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Source Apportionments of Aerosols and Their Direct Radiative Forcing and Long-Term Trends Over Continental United States

Yang Yang[1], Hailong Wang[1], Steven J. Smith[2], Rudong Zhang[1], Sijia Lou[1], Hongbin Yu[3], Can Li[3,4], and Philip J. Rasch[1]

[1]Atmospheric Sciences and Global Change Division, Pacific Northwest National Laboratory, Richland, WA, USA, [2]Joint Global Change Research Institute, Pacific Northwest National Laboratory, College Park, MD, USA, [3]NASA Goddard Space Flight Center, Greenbelt, MD, USA, [4]Earth System Science Interdisciplinary Center, University of Maryland, College Park, MD, USA

Key Points:

- Increases in emissions from East Asia mitigated the warming effect induced by reductions in U.S. emissions by 25% in western United States
- Changes in non-U.S. emissions have a modest impact on U.S. air quality improvement
- Foreign emissions may become increasingly important to radiative forcing over the United States

Supporting Information:

- Supporting Information S1

Correspondence to:
Y. Yang and H. Wang,
yang.yang@pnnl.gov;
hailong.wang@pnnl.gov

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Abstract Due to U.S. air pollution regulations, aerosol and precursor emissions have decreased during recent decades, while changes in emissions in other regions of the world also influence U.S. aerosol trends through long-range transport. We examine here the relative roles of these domestic and foreign emission changes on aerosol concentrations and direct radiative forcing at the top of the atmosphere over the continental United States. Long-term (1980–2014) trends and aerosol source apportionment are quantified in this study using a global aerosol-climate model equipped with an explicit aerosol source tagging technique. Due to U.S. emission control policies, the annual mean near-surface concentration of particles, consisting of sulfate, black carbon, and primary organic aerosol, decreases by about −1.1 (±0.1)/−1.4 (±0.1) $\mu g/m^3$ in western United States and −3.3 (±0.2)/−2.9 (±0.2) $\mu g/m^3$ in eastern United States during 2010–2014, as compared to those in 1980–1984. Meanwhile, decreases in U.S. emissions lead to a warming of +0.48 (±0.03)/+0.46 (±0.03) W/m^2 in western United States and +1.41 (±0.07)/+1.32 (±0.09) W/m^2 in eastern United States through changes in aerosol direct radiative forcing. Increases in emissions from East Asia generally have a modest impact on U.S. air quality but mitigated the warming effect induced by reductions in U.S. emissions by 25% in western United States and 7% in eastern United States. As U.S. domestic aerosol and precursor emissions continue to decrease, foreign emissions may become increasingly important to radiative forcing over the United States.

1. Introduction

Sulfate, black carbon (BC), and organic aerosols (OA) are important constituents of air pollution. They affect air quality and human health, cause morbidity and premature mortality, reduce visibility, and contribute to the ecosystem acidification (Dockery et al., 1993; Driscoll et al., 2001; Fajersztajn et al., 2013). Aerosols also substantially perturb the Earth's radiation balance directly through scattering and absorbing incoming solar radiation and indirectly through modifying cloud microphysical properties. Since industrialization, anthropogenic aerosols have led to a global mean negative direct radiative forcing (DRF) of -0.35 ± 0.50 W/m^2, which has dampened the warming effect from increased greenhouse gas concentrations (Boucher et al., 2013). Understanding aerosols and their radiative impacts is essential for both climate mitigation and improving regional air quality (Shindell et al., 2012).

Air quality is affected not only by local emissions but also by long-range transport of pollutants from distant source regions (Zhang et al., 2017). Quantifying regional source attribution for aerosol species and their radiative influences is, therefore, of interest from both scientific and policy perspectives (Yu et al., 2012). Recent studies have examined the contribution to North American aerosols from distant source regions, especially from East Asia (Chin et al., 2007; Huang et al., 2013; Jacob et al., 2003; Jaffe et al., 2003; Liu et al., 2009; Stjern et al., 2016; Wang et al., 2014; Yang et al., 2015; Yu et al., 2013). Averaged over North America, about one third of the near-surface sulfate concentration comes from distant source emissions for 2010–2014 (Yang, Wang, Smith, Easter, et al., 2017). Sources from East Asia account for 30–50% of the total nonlocal near-surface sulfate concentrations over western United States and 10–30% over eastern United States (Heald et al., 2006; Liu et al., 2008; Park et al., 2004). In western United States, in contrast, only 8% of the near-surface BC concentration is due to emissions from China (Yang, Wang, Smith, Ma, & Rasch, 2017).

