Supplementary Information

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

Taufiq Hassan, Kai Zhang, Jianfeng Li, Balwinder Singh, Shixuan Zhang, Hailong Wang, & Po-Lun Ma

Atmospheric Sciences and Global Change Division, Pacific Northwest National Laboratory, Richland, WA, USA.

Correspondence to Taufiq Hassan (taufiq.hassan@pnnl.gov) and Kai Zhang (kai.zhang@pnnl.gov)

Contents of this file

1. **Table S1 and S2**
2. **Figures S1 to S13**

Table S1. EAMv2 anthropogenic aerosol emissions data statistics in the default emission treatment for present-day (PD) LR simulations. Statistics are shown for both the surface and elevated emissions of different aerosol species. All estimates are over the North America land surface. 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 "accurate" subscript indicates data that preserve spatial heterogeneity and conserve mass. The "lin" subscript indicates linearly interpolated data used in the default treatment. 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 [$\times 10^{-12}$ 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	5	2.504	10	-0.209	4	45
	elevated	2	0.902	10	-0.102	4	34
POM	surface	20	2.924	40	-0.204	20	42
	elevated	40	0.285	300	-0.100	100	33
SO4	surface	0.7	0.219	1	-0.140	0.4	36
	elevated	5	2.052	20	-0.276	9	57

