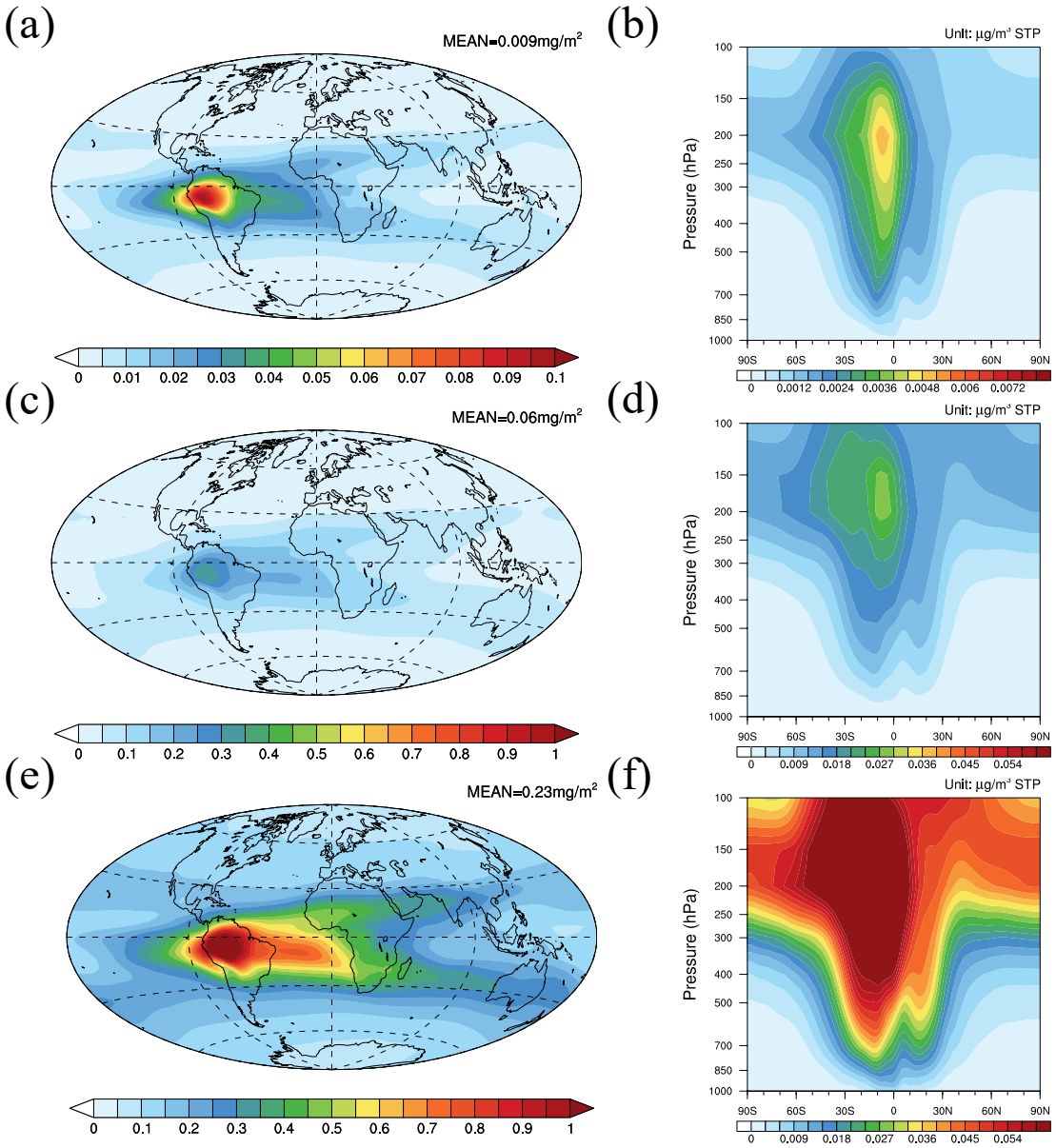


Figure S11 The annual average burden (a,c,e) and zonal average mass concentration (b,d,f) of newSOA in the nucleation mode (a,b), Aitken mode (b,d) and accumulation mode (e,f) for EX2 case.

