Geoscientific
Model Development

Supplement of

Impacts of spatial heterogeneity of anthropogenic aerosol emissions in a regionally refined global aerosol–climate model

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Aerosol	Emission space	Mean [$\times 10^{-12}$ kg/m^2/s] (accurate)	NMB [%]	StdDev [$\times 10^{-12}$ kg/m^2/s] (accurate)	NStdDevB	RMSE [$\times 10^{-12}$ kg/m^2/s]	N_RMSE [%]
BC	surface	4.93	-2.669/-1.847	9.5	-0.209/0.083	4.29/3.6	45.1/37.8
	elevated	1.53	-1.01/-25.053	10.5	-0.102/-0.137	3.58/6.52	34/61.9
POM	surface	17.5	-3.099/-0.472	38.9	-0.204/0.1	16.6/13.2	42.7/33.9
	elevated	39.1	-0.37/-26.966	301	-0.1/-0.146	102/186	33.7/61.8
SO4	surface	0.67	-0.138/-0.903	1.16	-0.140/0.087	0.42/0.3	36.1/26
	elevated	4.91	-2.194/-0.174	15.3	-0.276/0.525	8.9/11.3	57.9/73.6

Table S2. EAMv2 anthropogenic aerosol emissions data statistics in the default emission treatment for present-day (PD) RRM simulations. Statistics are shown for both the surface and elevated emissions of different aerosol species. All estimates are over the South Asian land surface (bounded by $0° - 30°$N and $60°$E $- 120°$E). Mean values indicate the area-weighted mean emission fluxes. NMB, NStdDevB, and N_RMSE are defined as $\left(\frac{\sum(emis_{lin}-emis_{accurate})}{\sum emis_{accurate}}\right) \times 100\%$, $\frac{stdDev_{lin}-stdDev_{accurate}}{stdDev_{accurate}}$, $\frac{RMSE}{stdDev_{accurate}} \times 100\%$ respectively. The subscript "accurate" indicates data that preserve spatial heterogeneity and conserve mass. The subscript "lin" indicates linearly interpolated data used in the default treatment. NMB, NStdDevB, RMSE, and N_RMSE before (after) the slash are estimates for RRM-PD (RRM-EHR-PD). Units of Mean, StdDev, and RMSE are in kg/m^2/s. N_RMSE and NMB are in percentage (%). NStdDevB is unitless.

Aerosol	Emission space	Mean [$\times10^{-12}$ kg/m^2/s] (accurate)	NMB [%]	StdDev [$\times10^{-12}$ kg/m^2/s] (accurate)	NStdDevB	RMSE [$\times10^{-12}$ kg/m^2/s]	N_RMSE [%]
BC	surface	76.3	-3.091/ 0.734	84.6	-0.236/ 0.192	42.6/ 30.3	50.3/ 35.7
	elevated	4.1	-2.566/ 4.997	9.49	-0.244/ 0.340	4.48/ 4.89	47.1/ 51.4
POM	surface	286	-3.088/ 0.769	278	-0.202/ 0.151	120/ 85.8	43.3/ 30.9
	elevated	59.6	-2.414/ 2.032	177	-0.285/ 0.306	94.4/ 86	53.4/ 48.6
SO4	surface	5.56	-2.927/ 0.671	7.38	-0.223/ 0.168	3.62/ 2.27	49/ 30.7
	elevated	32.4	-4.259/ 0.023	64.8	-0.381/ 0.598	43.8/ 57.5	67.6/ 88.7

Table S3. EAMv2 simulated source-sink statistics from RRM-SE-PD and RRM-PD simulations. Statistics are based on annual mean estimates for both the total and contributing processes for different aerosol species. All estimates are over the North America land surface. N_RMSE is defined as in Table 1.

| Aerosol species	Sources / Sinks	Decomposed process	Mean [kg m^{-2} s^{-1}]	RMSE [kg m^{-2} s^{-1}]	N_RMSE [%]
BC	Sources	Total	0.00002	0.000046	71
		Surf emis	0.000015	0.000023	67
		Elev emis	0.000006	0.00004	72
	Sinks	Total	-0.000017	0.000006	45
		Dry dep (Grav)	0.0	0.0	15
		Dry dep (Turb)	0.000007	0.000006	58
		Wet dep (incloud, strat)	-0.000007	0.0	13
		Wet dep (incloud, conv)	-0.000003	0.0	12
		Wet dep (belowcloud)	-0.0	0.0	13
POM	Sources	Total	0.000197	0.001136	72
		Surf emis	0.000052	0.000084	62
		Elev emis	0.000144	0.001133	72
	Sinks	Total	-0.000131	0.000029	34
		Dry dep (Grav)	0.000003	0.0	17
		Dry dep (Turb)	0.000048	0.000028	45
		Wet dep (incloud, strat)	-0.000056	0.000005	13
		Wet dep (incloud, conv)	-0.000023	0.000003	19
		Wet dep (belowcloud)	-0.000001	0.0	18
Sulfate	Sources	Total	0.0001	0.000037	36
		Surf emis	0.0	0.0	54
		Elev emis	0.000005	0.000014	85
		Gas-aero exchange	0.000023	0.00001	32
		AQ chem (H2SO4)	0.000001	0.000001	45
		AQ chem (SO4)	0.00007	0.000017	25
	Sinks	Total	-0.000103	0.000006	9
		Dry dep (Grav)	0.000004	0.000001	20
		Dry dep (Turb)	0.000015	0.000001	11
		Wet dep (incloud, strat)	-0.000067	0.000005	11
		Wet dep (incloud, conv)	-0.000017	0.000002	10
		Wet dep (belowcloud)	-0.000001	0.0	11
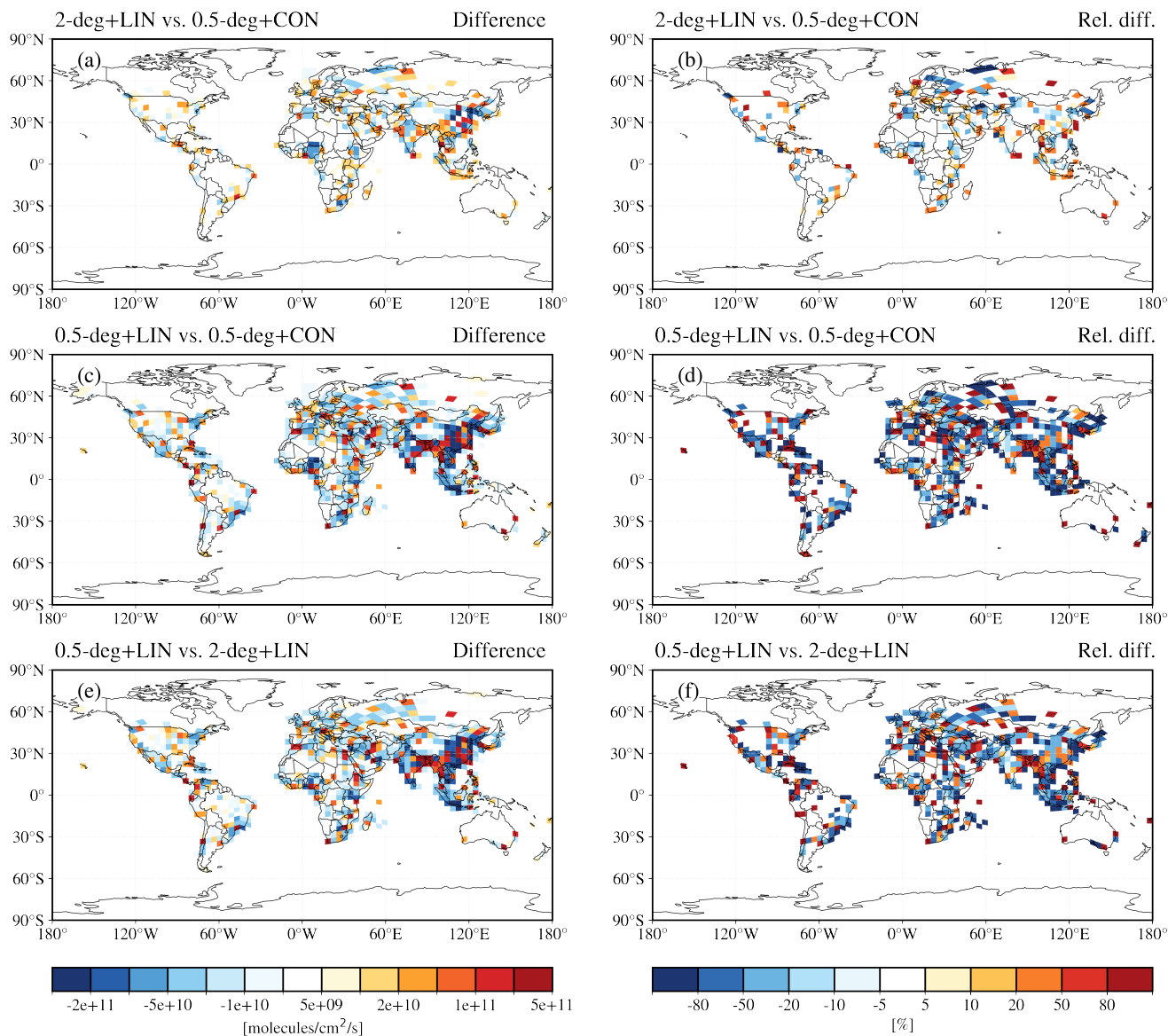
Figure S1. Spatial distributions of surface Black Carbon (BC) emission differences among different remapping configurations. Three remapping configurations are exploited, including the conservative remapping of the high-resolution (~0.5°) emission data onto the EAM ~4° physical grids (named "0.5-deg+CON"), the non-conservative linear remapping of the 0.5° emission data onto the ~4° grids (named "0.5-deg+LIN"), and the non-conservative linear remapping of the low-resolution (~2°) emission data onto the ~4° grids (named "2-deg+LIN"). The first row (a, b) shows the differences between the "2-deg+LIN" and "0.5-deg+CON" remapping configurations, the second row (c, d) is for the differences between the "0.5-deg+LIN" and "0.5-deg+CON" remapping configurations, and the third row (e, f) compares the "0.5-deg+LIN" and "2-deg+LIN" configurations. The emission differences (a, c, e) are shown in the left panels in molecules/cm2/s and the relative differences (b, d, f) are shown in the right panels in percent (%).