Table S2. 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
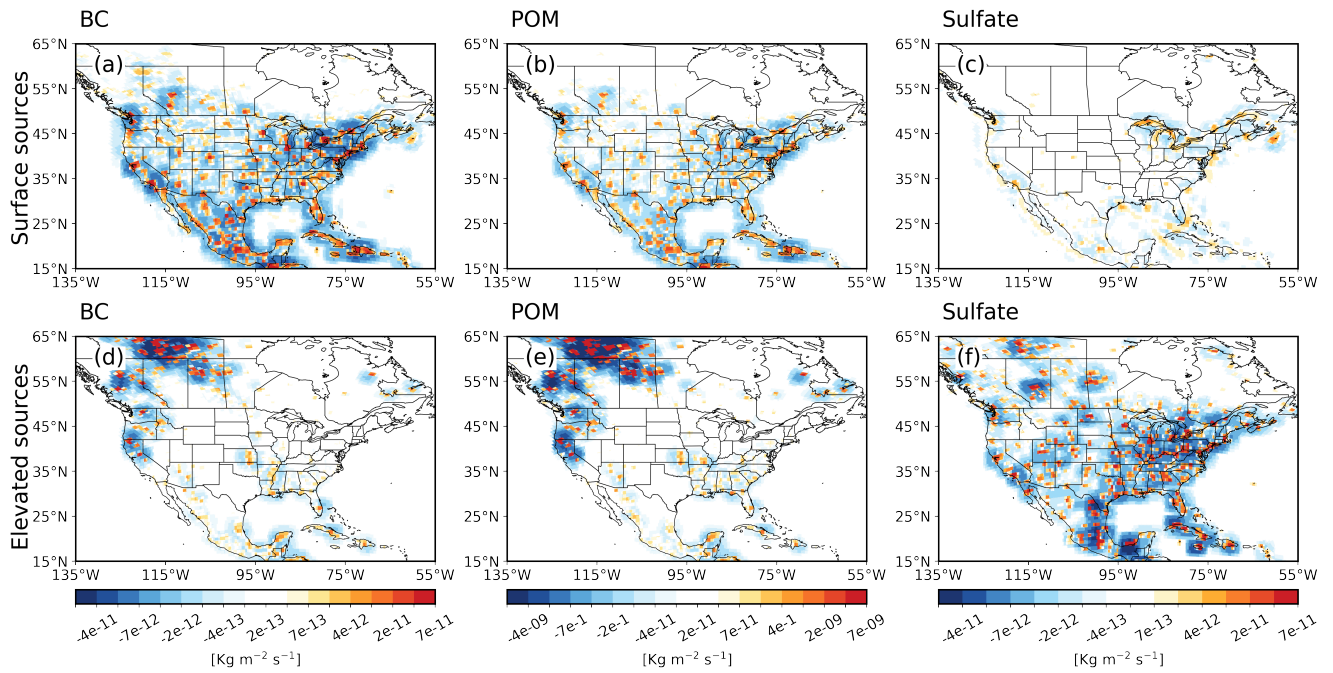
Figure S1. Spatial distribution of difference in surface (top) and elevated (bottom) emissions used in RRM-SE-PD and RRM-PD over North America for (a) Black Carbon (BC), (b) Primary Organic Matter (POM), and (c) Sulfate (SO4) aerosols. The units are in kg/m^2/s.
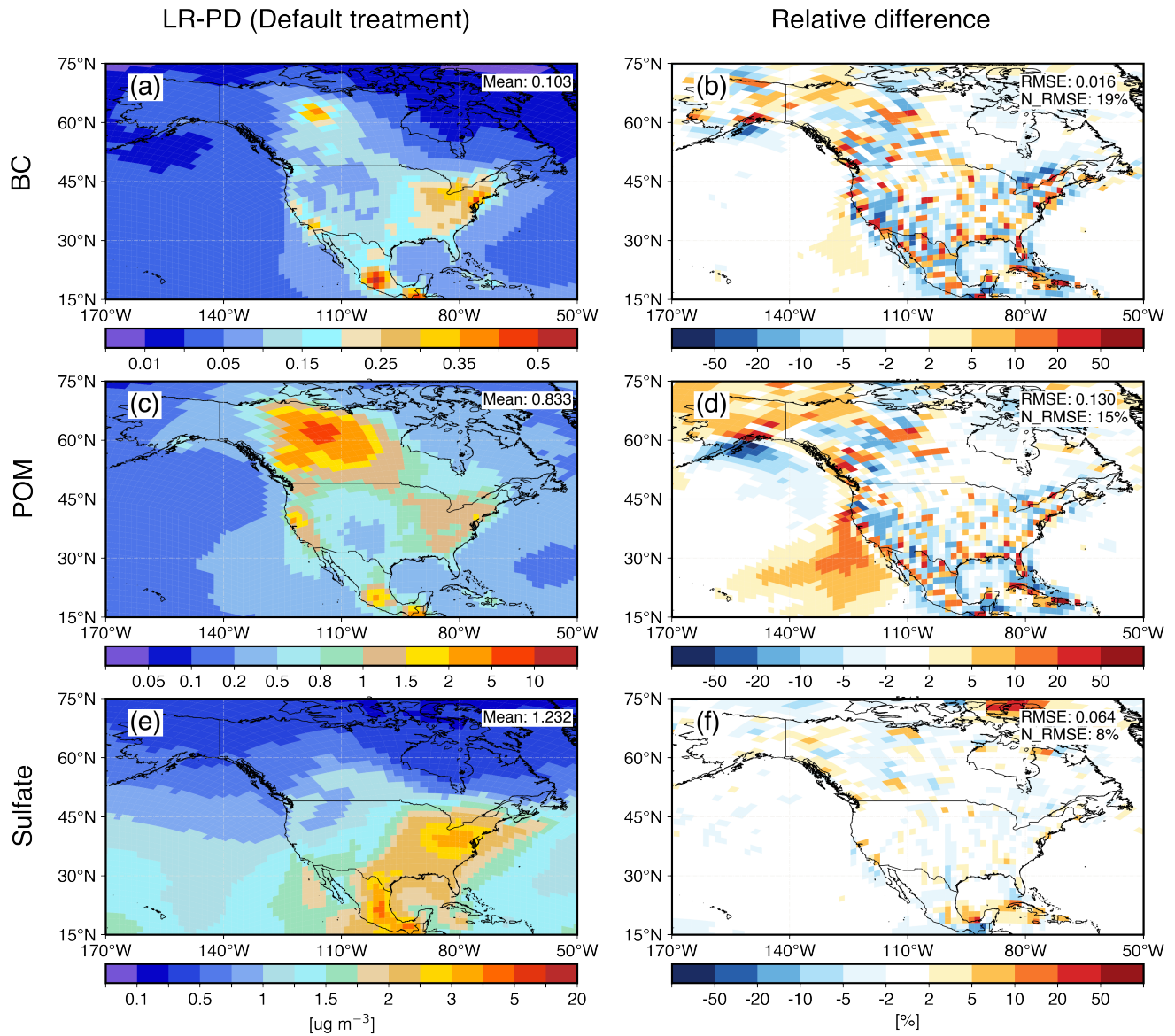
Figure S2. Simulated spatial distribution of annual mean aerosol surface concentration from LR-PD (left column) and the relative difference between LR-SE-PD and LR-PD (right column) over North America. Distributions are shown for (a, b) Black Carbon (BC), (c, d) Primary Organic Matter (POM), and (e, f) Sulfate aerosols. The relative difference for field X is calculated as: $\left(\frac{X_{se} - X_{def}}{X_{def}}\right) \times 100\%$, 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.
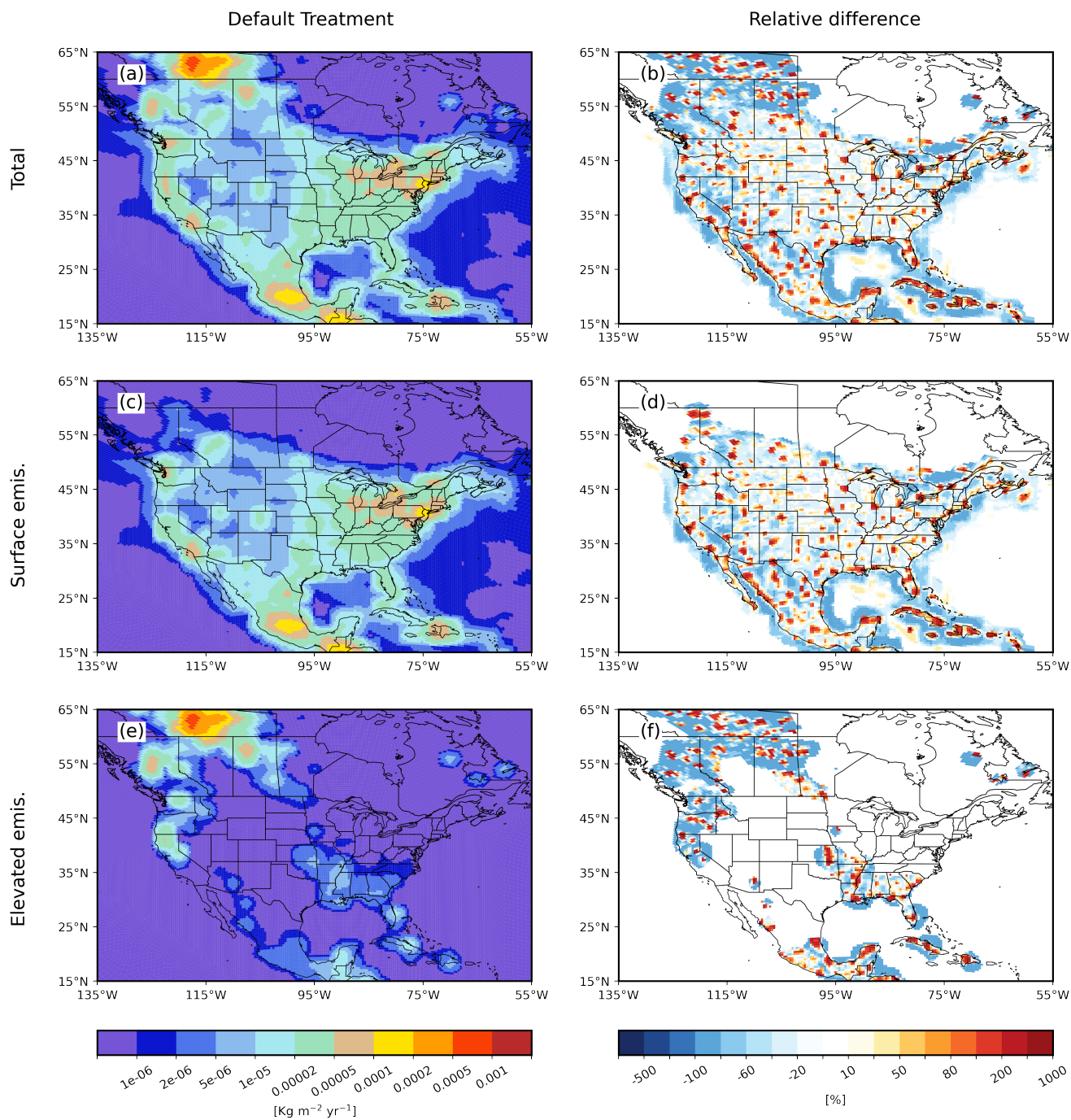
Figure S3. Spatial distribution of simulated annual mean source and major contributing components of BC over North America from PD RRM simulations. The distribution from RRM-PD (with default emission treatment) (left column) and the associated relative difference between RRM-SE-PD (with new emission treatment) and RRM-PD (right column) are shown.