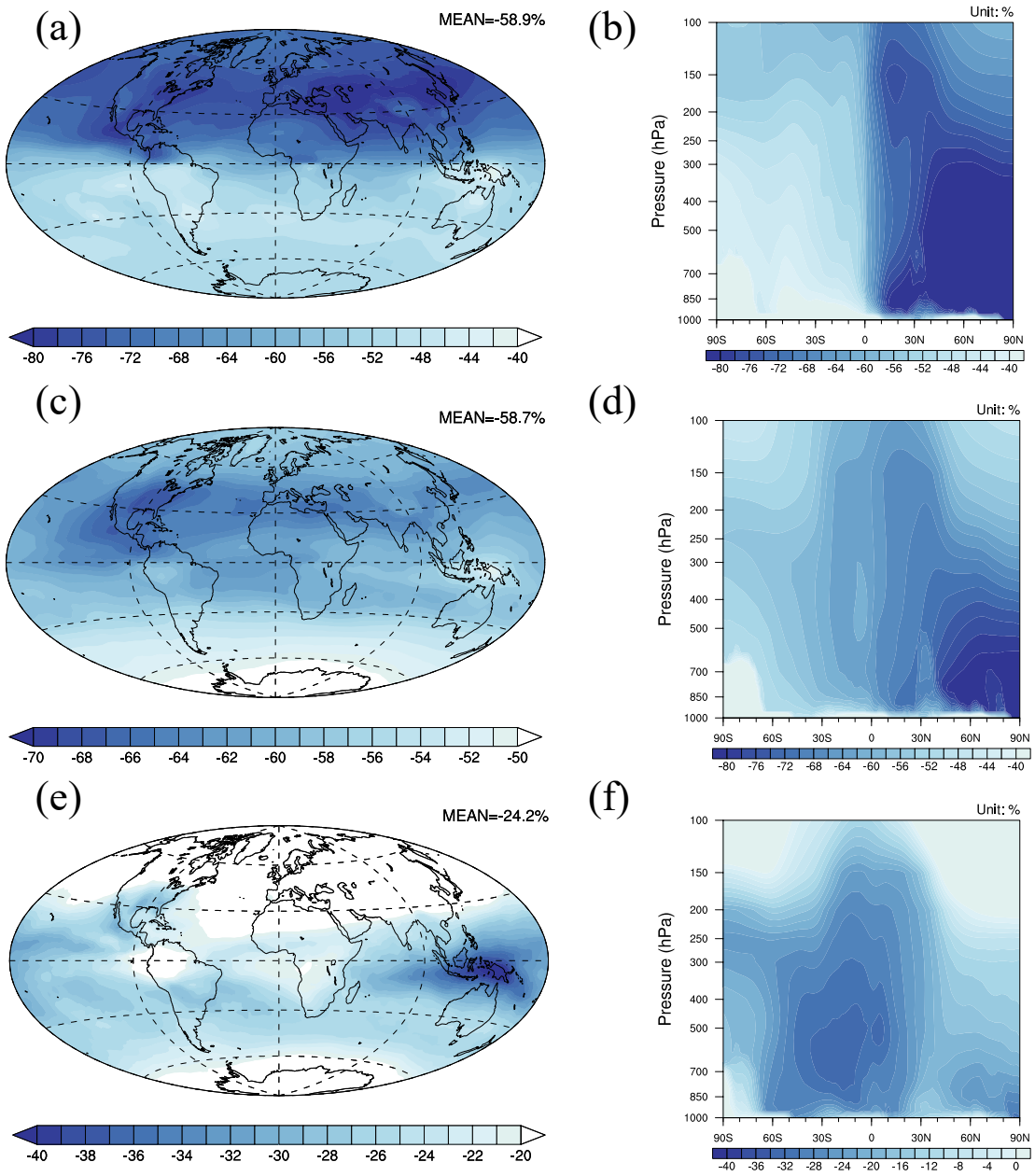


Figure S12 The annual average percentage difference in column number concentration (a,c,e) and zonal average number concentration (b,d,f) of newSOA in the nucleation mode (a,b), Aitken mode (b,d) and accumulation mode (e,f) between EX2 and BASE.