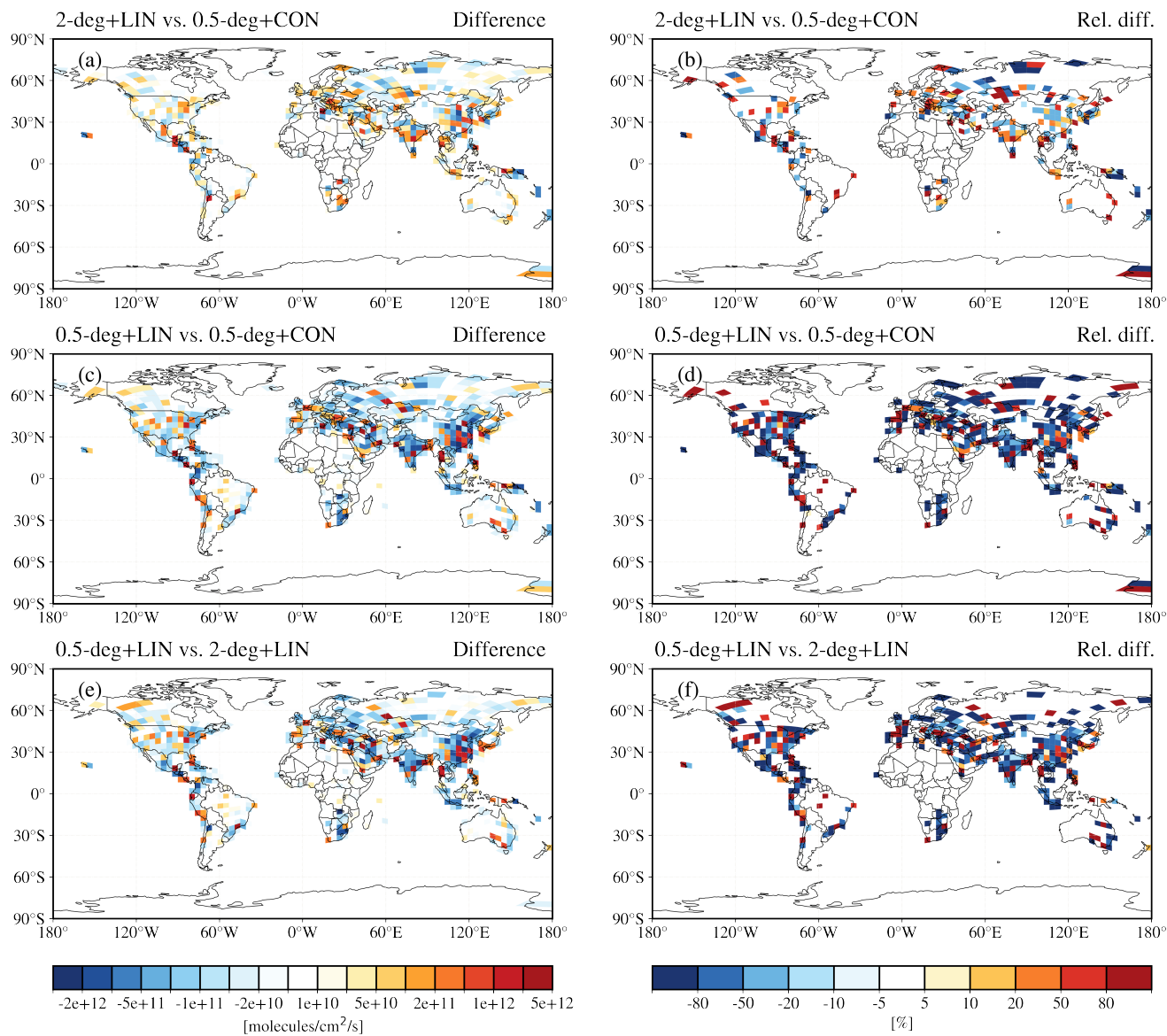
Figure S2. Same as Figure S2 but for elevated SO2 emissions (i.e., energy, industrial, biomass burning, and volcanic sources).
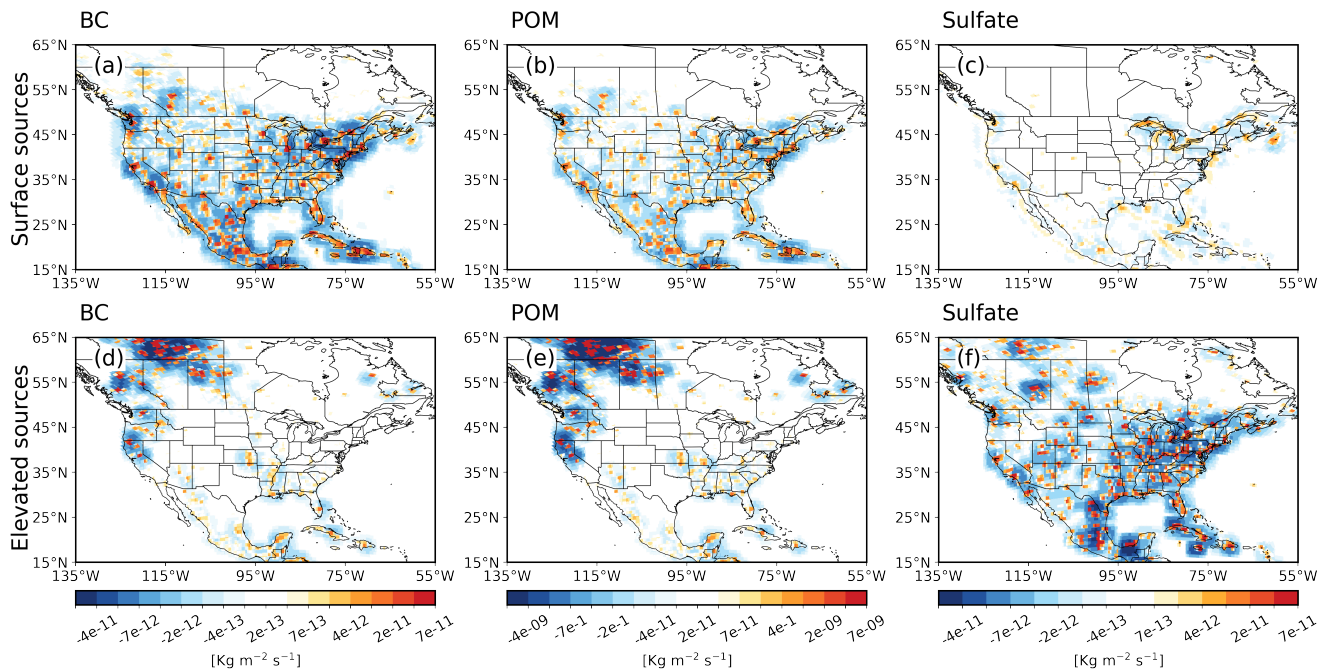
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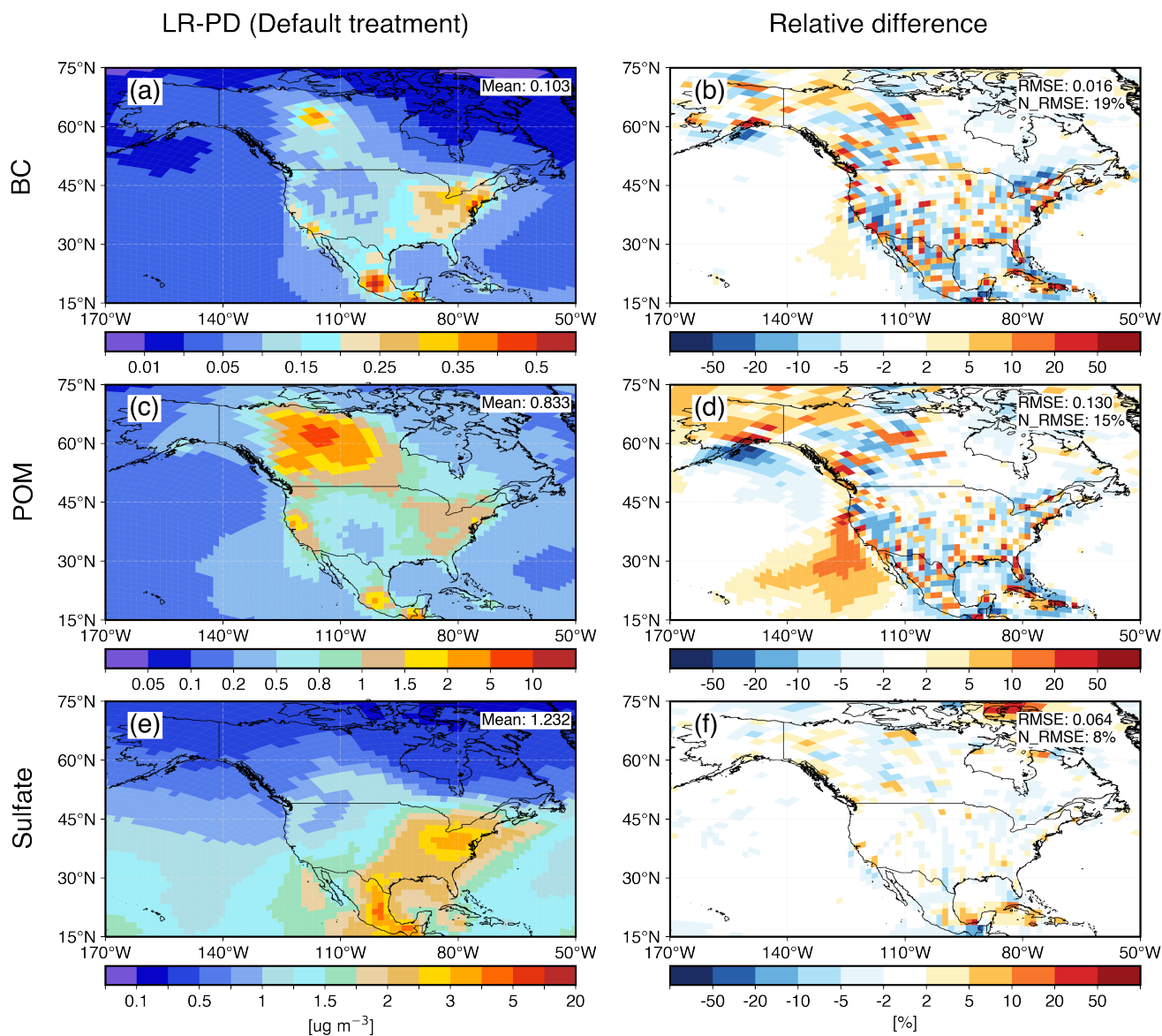
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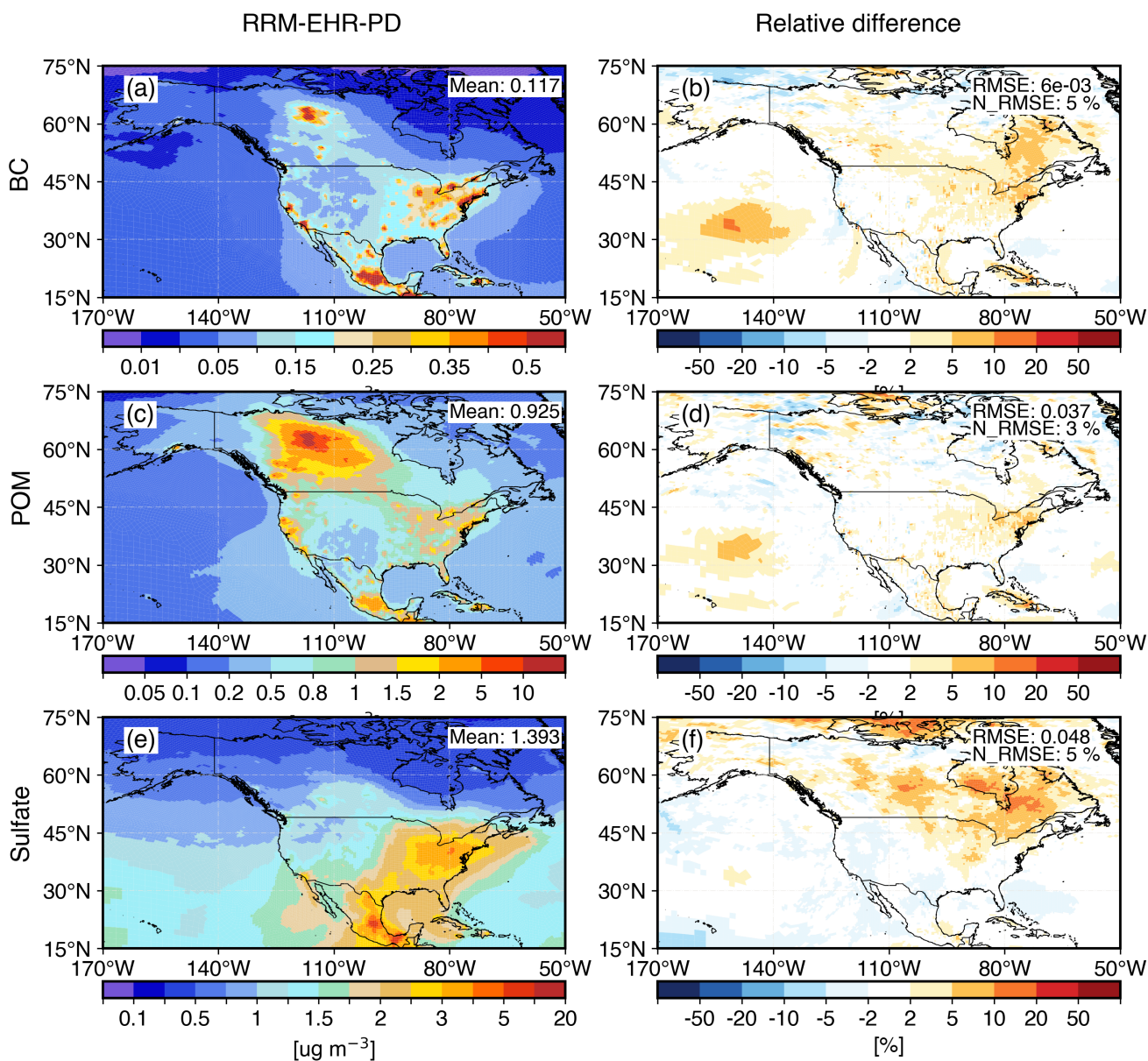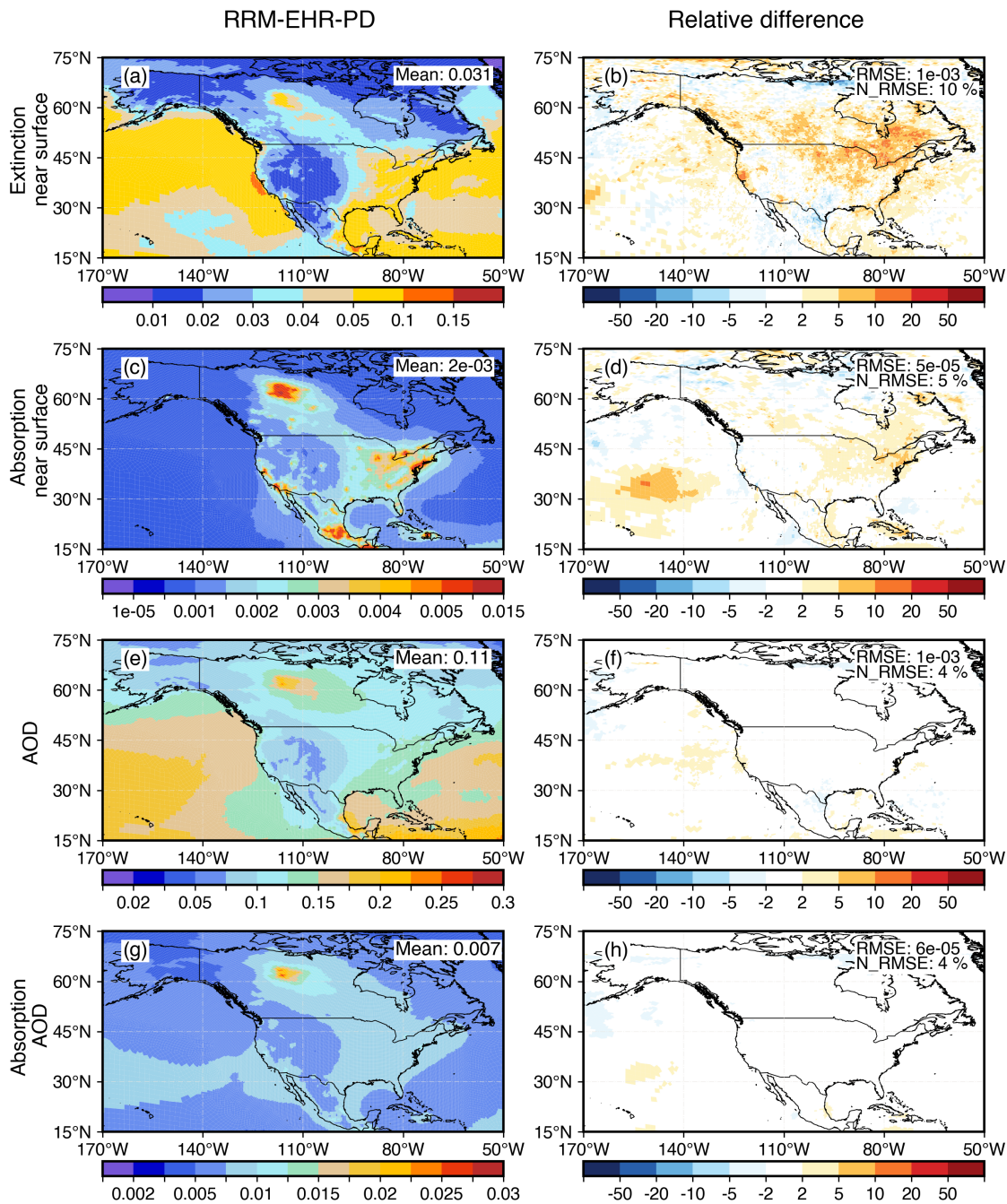
Figure S5. Simulated spatial distribution of annual mean aerosol surface concentration from RRM-EHR-PD (left column) and the relative difference between RRM-SE-PD and RRM-EHR-PD (right column) over North America. Distributions are shown for (a, b) Black Carbon (BC), (c, d) Primary Organic Matter (POM), and (e, f) Sulfate aerosols. The relative difference for field X is calculated as: $\left(\frac{X_{def}-X_{se}}{X_{se}}\right) \times$ 100%, where "se" and "def" subscripts refer to the simulations with new and default emission treatment respectively. Mean, RMSE and normalized RMSE (N_RMSE) are indicated at the top right corner of the panels. Mean and RMSE has a unit of $\mu g\ m^{-3}$. N_RMSE is defined as in Table 2.

Figure S6. Spatial distribution of annual mean simulated (a, b) aerosol extinction at surface, (c, d) aerosol absorption at surface, (e, f) Aerosol Optical Depth (AOD), and (g, h) absorbing AOD from RRM-EHR-PD (left column) and the relative difference between RRM-SE-PD and RRM-EHR-PD (right column) over North America. The relative difference for field X is calculated as: $\left(\frac{X_{def} - X_{se}}{X_{se}}\right) \times 100\%$, where "se" and "def" subscripts refer to the simulations with new and default emission treatment respectively. Mean, RMSE and normalized RMSE (N_RMSE) are indicated at the top right corner of the panels. Mean and RMSE has a unit of $\mu g\ m^{-3}$. N_RMSE is defined as in Table 2.