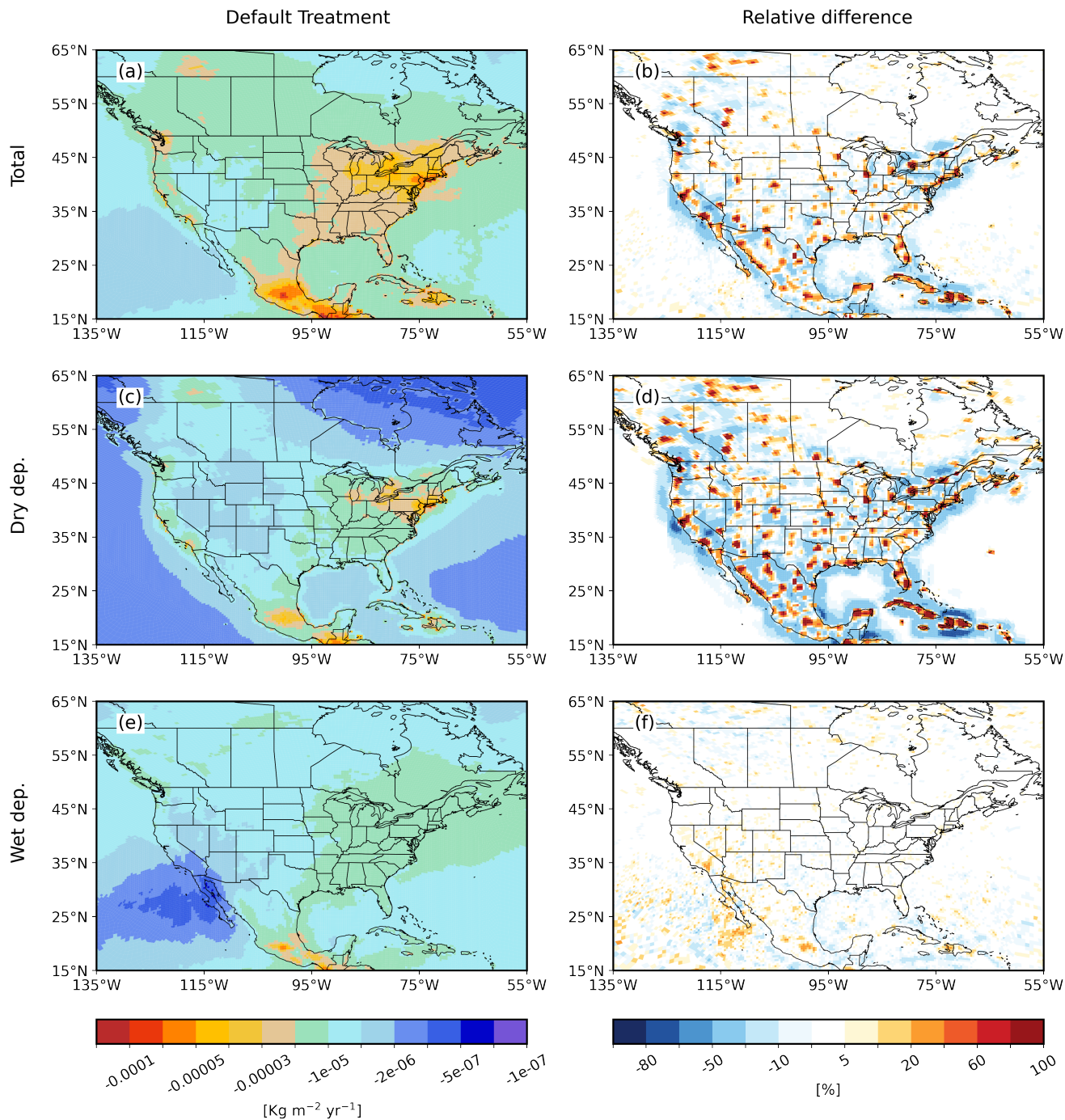
Figure S4. Spatial distribution of simulated annual mean sinks and major contributing components of BC over North America from PD RRM simulations. The distribution from RRM-PD (with default emission treatment) (left column) and the associated relative difference between RRM-SE-PD (with new emission treatment) and RRM-PD (right column) are shown.
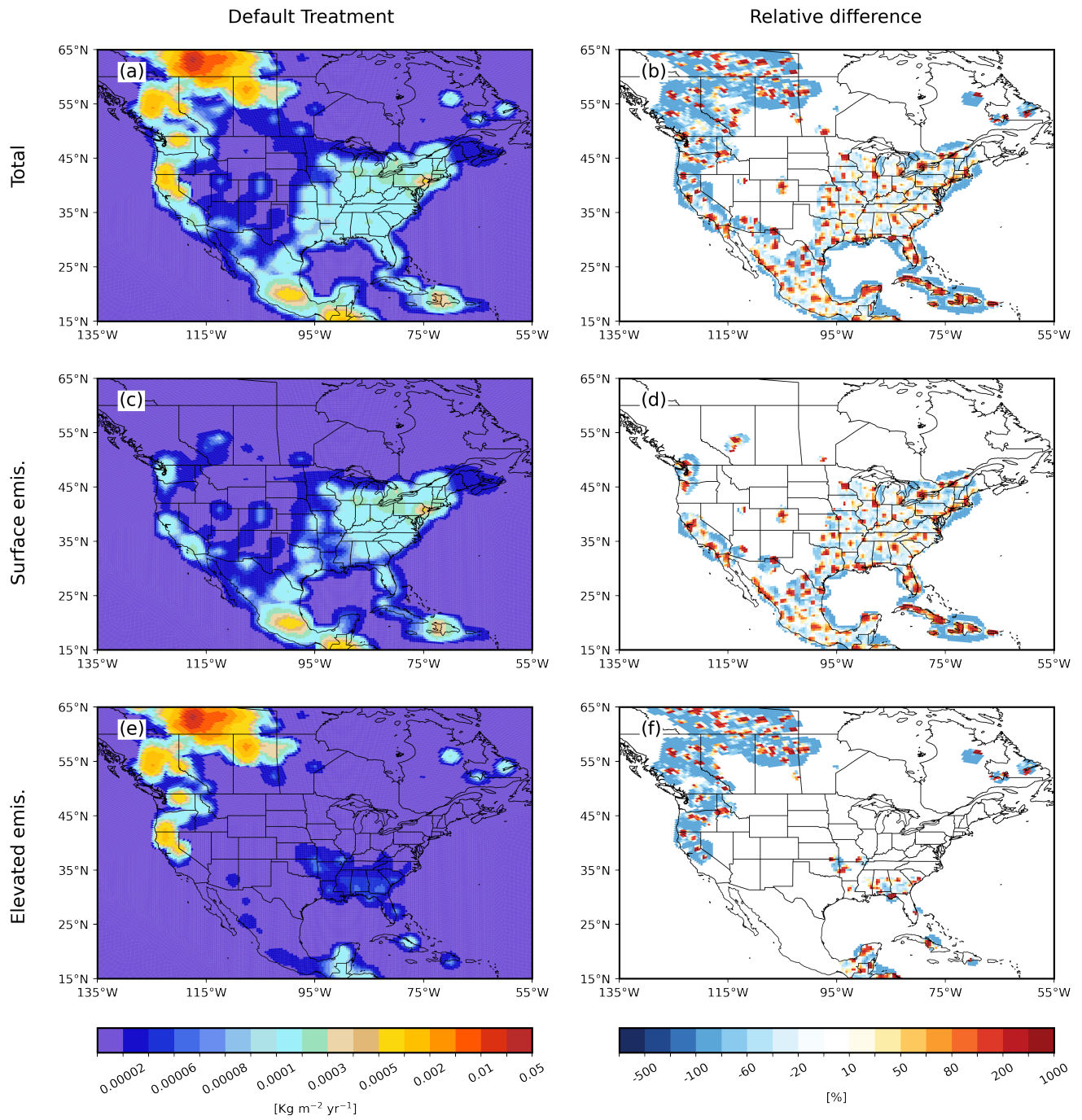
Figure S5. Same as Fig. S3, except for POM.