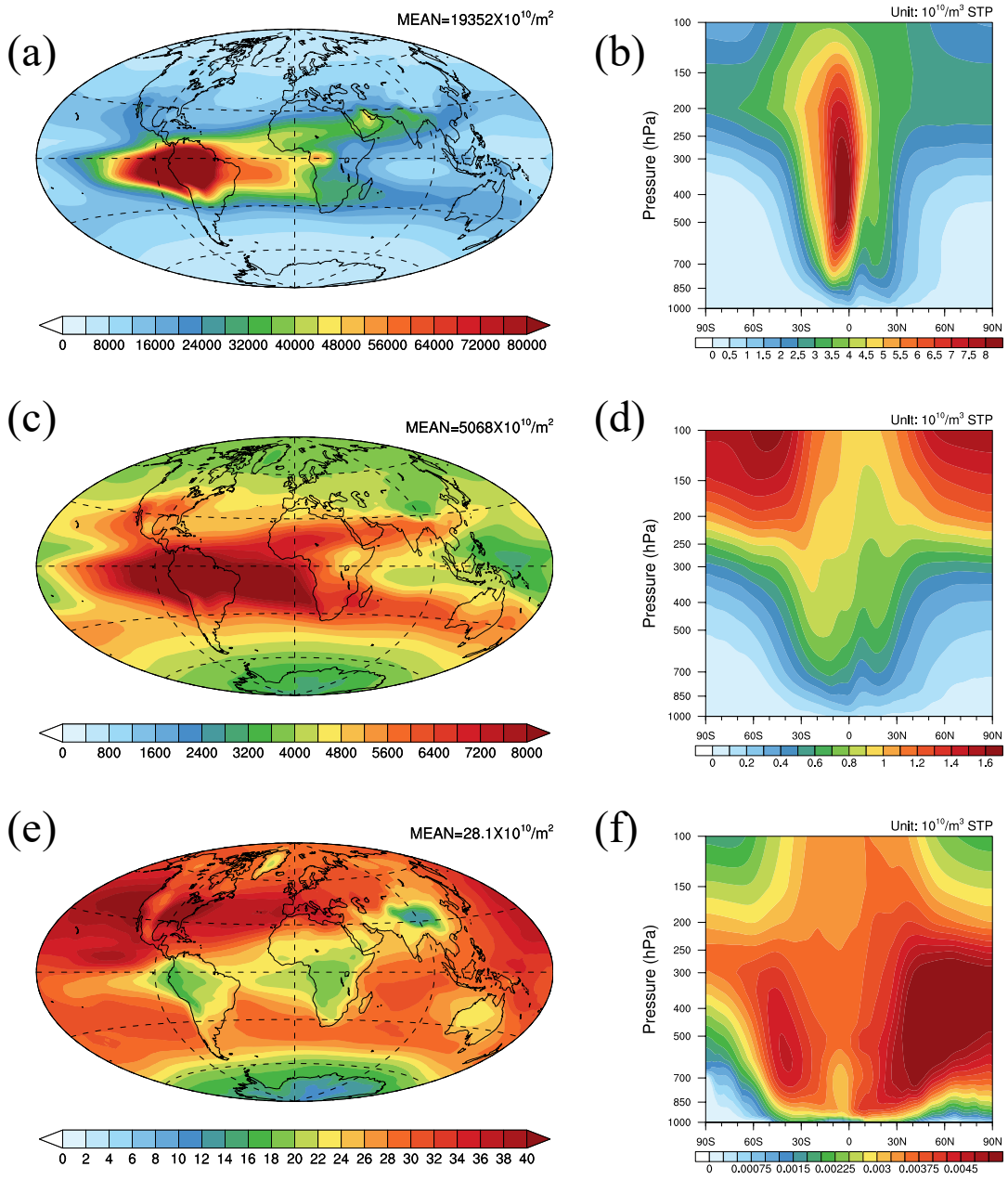


Figure S13. The annual average column number concentration (a,c,e) and zonal average number concentration (b,d,f) of newSOA in the nucleation mode (a,b), Aitken mode (b,d) and accumulation mode (e,f) for EX3 case.