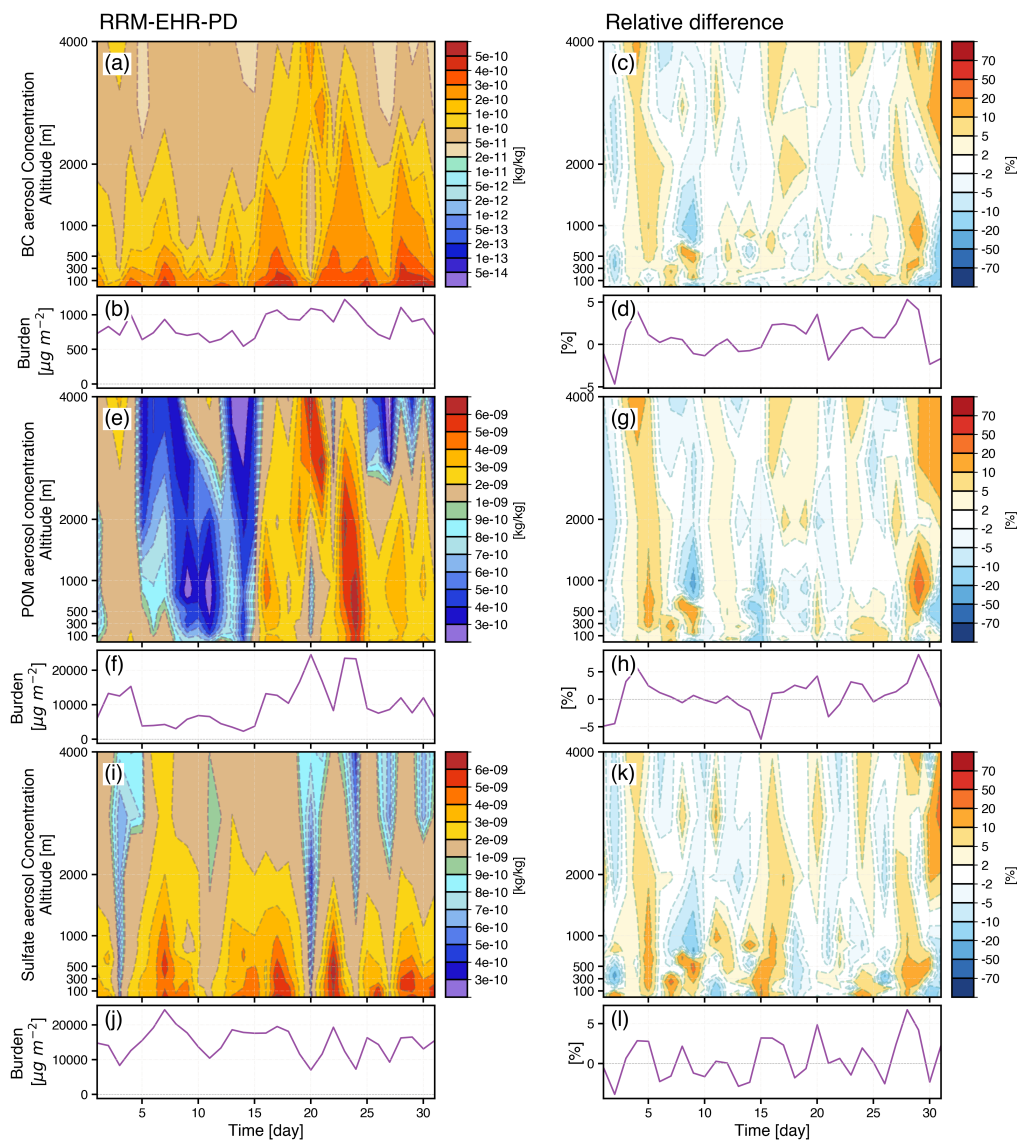
Figure S7. Daily mean concentration profile and burden time-series of (a-d) BC, (e-h) POM, and (i-l) Sulfate aerosols. All profiles are shown during the month of July 2016 over highly polluted locations in the eastern United States (42°N and 70°W). Simulated vertical distribution and burden time series from RRM-EHR-PD (left column) and the relative difference between RRM-EHR-PD and RRM-SE-PD (right column) are shown.
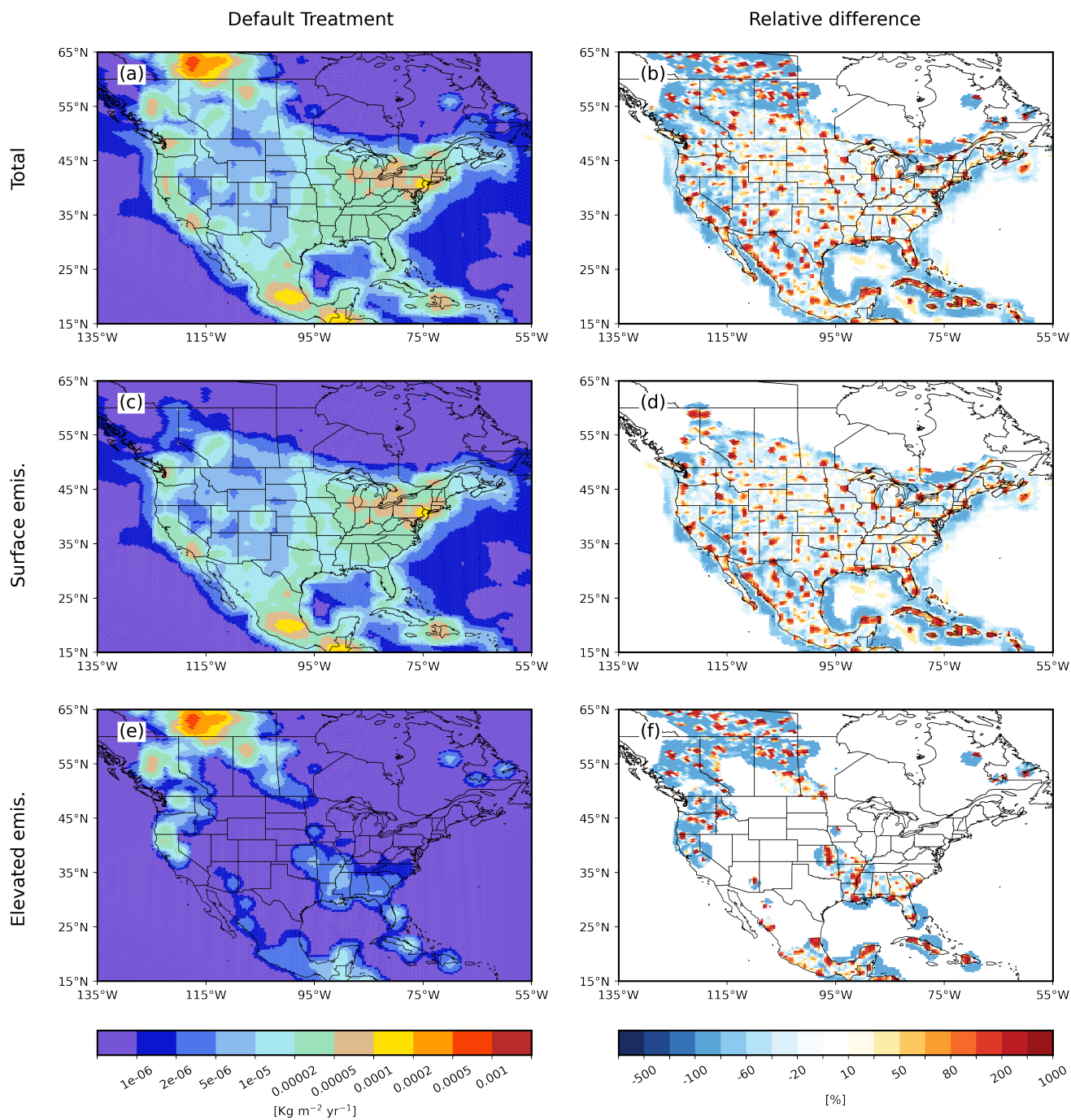
Figure S8. Spatial distribution of simulated annual mean source and major contributing components of BC over North America from PD RRM simulations. The distribution from RRM-PD (with default emission treatment) (left column) and the associated relative difference between RRM-SE-PD (with new emission treatment) and RRM-PD (right column) are shown.