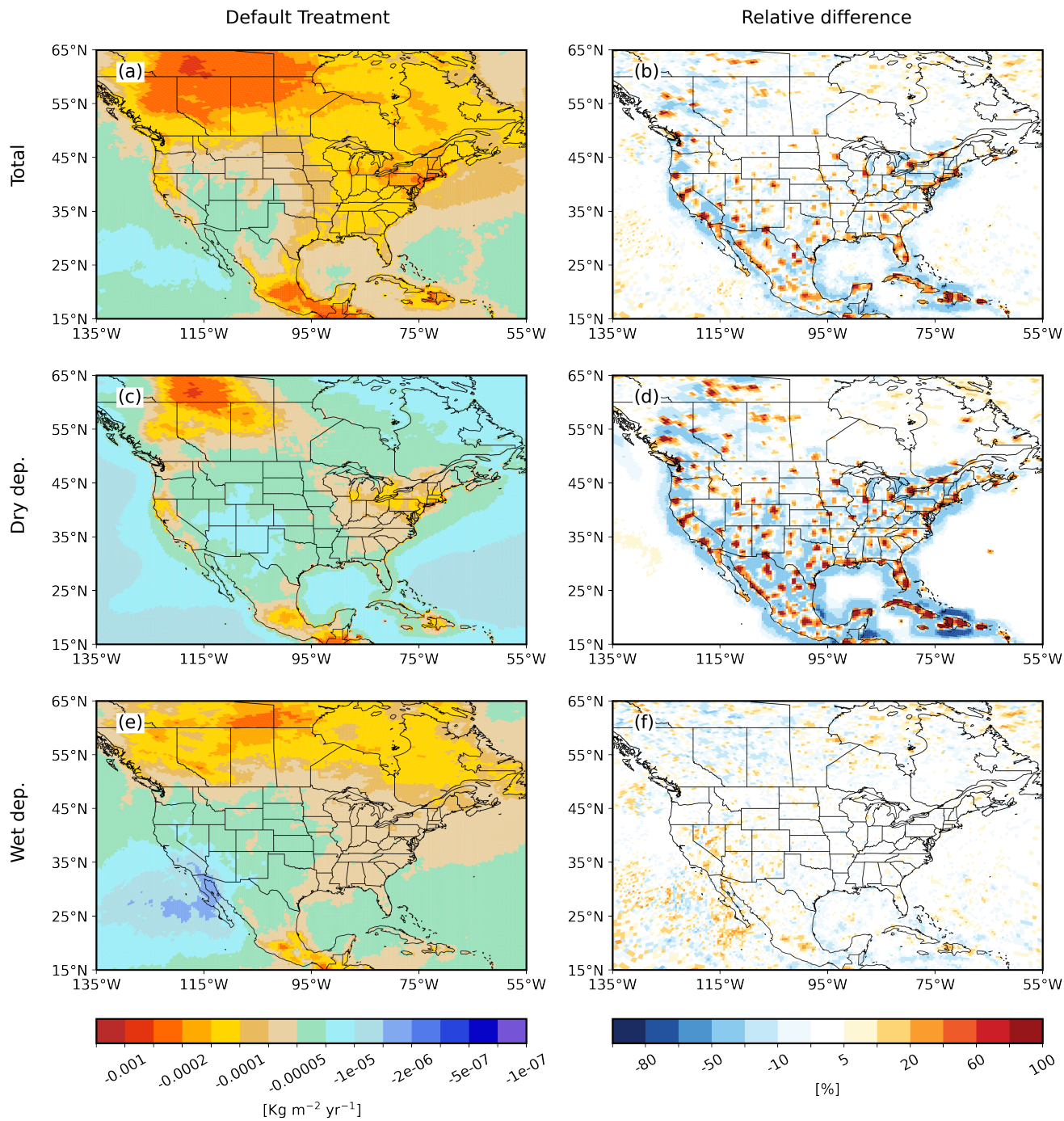
Figure S6. Same as Fig. S4, except for POM.