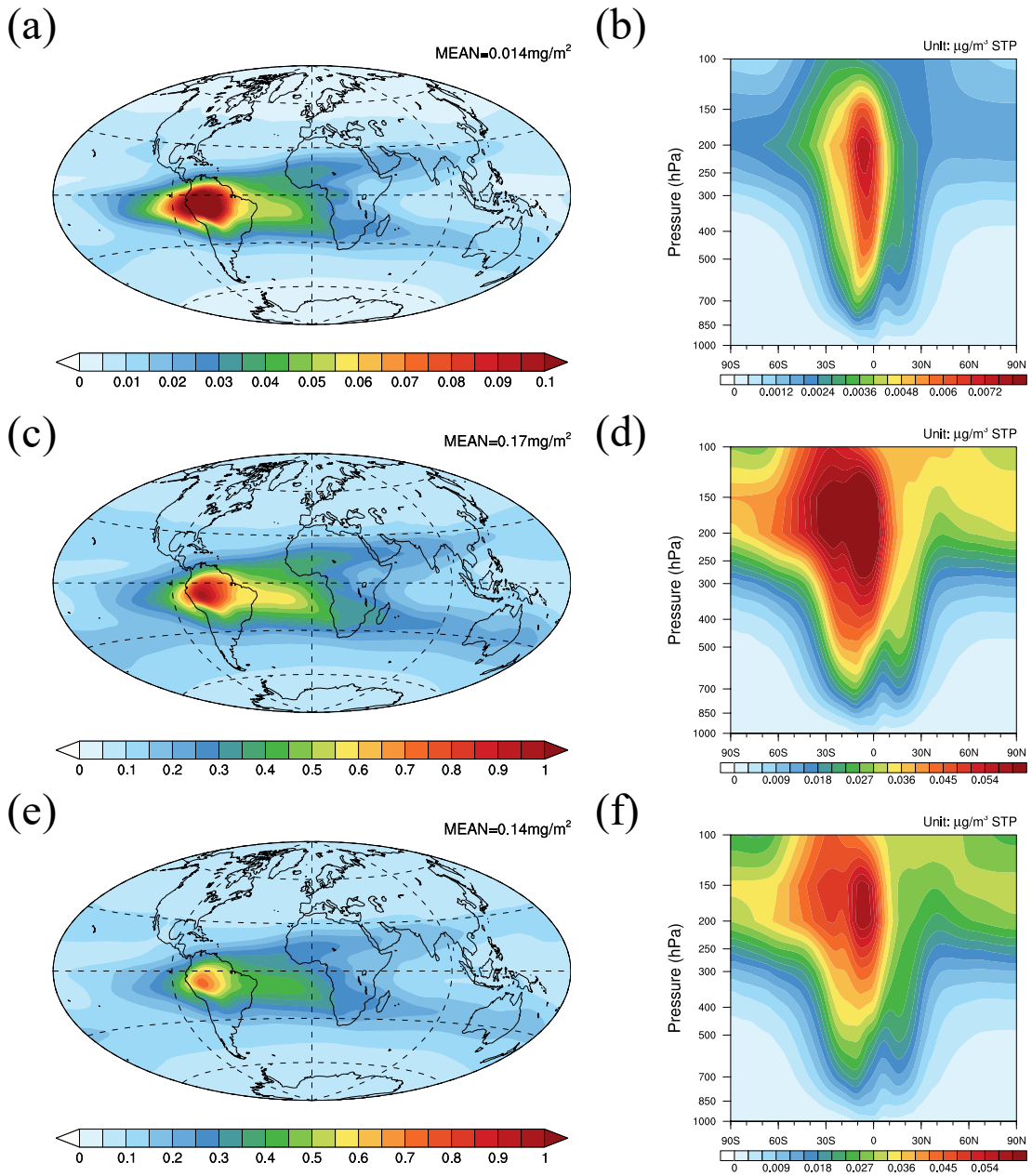


Figure S14. The annual average burden (a,c,e) and zonal average mass concentration (b,d,f) of newSOA in the nucleation mode (a,b), Aitken mode (b,d) and accumulation mode (e,f) for EX3 case.