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In the upper troposphere distant sources are an even more important contributor to aerosol concentrations (Hadley et al., 2007; Zhang et al., 2015), resulting in large nonlocal source contributions to column burden, aerosol optical depth (AOD), and DRF. Over the west coast of North America, the contribution of emissions from East Asia to the BC column burden can be up to 40% (Wang et al., 2014).

As a result of air pollution regulations, U.S. aerosol and precursor emissions have decreased during recent decades (Smith et al., 2011). Aerosol concentrations and AOD in the United States, especially in eastern United States, have decreased since the inception of U.S. emission control programs (Chin et al., 2014; Zhang et al., 2014). Decreased AOD was also linked to increasing surface solar radiation (aka *surface brightening*, Wild, 2012). In the meantime, developing countries (e.g., in East Asia and South Asia) have significantly increased their emissions due to the rapid economic growth and accelerated urbanization (Streets et al., 2000). However, the relative roles that U.S. and non-U.S. sources played in changing aerosol concentrations and DRF over the United States during the recent decades are still not clear, which are important for the prediction of future air quality and climate change.

In this study, we quantitatively examine the source attributions of aerosol concentrations, DRF at the top of the atmosphere (TOA), and their long-term trends over northwestern, southwestern, northeastern, and southeastern United States during 1980–2014 and estimate the relative contributions of U.S. and non-U.S. source emissions using a global aerosol-climate model equipped with an aerosol tagging technique.

2. Materials and Methods

2.1. Model Description and Experimental Design

The Community Atmosphere Model version 5 (CAM5), which has the ability to simulate sulfate, BC, primary organic aerosol (POA), and secondary organic aerosol (SOA) with a three-mode modal aerosol module (MAM3, Liu et al., 2012), is used to simulate the long-term variation of aerosols and their DRF at the TOA. In CAM5-MAM3, aerosols are internally mixed in the same aerosol mode and then externally mixed between modes. BC is mixed with other aerosol species (e.g., sulfate, POA, SOA, sea salt, and dust) in the accumulation mode immediately after emission into the atmosphere neglecting aging processes. A set of model improvements is also included to better represent the convective transport and wet scavenging of aerosols (Wang et al., 2013). In this study, the model is configured at a horizontal grid of 1.9° latitude × 2.5° longitude with 30 vertical layers up to 3.6 hPa. A 36-year simulation (1979–2014) driven by time-varying insolation, surface conditions, greenhouse gases, and aerosol and precursor emissions has been conducted. The model version here is identical to that used in recent work (Yang, Wang, Smith, Ma, & Rasch, 2017; Yang, Wang, Smith, Easter, et al., 2017). This coarse-resolution climate model is configured for studying long-term continent-scale mean air quality and climatic effect of aerosols rather than finer-scale air quality effects. Monthly aerosol and precursor emissions, including anthropogenic (Hoesly et al., 2018; version 20160726) and open biomass burning (van Marle et al., 2017; version 20161213) emissions, are obtained from the CMIP6 (Coupled Model Intercomparison Project Phase 6) data sets. Seasonally varying biogenic emissions are calculated offline by the MEGAN2.1 model. Atmospheric circulation patterns are important in driving the transport of aerosols (Ma et al., 2013). To minimize the impact of potential model biases on the aerosol transport, we nudge the model wind fields toward the MERRA (Modern Era Retrospective-Analysis for Research and Applications) reanalysis (Rienecker et al., 2011). The first year is for spin-up, and the remaining 35-year results are used in our analysis. In this study, we mainly focus on sulfate, BC, and POA aerosols and their DRF. DRF for any tagged aerosol is calculated as the difference in clear-sky shortwave radiation fluxes at the TOA using a radiation diagnostic package in CAM5, where the radiative transfer model is run twice, one with the specific tagged aerosol included and the other having the tagged aerosol excluded from the aerosol mixture.