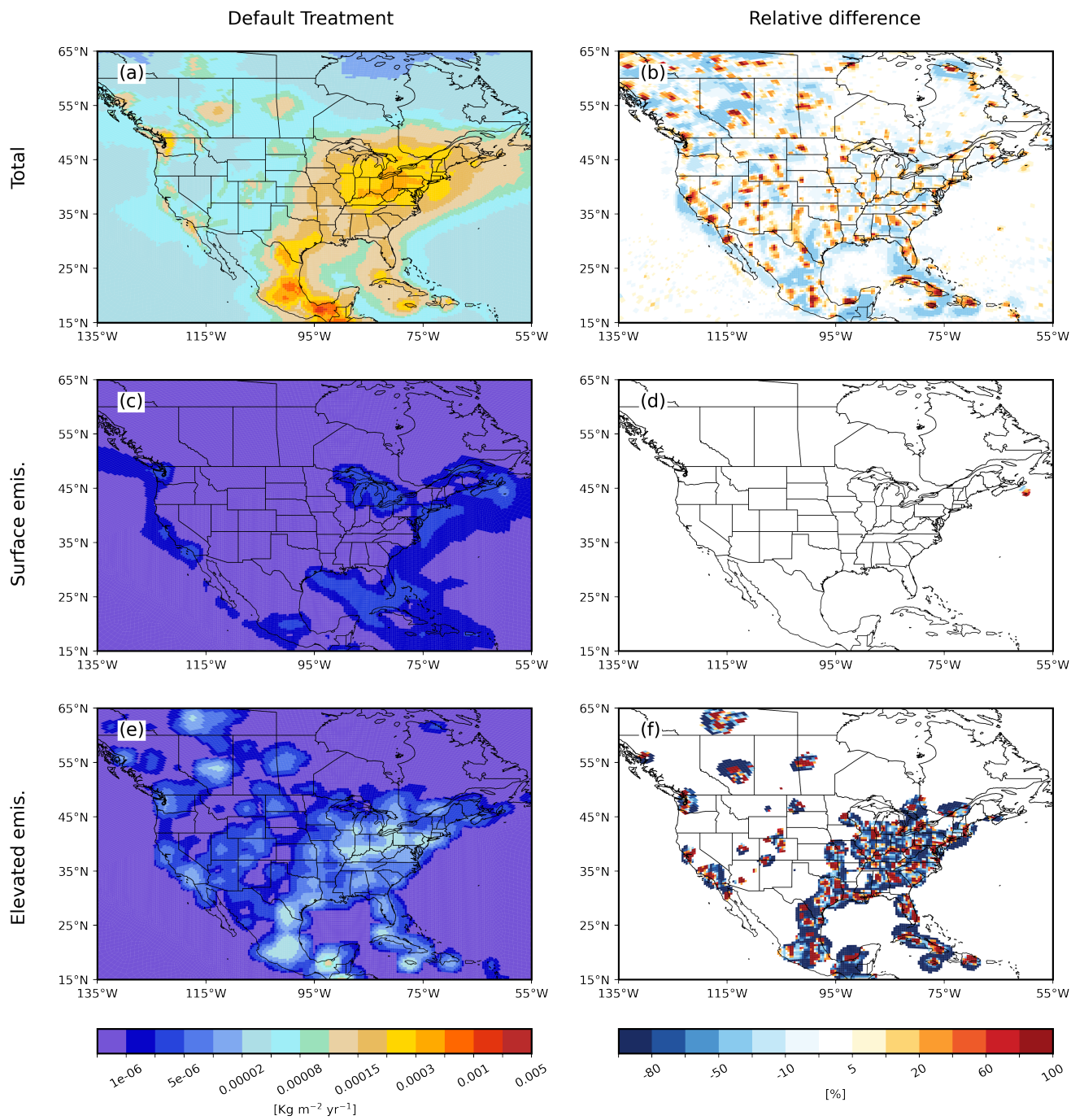
Figure S7. Same as Fig. S3, except for Sulfate.
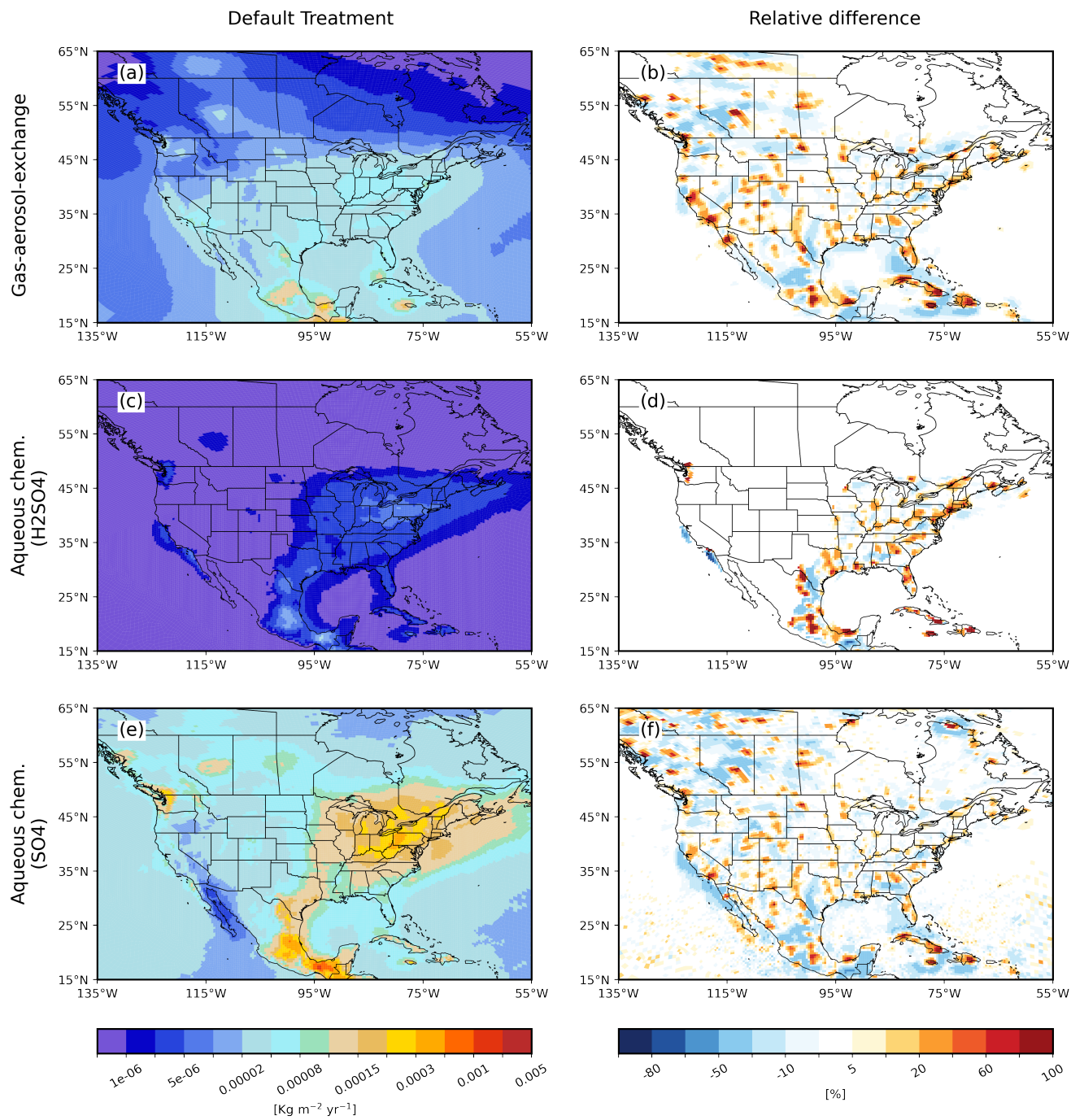
Figure S8. Same as Fig. S3, except for sulfate components.