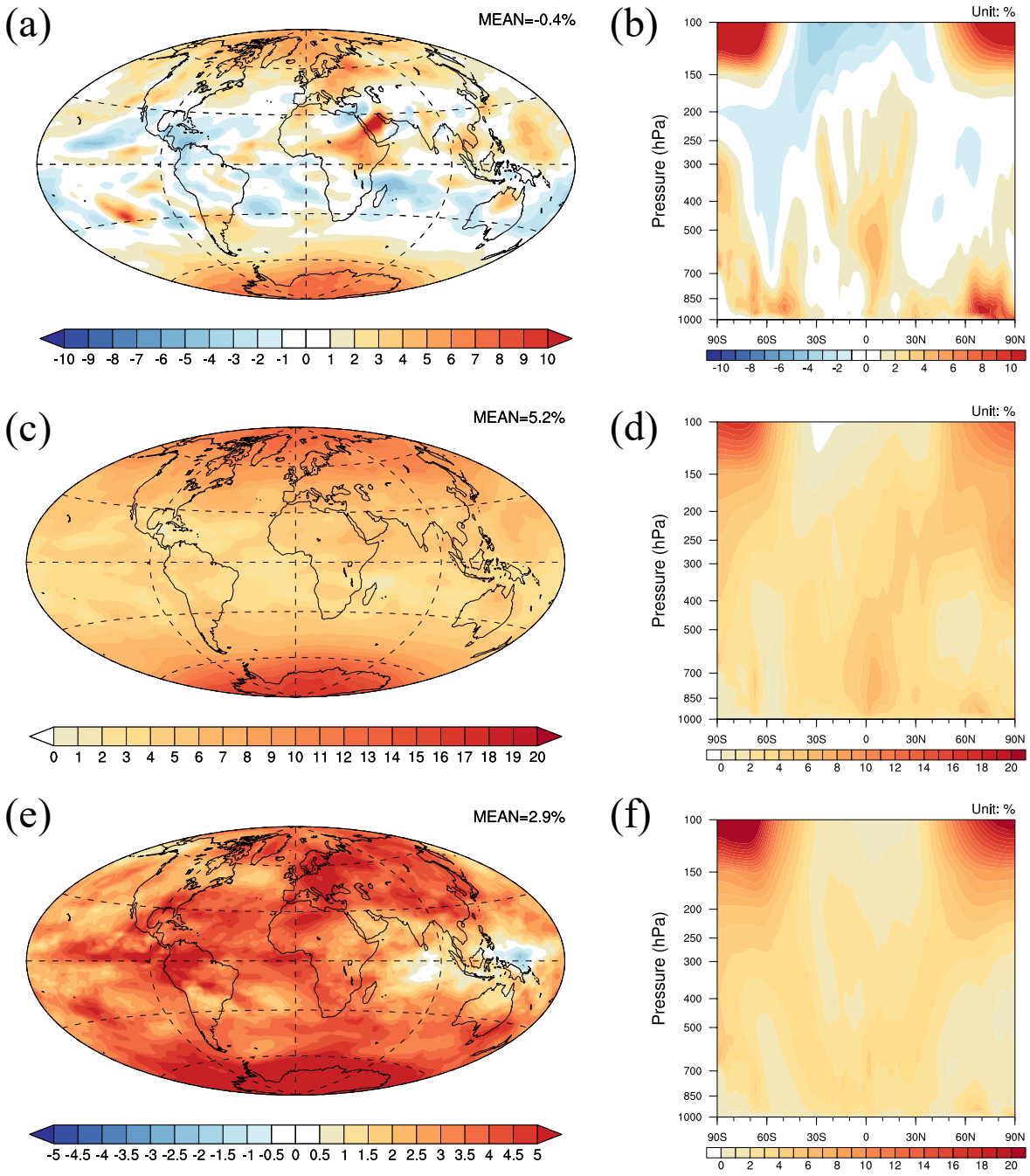


Figure S15 The annual average percentage difference in column number concentration (a,c,e) and zonal average number concentration (b,d,f) of newSOA in the nucleation mode (a,b), Aitken mode (b,d) and accumulation mode (e,f) between EX3 and BASE.

Table

Table S1. The comparison of OC concentration in observations and the BASE case simulation in the regions of the IMPROVE network

NO.	Region	Observation (μg C/m3)	Simulation (μg C/m3)	NMB	R
1	Alaska	0.364	0.090	-75%	0.900
2	Appalachia	1.569	1.910	22%	0.798
3	Boundary Waters	0.907	0.582	-36%	0.913
4	California Coast	0.869	0.833	-4%	0.776
5	Central Great Plains	1.171	0.762	-35%	0.775
6	Central Rockies	0.532	0.347	-35%	0.879
7	Colorado Plateau	0.611	0.375	-39%	0.767
8	Columbia River Gorge	1.269	0.503	-60%	0.452
9	Death Valley	0.715	0.415	-42%	0.866
10	East Coast	1.345	1.019	-24%	0.650
11	Great Basin	0.645	0.297	-54%	0.878
12	Hawaii	0.145	0.154	6%	0.186
13	Hells Canyon	1.232	0.343	-72%	0.884
14	Mid South	1.475	1.013	-31%	0.800
15	Mogollon Plateau	0.807	0.484	-40%	0.730
16	Northeast	1.049	1.231	17%	0.754
17	Northern Great Plains	0.816	0.306	-62%	0.956
18	Northern Rockies	1.115	0.345	-69%	0.977
19	Northwest	0.741	0.522	-30%	0.721
20	Ohio River Valley	1.689	1.610	-5%	0.797
21	Oregon/N.California	1.118	0.596	-47%	0.835
22	Sierra Nevada	1.278	0.834	-35%	0.751
23	Southeast	1.785	0.890	-50%	0.598
24	S. Arizona	0.703	0.567	-19%	0.154
25	S. California	0.939	1.139	21%	0.778
26	Virgin Islands	0.123	0.059	-53%	0.573
27	West Texas	0.687	0.462	-33%	0.647
28	Ontario	1.119	1.396	25%	0.721