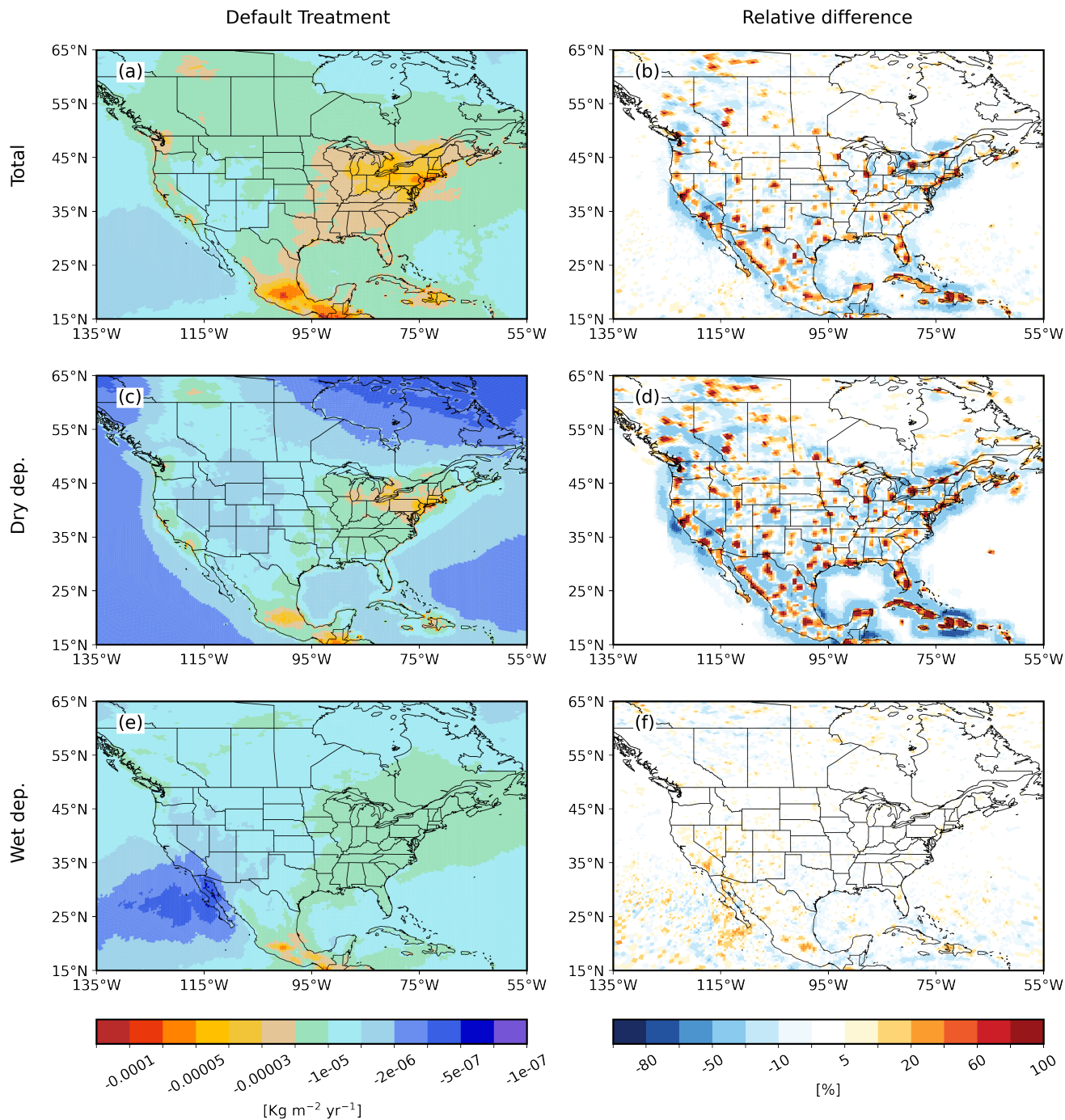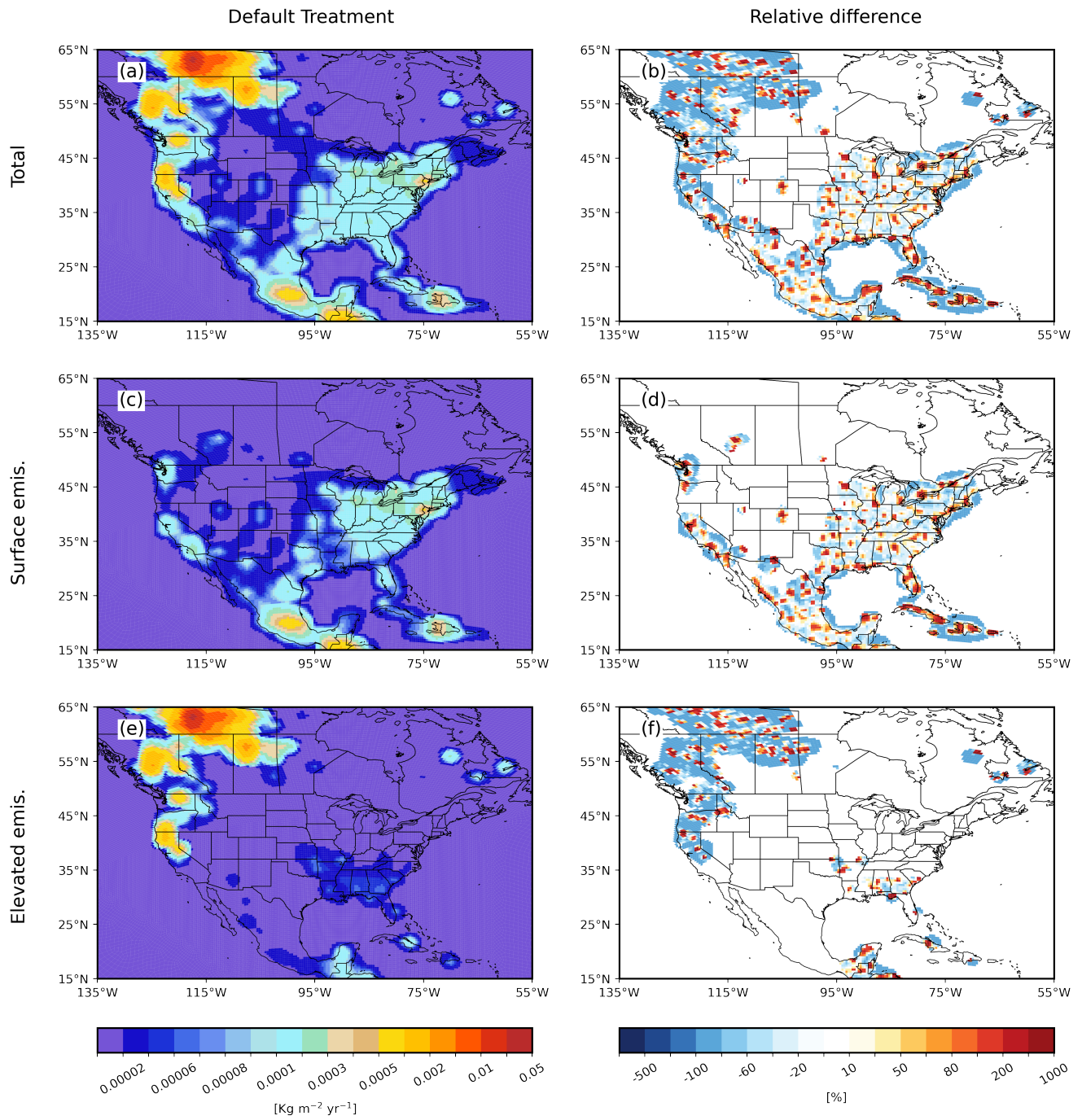
Figure S9. Spatial distribution of simulated annual mean sinks and major contributing components of BC over North America from PD RRM simulations. The distribution from RRM-PD (with default emission treatment) (left column) and the associated relative difference between RRM-SE-PD (with new emission treatment) and RRM-PD (right column) are shown.

Figure S10. Same as Fig. S8, except for POM.
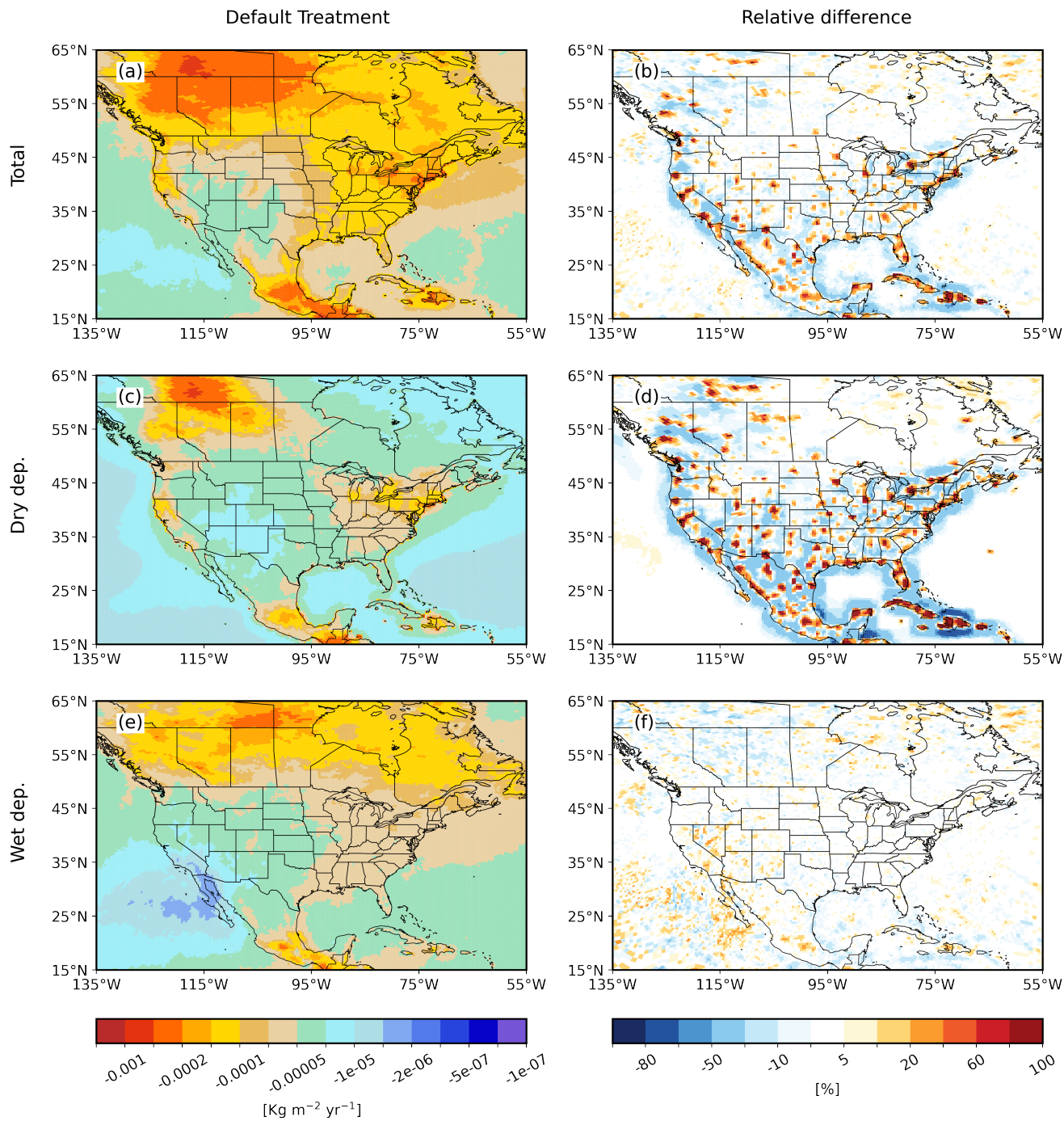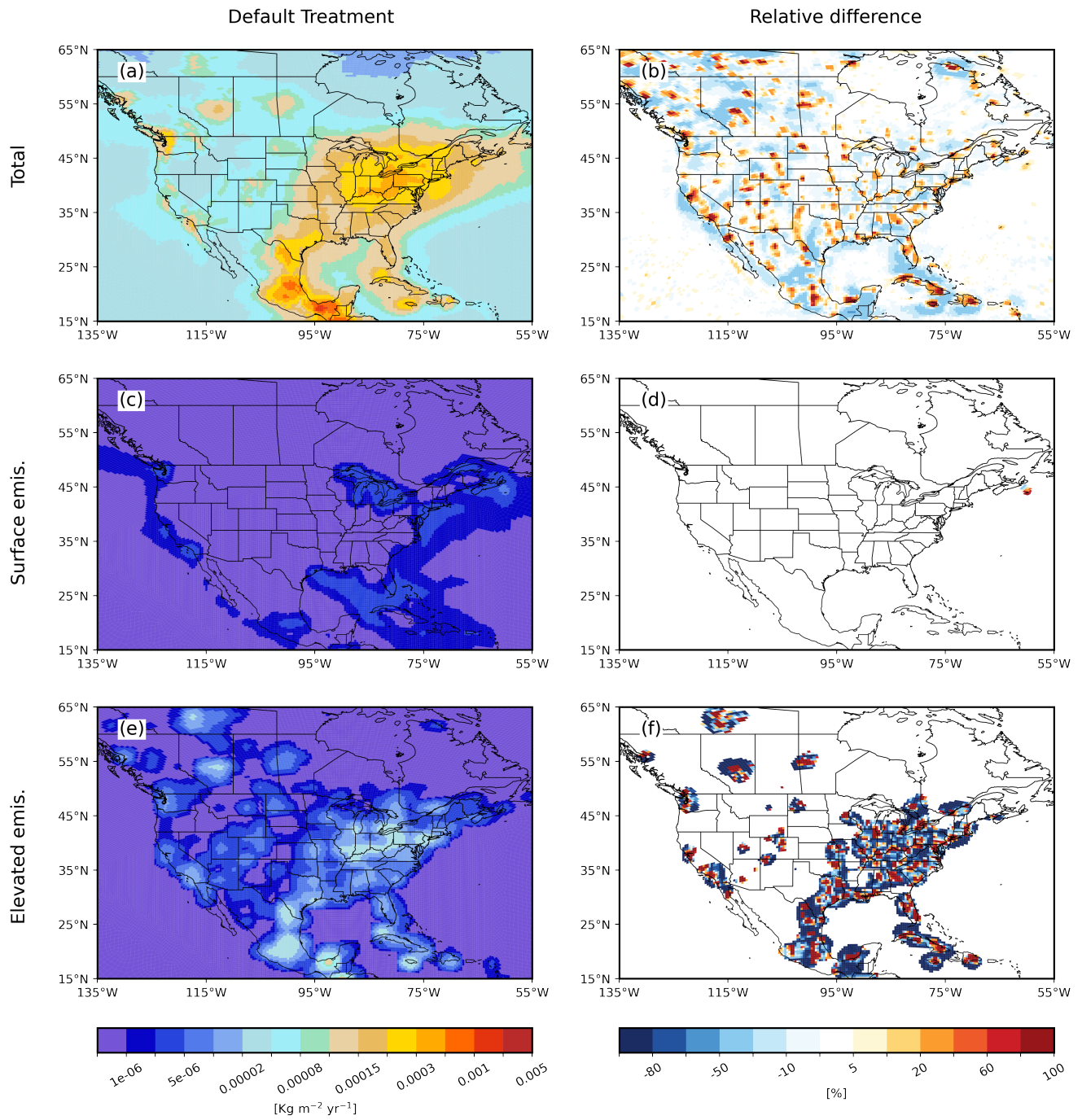
Figure S11. Same as Fig. S9, except for POM.