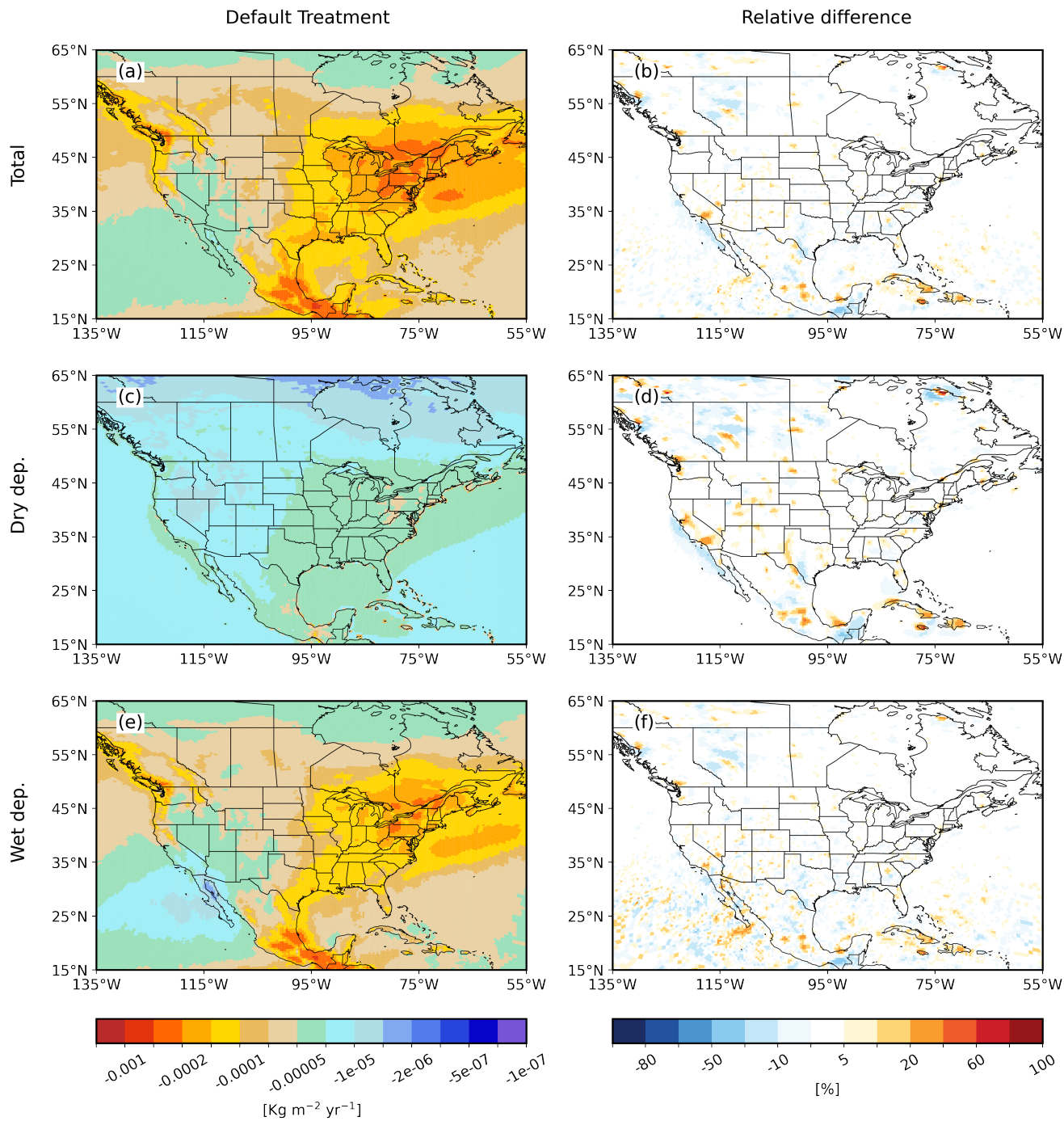
Figure S9. Same as Fig. S4, except for Sulfate.
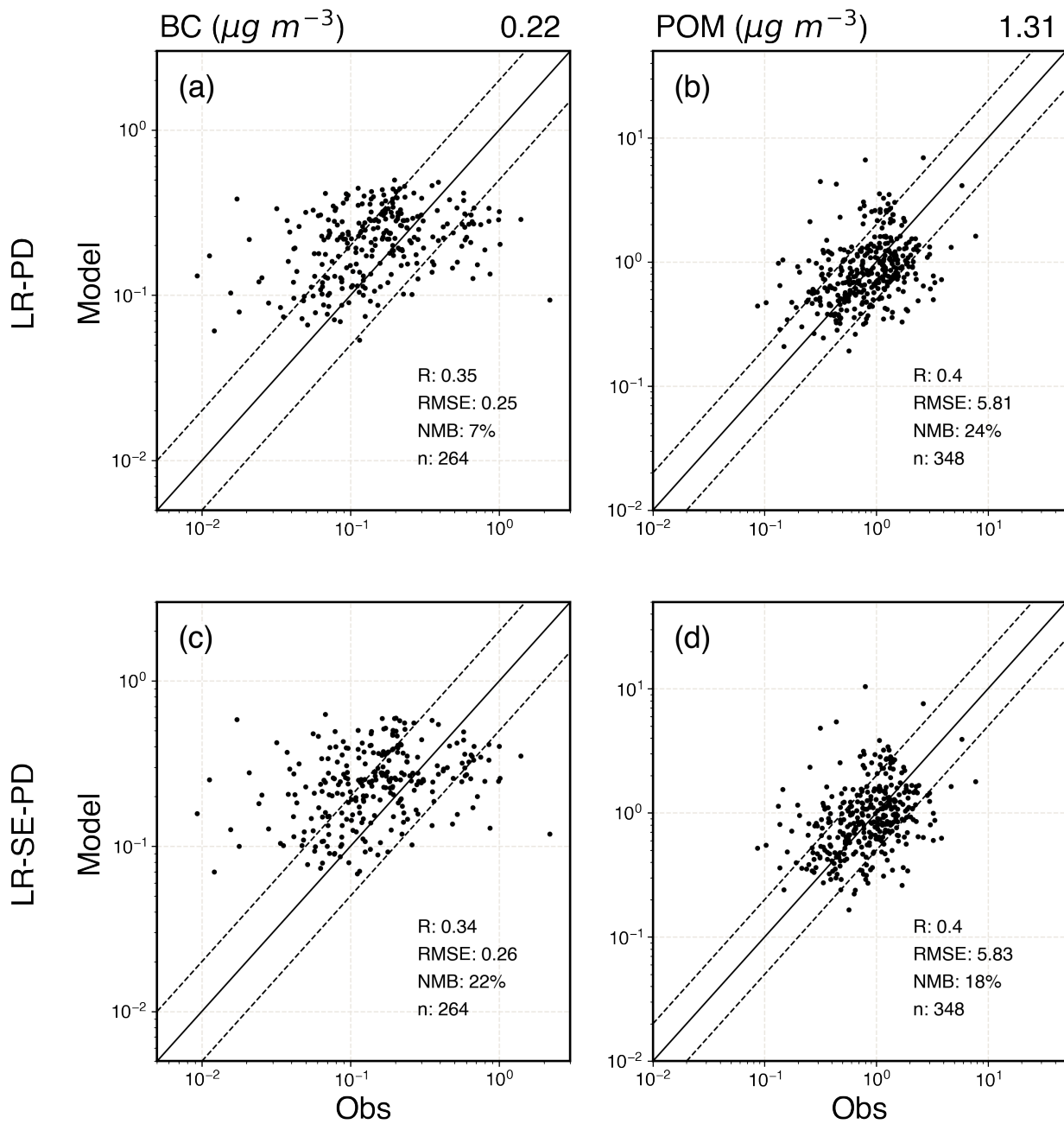
Figure S10. Scatter plots between simulated and observed monthly mean surface concentrations of (a, c) Black Carbon (BC) and (b, d) Primary Organic Matter (POM). Observations of the surface concentrations are from IMPROVE for the simulation year of 2016. Scatter plot statistics compare the spearman's correlation (R), number of data points (n), RMSE, NMB values between (a, b) LR-PD and (c, d) 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.