NMB: normalized mean bias

R: temporal correlation coefficient between the OC concentration in the simulation and observation

Table S2. The comparison of OC concentration in observations and the BASE case simulation at the sites of the EMEP network

NO.	Region	Observation (μg C/m^3)	Simulation (μg C/m^3)	NMB	R
1	Illmitz	4.690	2.460	-48%	-0.461
2	Payerne	2.318	1.201	-48%	-0.389
3	Rigi	0.997	0.781	-22%	0.690
4	Kosetice	3.563	1.313	-63%	-0.696
5	Waldhof	4.342	1.105	-75%	-0.197
6	Melpitz	2.902	1.200	-59%	-0.340
7	Campisabalos	2.147	0.552	-74%	0.901
8	Montseny	2.030	0.732	-64%	0.601
9	Virolahti II	2.116	1.594	-25%	0.445
10	Puy de Dome	0.965	0.518	-46%	0.092
11	Harwell	1.938	0.678	-65%	-0.120
12	Edingburgh	1.510	0.363	-76%	-0.101
13	Mace Head	1.310	0.161	-88%	-0.268
14	Ispra	9.033	1.201	-87%	-0.691
15	Belogna	6.031	1.064	-82%	-0.418
16	Kollumerwaard	2.440	0.685	-72%	-0.066
17	Birkennes	0.957	0.369	-61%	0.686
18	Birkenne II	0.898	0.369	-59%	0.221
19	Diabla Gora	1.641	1.880	15%	0.012
20	Braganca	4.087	0.615	-85%	-0.459
21	Vavihill	1.617	0.911	-44%	-0.310
22	Aspvreten	1.849	1.191	-36%	0.344
23	Iskrba	3.379	1.747	-48%	-0.267
24	Stará Lesná	4.389	1.221	-72%	0.508

NMB: normalized mean bias

R: temporal correlation coefficient between the OC concentration in the simulation and observation

Table S3. The bias and temporal correlation coefficient between the aerosol number concentration in the BASE case simulation and observations at all available sites over the world

NO.	Site name	w/o organic nucleation		w/ organic nucleation		
		NMB	R	NMB	R	R_SOA
1	Whistler Mountain	12%	0.717	56%	0.941	0.826
2	Alert	-73%	-0.150	-65%	0.025	0.209
3	Jungfraujoch	18%	0.248	129%	0.315	0.637
4	Hohenpeissenberg	-1%	-0.047	2%	0.087	0.050
5	Neumayer	-90%	0.870	-85%	0.943	0.756
6	Izana	-82%	0.410	-47%	-0.546	0.527
7	Varrio	-44%	-0.669	-39%	-0.510	-0.345
8	Hyytiala	-16%	-0.936	-14%	-0.805	0.013
9	Pallas	-48%	-0.705	-35%	-0.129	-0.531
10	Puy de Dome	-55%	0.786	-43%	0.590	0.554
11	Harwell	68%	-0.138	72%	-0.037	-0.119
12	Mace Head	-63%	-0.169	-57%	0.213	0.026
13	Mt Cimone	-7%	0.706	13%	0.854	-0.240
14	Preila	-2%	-0.135	-4%	-0.112	0.040
15	Zeppelin mountain	-78%	-0.592	-48%	0.291	0.628
16	Cape San Juan	-31%	0.091	10%	0.094	0.614
17	Lulin	-69%	0.280	9%	-0.102	-0.061
18	Barrow	149%	-0.510	150%	-0.183	-0.226
19	Bondville	171%	-0.620	167%	-0.669	-0.128
20	Mauna Loa	-63%	-0.325	5%	0.316	-0.007
21	Boone	21%	0.414	37%	0.359	-0.210
22	Matatula	-74%	0.346	170%	0.342	0.722
23	Southern Great Plains	-10%	0.153	-3%	0.033	-0.320
24	South Pole	65%	0.835	151%	0.913	0.748
25	Trinidad Head	25%	0.064	50%	0.544	-0.175
26	Steamboat Spring	-38%	0.628	-17%	0.466	-0.527
27	Cape Point	-73%	0.126	-43%	0.073	-0.145