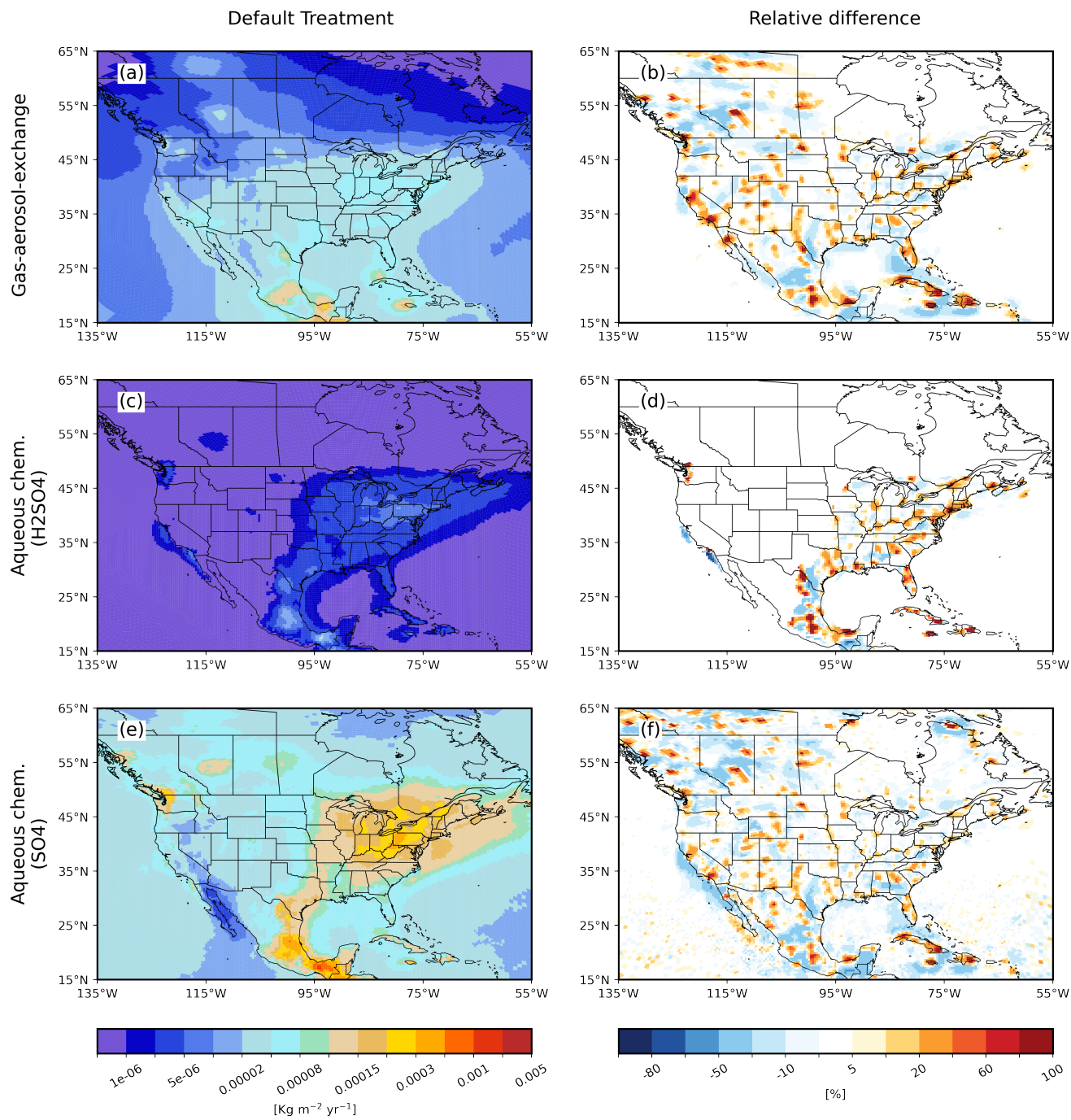**Figure S12.** Same as Fig. S8, except for Sulfate.

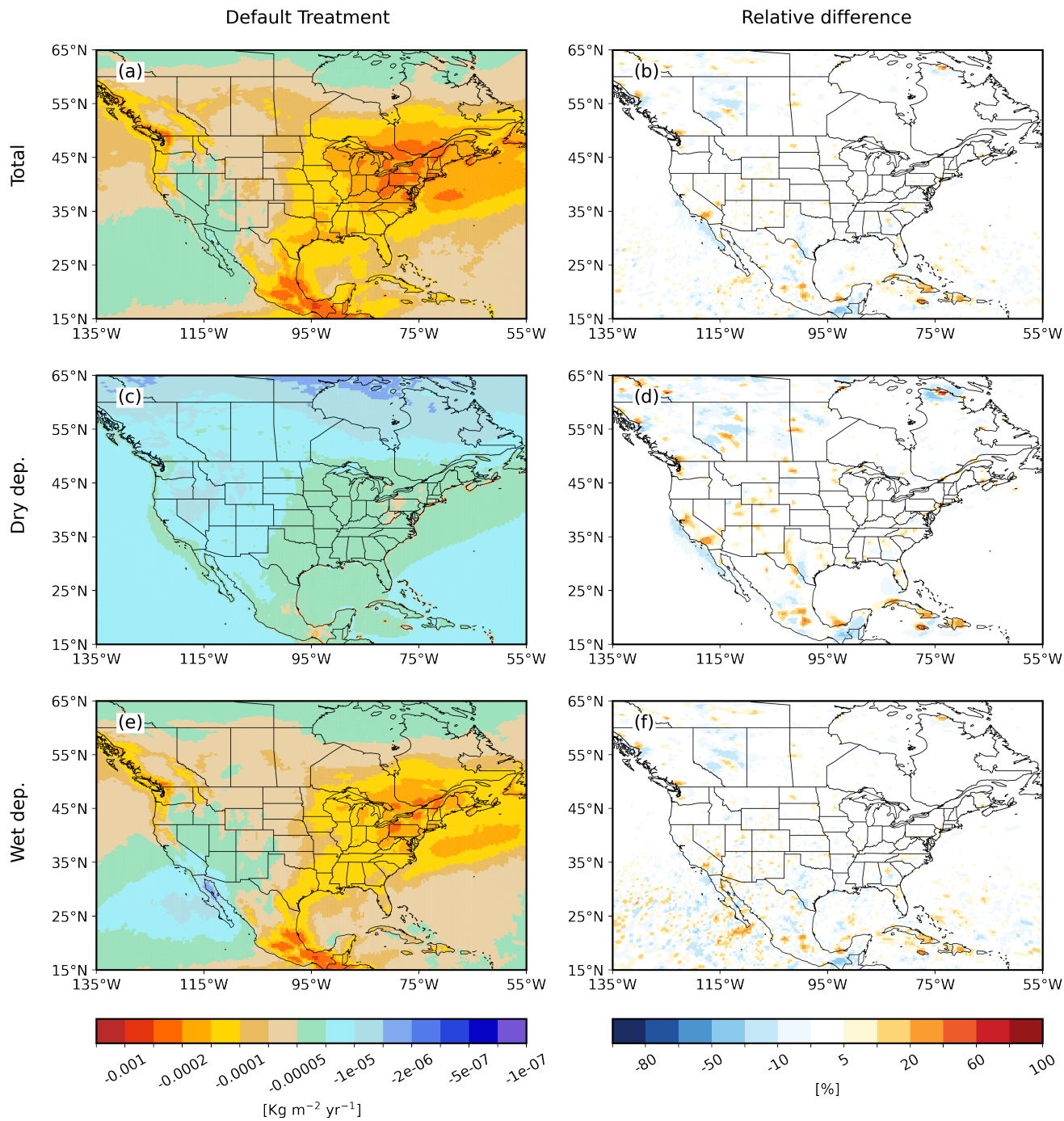**Figure S13.** Same as Fig. S8, except for sulfate components.