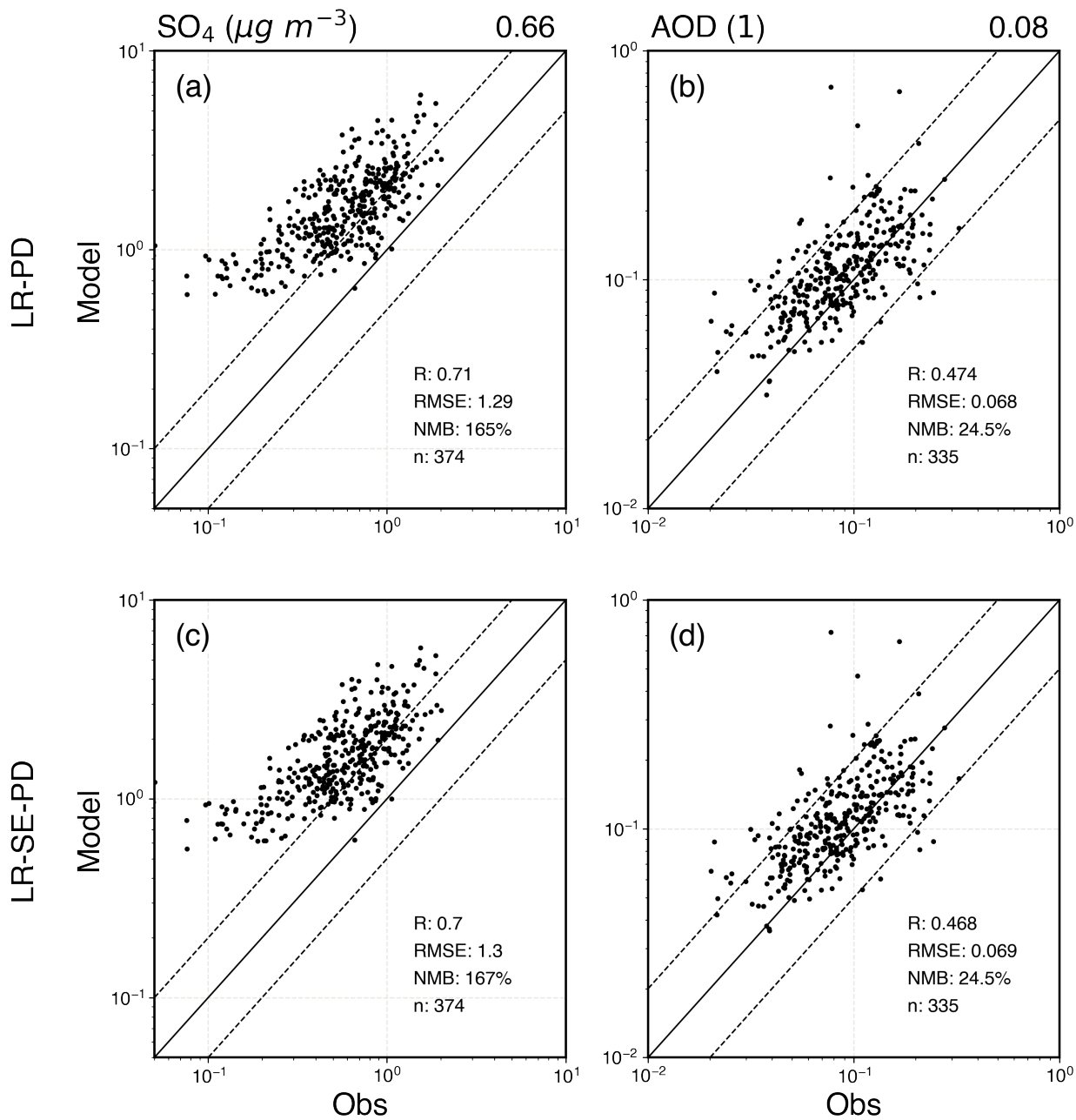
Figure S11. Scatter plots between simulated and observed monthly mean surface concentrations of (a, c) sulfate (SO_4) aerosols and (b, d) Aerosol Optical Depth (AOD) at 550 nm. Observations of the surface concentrations and AOD are from IMPROVE and AERONET respectively for the simulation year of 2016. Scatter plot statistics compare the spearman's correlation (R), number of data points (n), RMSE, NMB values between (a, b) LR-PD and (c, d) 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.