NMB: normalized mean bias

R: temporal correlation coefficient between the total aerosol number concentration in the simulation and observation

R_SOA: temporal correlation coefficient between the newSOA number concentration in the simulation and the total aerosol number concentration in the observation

Table S4. The bias and temporal correlation coefficient between the aerosol number concentration in different sensitivity experiments and observations at all available sites over the world

NO.	Site name	EX1		EX2		EX3	
		NMB	R	NMB	R	NMB	R
1	Whistler Mountain	123%	0.957	10%	0.893	47%	0.970
2	Alert	-66%	0.065	-73%	-0.097	-67%	0.044
3	Jungfraujoch	245%	0.723	51%	0.326	126%	0.371
4	Hohenpeissenberg	15%	0.116	0%	-0.303	2%	-0.222
5	Neumayer	-85%	0.960	-86%	0.953	-84%	0.953
6	Izana	-11%	-0.316	-73%	-0.139	-46%	-0.527
7	Varrio	-26%	0.451	-44%	-0.522	-41%	-0.350
8	Hyytiala	1%	-0.430	-14%	-0.788	-12%	-0.800
9	Pallas	-12%	0.637	-45%	-0.557	-35%	-0.122
10	Puy de Dome	-18%	0.489	-53%	0.672	-45%	0.610
11	Harwell	63%	-0.012	59%	-0.039	60%	-0.030
12	Mace Head	-57%	0.439	-65%	0.357	-61%	0.406
13	Mt Cimone	68%	0.802	-4%	0.895	11%	0.883
14	Preila	9%	0.770	-6%	0.363	-3%	0.544
15	Zeppelin mountain	-42%	0.597	-73%	0.000	-49%	0.619
16	Cape San Juan	25%	0.555	-24%	0.363	11%	0.368
17	Lulin	114%	0.202	-40%	0.195	13%	-0.075
18	Barrow	169%	-0.115	148%	-0.261	161%	-0.160
19	Bondville	195%	-0.318	158%	-0.652	161%	-0.652
20	Mauna Loa	49%	0.466	-43%	0.031	6%	0.287
21	Boone	126%	-0.629	26%	0.462	38%	0.329
22	Matatula	291%	0.087	13%	0.196	191%	0.169
23	Southern Great Plains	19%	-0.165	-7%	0.065	-4%	0.062
24	South Pole	153%	0.952	120%	0.970	163%	0.954
25	Trinidad Head	82%	-0.346	29%	-0.113	44%	-0.118
26	Steamboat Spring	52%	0.289	-34%	0.681	-17%	0.561
27	Cape Point	-35%	0.205	-64%	0.083	-44%	0.173

NMB: normalized mean bias

R: temporal correlation coefficient between the total aerosol number concentration in the simulation and observation

Table S5. Summary of the boreal seasonal and annual global average SOA number and burden in the sensitivity to the formation of SOA from IEPOX

		Spring	Summer	Fall	Winter	Annual
Aerosol Number (10^{10} m^{-2})	newSOA (nucleation)	21958	13182	20197	22038	19343
	newSOA (Aitken)	5122	4219	5153	5770	5066
	newSOA (accumulation)	27.5	30.3	27.4	27.4	28.2
Aerosol Burden (mg m^{-2})	newSOA (nucleation)	0.017	0.007	0.013	0.018	0.014
	newSOA (Aitken)	0.217	0.094	0.141	0.227	0.170
	newSOA (accumulation)	0.186	0.087	0.104	0.173	0.137
	mixSOA with sulfate	0.955	1.196	1.073	0.833	1.014
	mixSOA with soot (fossil/bio-fuel)	0.287	0.340	0.316	0.220	0.291
	mixSOA with soot (biomass burning)	0.159	0.333	0.405	0.181	0.269
	mixSOA with sea salt and dust	0.007	0.004	0.003	0.004	0.005
	Total SOA	1.83	2.06	2.06	1.66	1.90