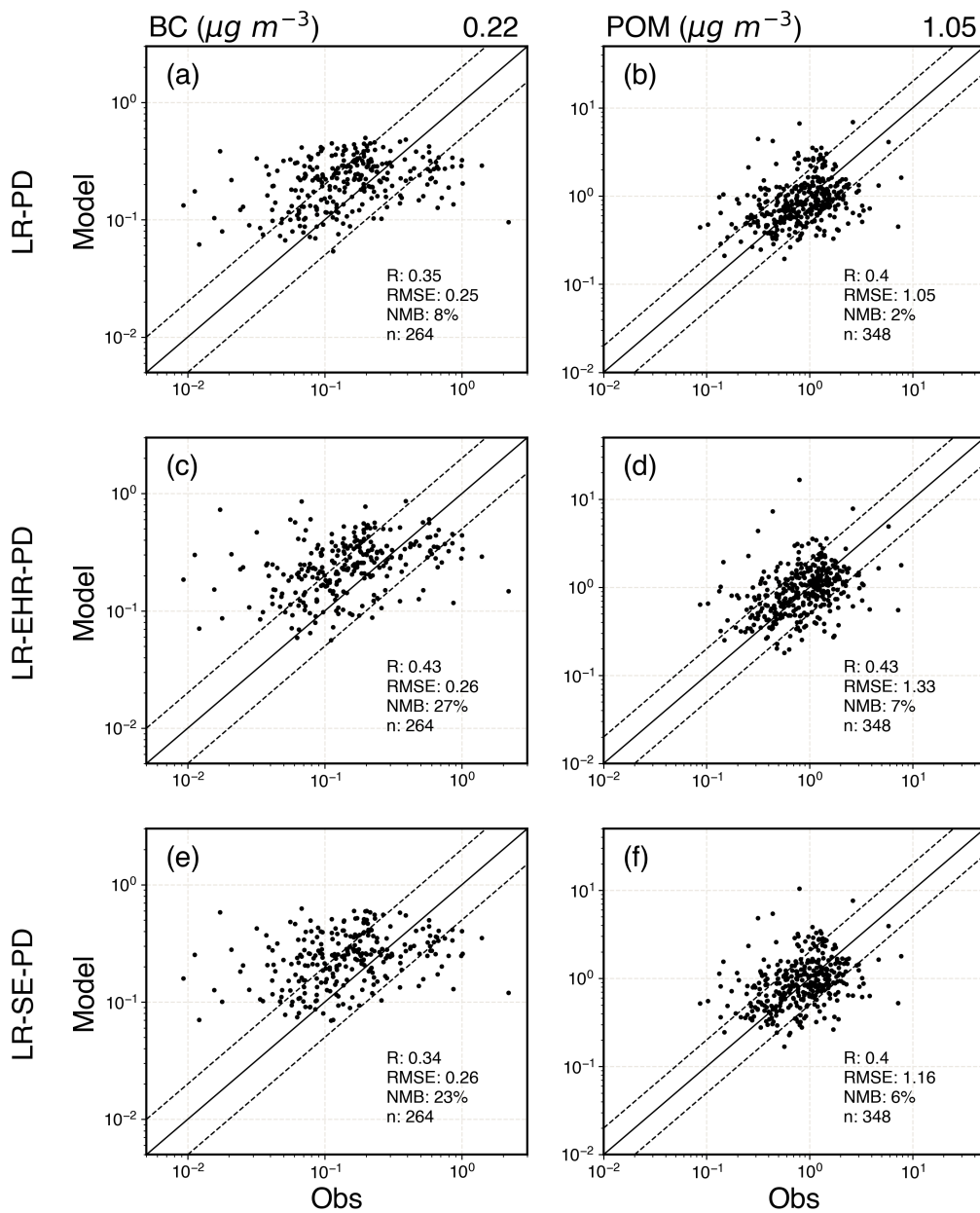**Figure S14.** Same as Fig. S9, except for Sulfate.

Figure S15. Scatter plots between simulated and observed monthly mean surface concentrations of (a, c) Black Carbon (BC) and (b, d) Primary Organic Matter (POM). Observations of the surface concentrations are from IMPROVE for the simulation year of 2016. Scatter plot statistics compare the spearman's correlation (R), number of data points (n), RMSE, NMB values between (a, b) LR-PD, (c, d) LR-EHR-PD, and (e, f) LR-SE-PD simulation. RMSE and NMB are defined as in Table 2. Solid lines indicate the 1:1 ratio, and the dashed lines indicate the 1:2 and 2:1 ratio. The values at the top of each column indicate the observed mean.
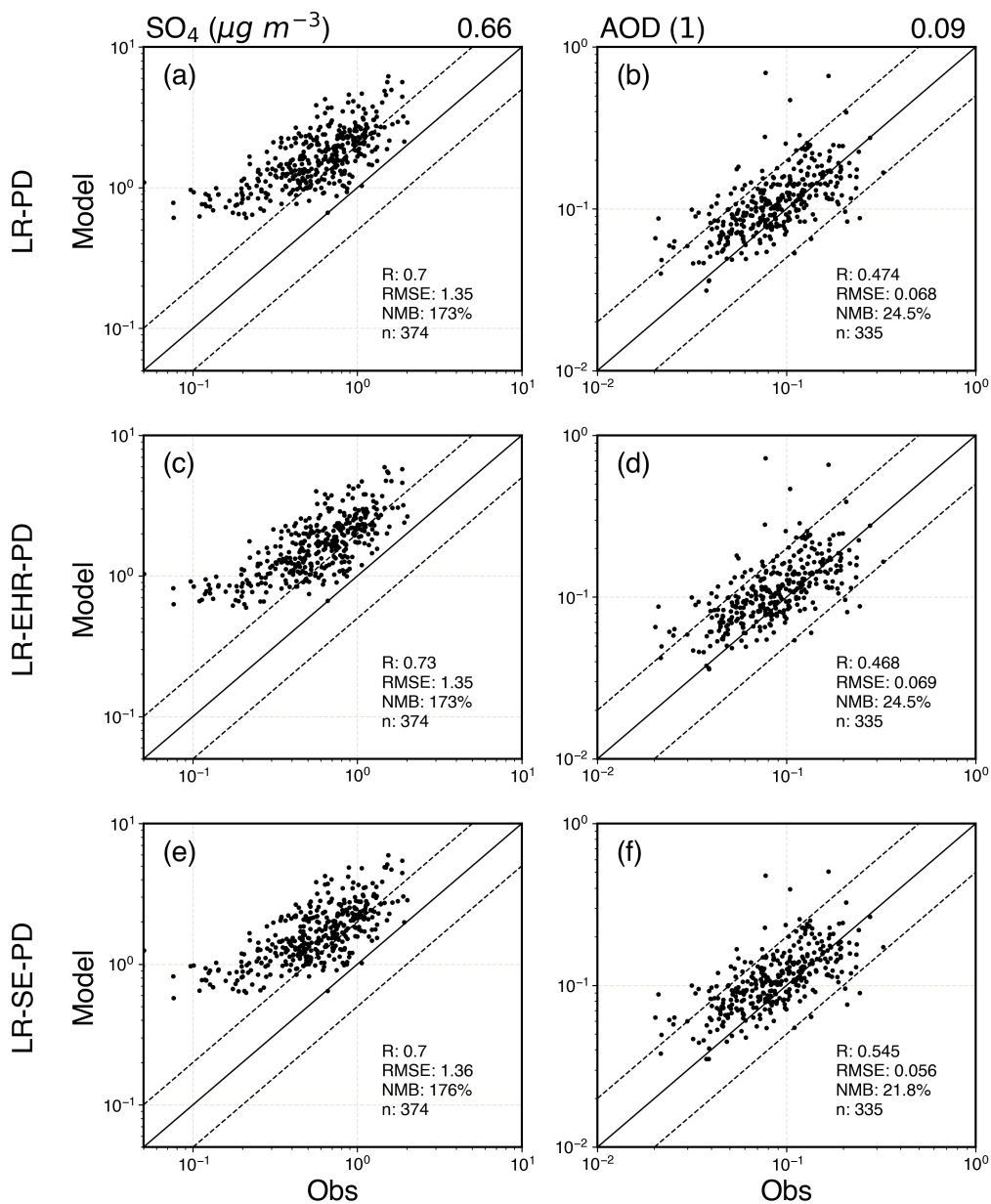
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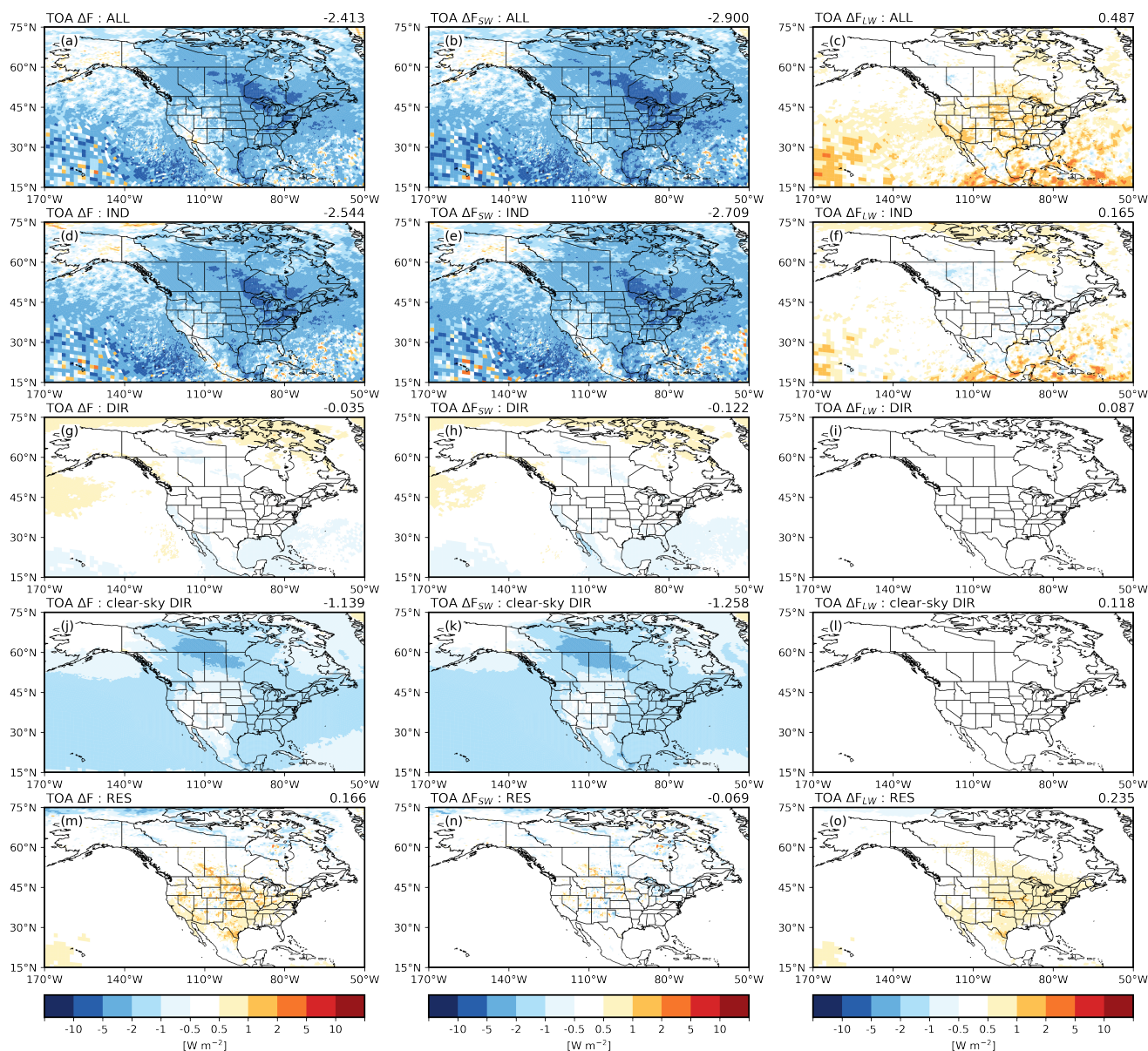
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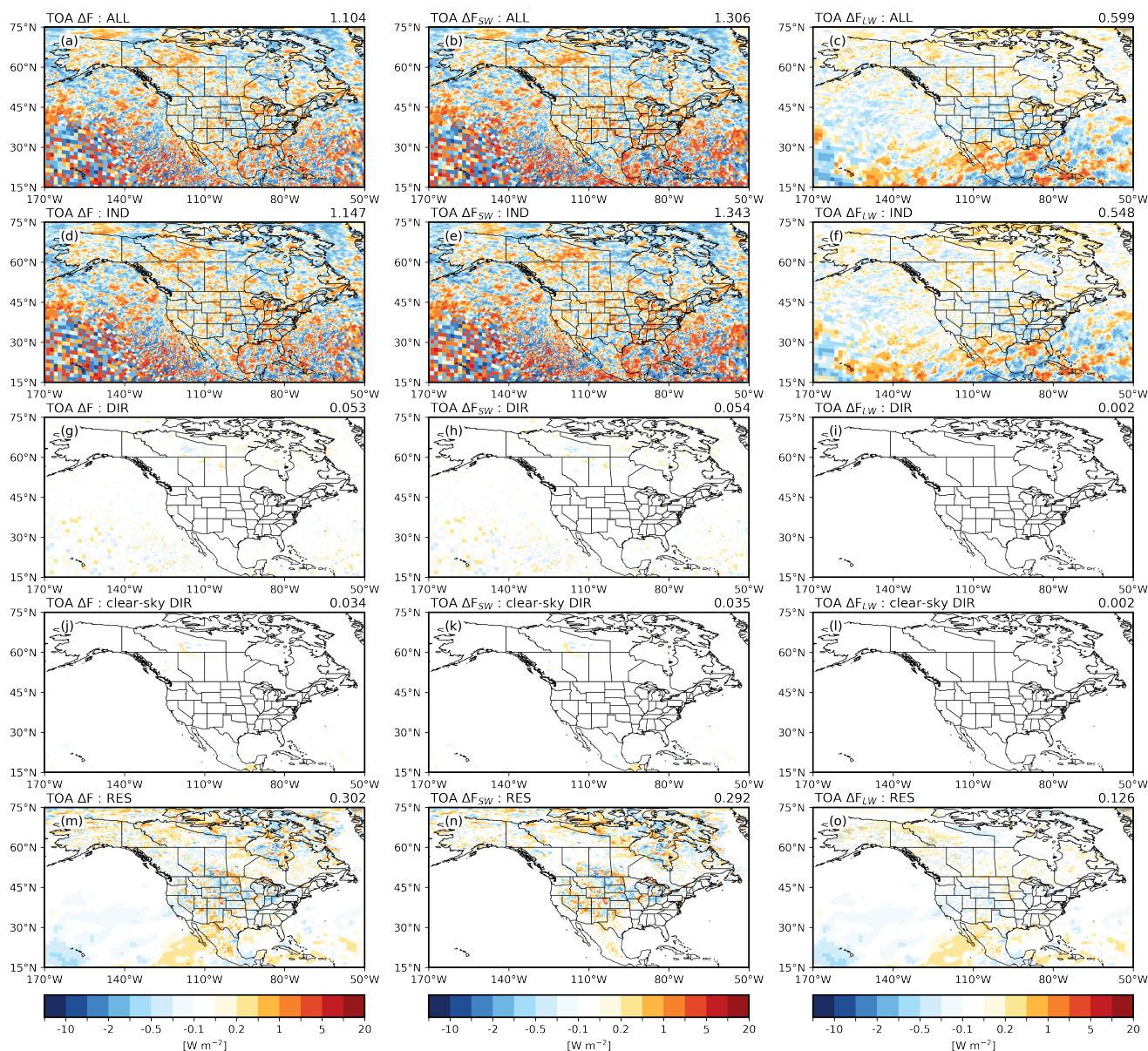