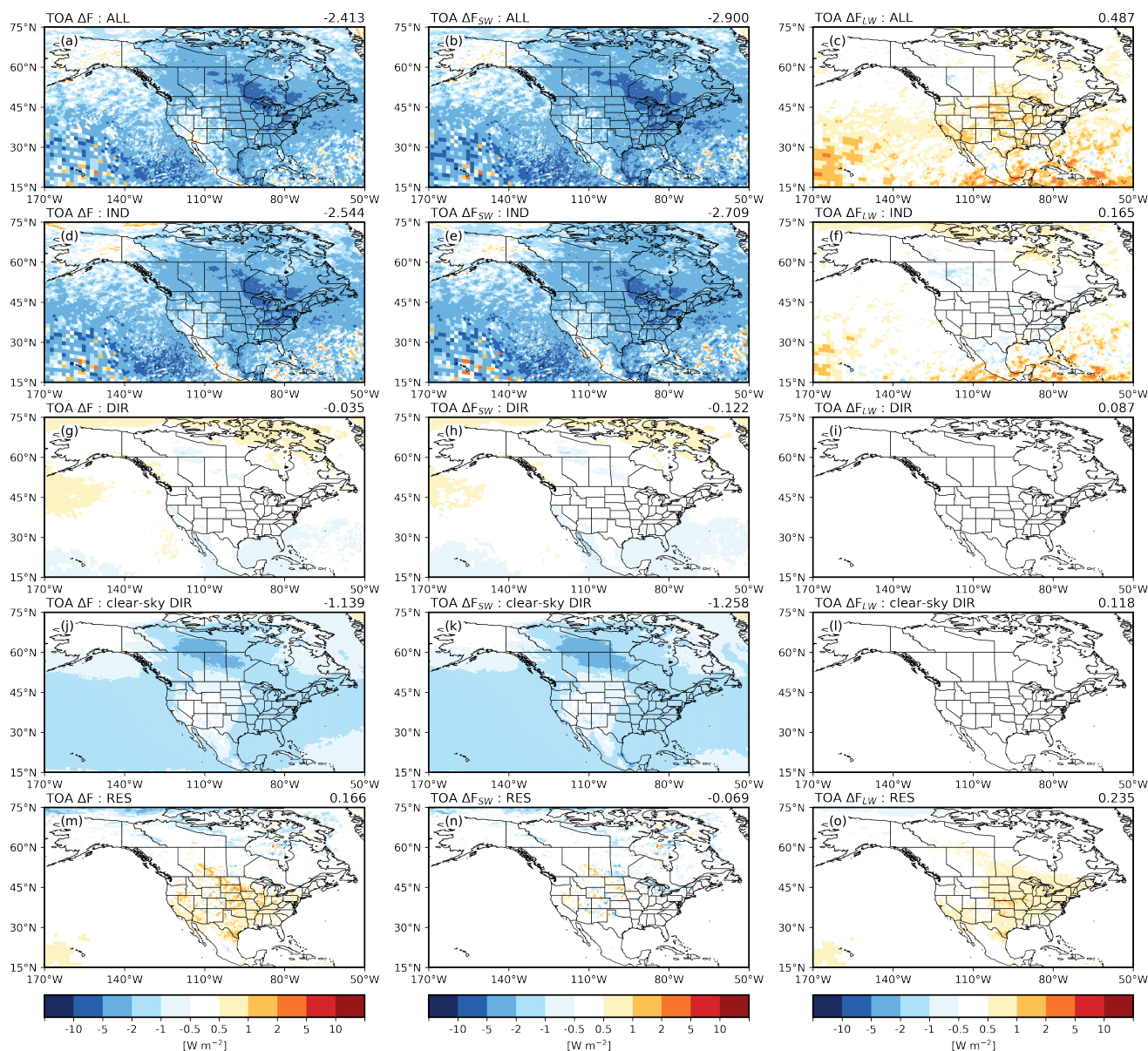
Figure S12. Spatial distribution of anthropogenic aerosol radiative forcing (ΔF) over North America at the top of atmosphere (TOA) from the simulations using the default emission treatment. Decomposed net (left column), shortwave (middle column), and longwave (right column) ΔF are calculated over North 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. Spatial annual mean over NA are indicated at the top right corner of the panels.
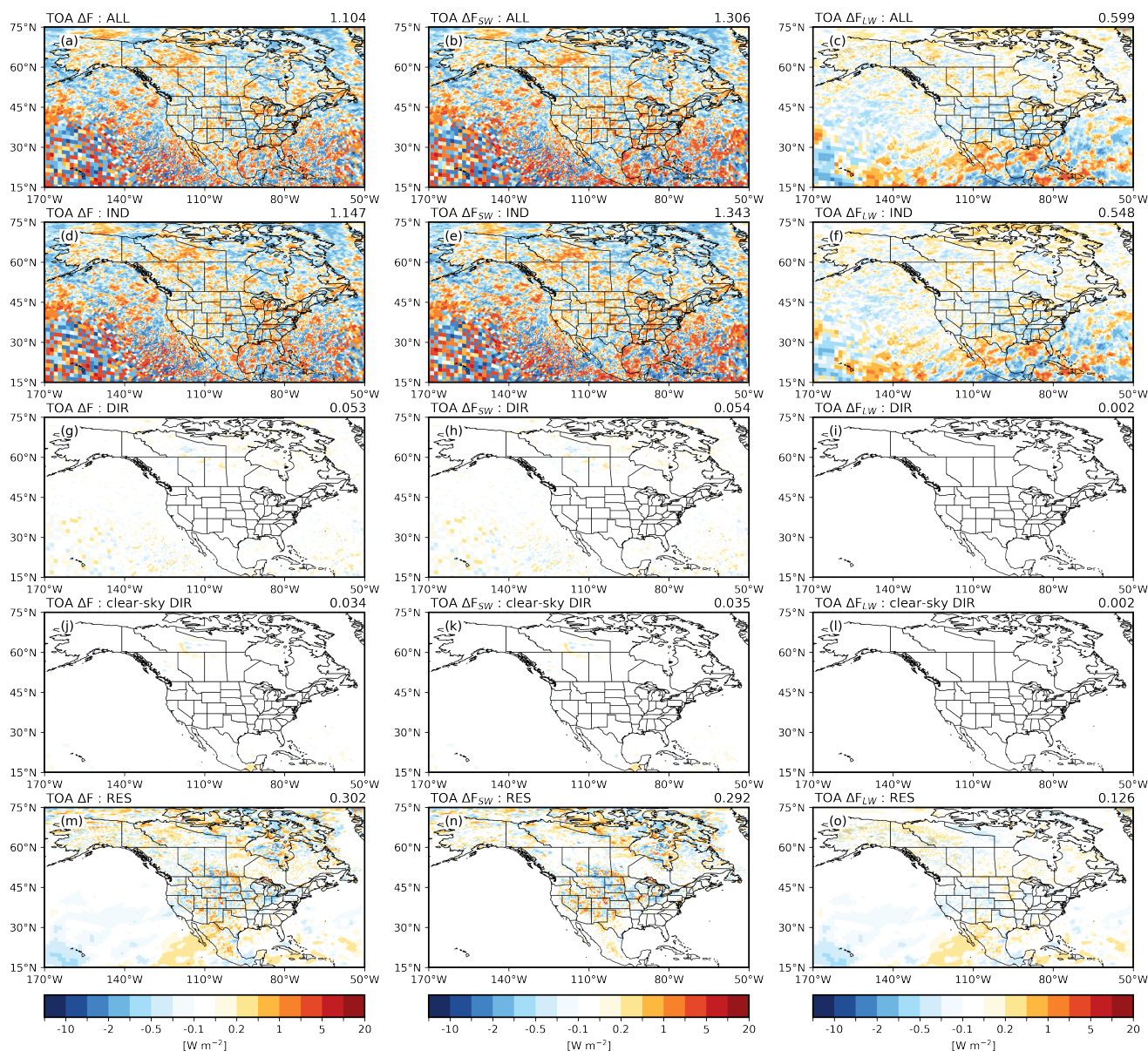
Figure S13. Spatial distribution of anthropogenic aerosol radiative forcing (ΔF) difference over North America at the top of atmosphere (TOA) between simulations with revised and default treatment. Decomposed net (left column), shortwave (middle column), and longwave (right column) ΔF are calculated 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.