Figure S18. Spatial distribution of anthropogenic aerosol radiative forcing (ΔF) difference over North America at the top of atmosphere (TOA) between simulations with revised and default treatment. Decomposed net (left column), shortwave (middle column), and longwave (right column) ΔF are calculated over North America (NA). ALL indicates the total ΔF calculated from PD (2014) - PI (1850) simulations. ACI refers to ΔF from aerosol-cloud interactions. "SW" and "LW" subscripts indicate shortwave and longwave ΔF. RMSE over NA are indicated at the top right corner of the panels.
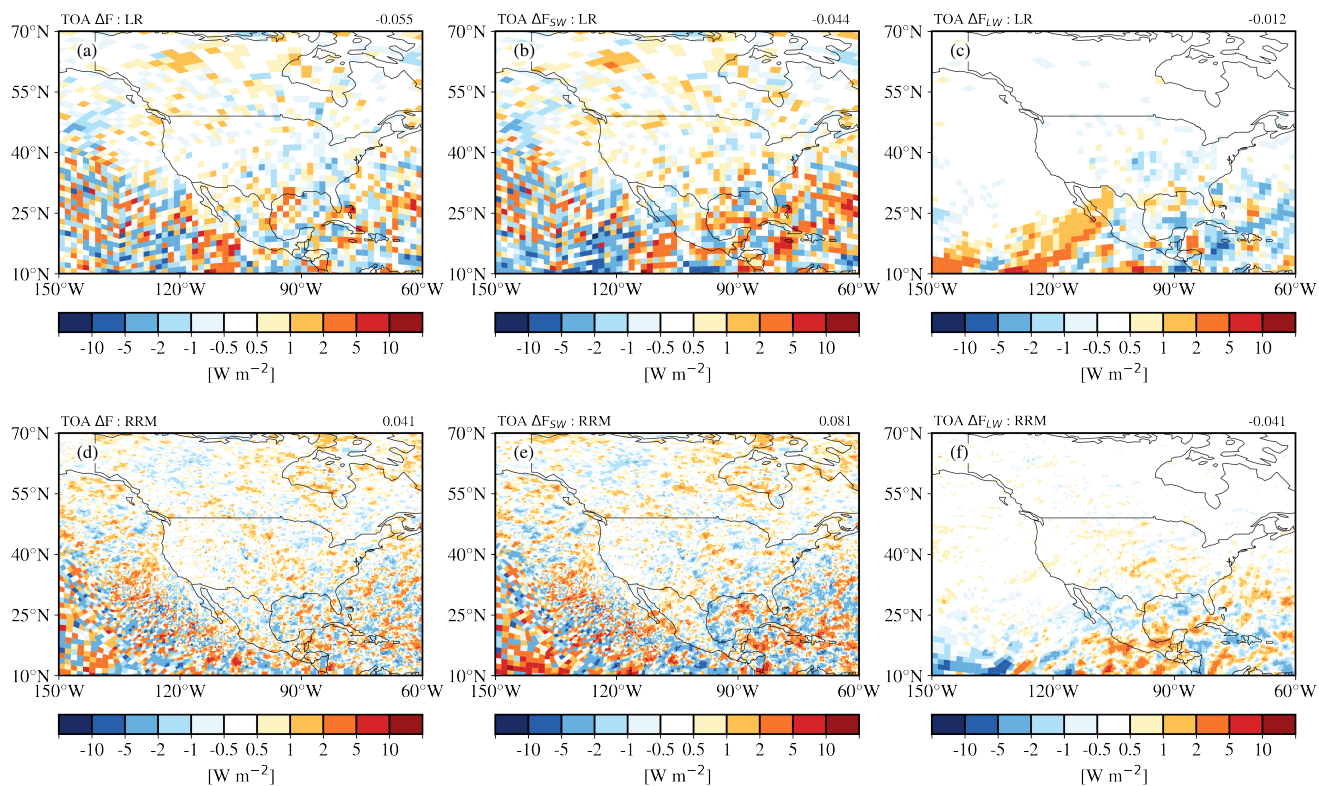
Figure S19. Spatial distribution of anthropogenic aerosol radiative forcing differences (net: a,d; shortwave: b,e; and longwave: c,f) at the top of atmosphere (TOA) between simulations with revised and default treatment from (a-c) LR (ne30pg2) and (d-f) RRM simulations. "SW" and "LW" subscripts indicate shortwave and longwave forcing. Area-weighted regional mean differences are indicated at the upper-right corner of each panel.