2.2. Aerosol Source Tagging in CAM5

To quantify the source-receptor relationship of aerosols and their DRF over the continental United States, here we employ an explicit aerosol source tagging technique. Emission, evolution, transport, and removal of aerosols including sulfate, BC, and POA from independent sources are calculated separately for each tagged region or sector. As described in Wang et al. (2014), this aerosol tagging method explicitly tracks precursor gases and aerosols emitted from predefined source regions/sectors using additional model variables within a single model simulation. The additional variables or tags undergo the same physical, chemical, and dynamical processes as the corresponding original ones. The simulation here is the first time that the three

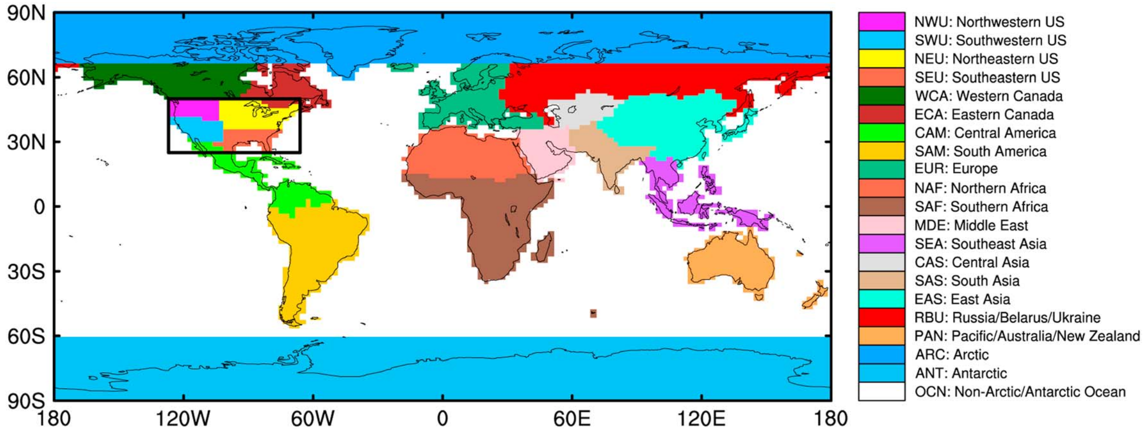

Figure 1. Tagged source regions, including northwestern, southwestern, northeastern and southeastern United States (NWU, SWU, NEU, and SEU), western and eastern Canada (WCA and ECA), Central America (CAM), Europe (EUR), the Middle East (MDE), South Asia (SAS), East Asia (EAS), Russia/Belarus/Ukraine (RBU, hereafter Russia), South America (SAM), North Africa (NAF), South Africa (SAF), Southeast Asia (SEA), Central Asia (CAS), Pacific/Australia/New Zealand (PAN), the Arctic (ARC), Antarctic (ANT), and Non-Arctic/Antarctic Ocean (OCN).

major anthropogenic species (i.e., sulfate, BC, and POA) are simultaneously tracked using source tagging in an aerosol-climate model. BC and sulfate were tagged separately in previous work, focusing on different source and receptor regions (Yang, Wang, Smith, Ma, & Rasch, 2017; Yang, Wang, Smith, Easter, et al., 2017). Based on these individual implementations, source tagging for BC, sulfate, and POA was merged into the model version used here to quantify source attributions of the different types of aerosols and their DRF over the United States on a multidecadal time scale. In this study, emissions from 21 individual source regions are tagged. They are northwestern, southwestern, northeastern, and southeastern United States (NWU, SWU, NEU, and SEU), western and eastern Canada (WCA and ECA), Central America (CAM), Europe (EUR), the Middle East (MDE), South Asia (SAS), East Asia (EAS), Russia/Belarus/Ukraine (RBU, hereafter Russia), South America (SAM), North Africa (NAF), South Africa (SAF), Southeast Asia (SEA), Central Asia (CAS), Pacific/Australia/New Zealand (PAN), the Arctic (ARC), Antarctic (ANT), and Non-Arctic/Antarctic Ocean (OCN), as shown in Figure 1. Natural sources of sulfate, oceanic dimethyl sulfide and volcanic eruptions, are tagged together and included in the OCN source.

2.3. Observational Data

The model is evaluated over the United States by comparing simulated near-surface aerosol concentrations with observations from the IMPROVE (Interagency Monitoring of Protected Visual Environments) (http://vista.cira.colostate.edu/IMPROVE/) network (Malm et al., 1994) and AOD with that from MODIS (Moderate Resolution Imaging Spectroradiometer, https://modis.gsfc.nasa.gov) Deep Blue retrieval (Hsu et al., 2013) and AERONET (Aerosol Robotic Network, https://aeronet.gsfc.nasa.gov) measurements (Holben et al., 1998). U.S. EPA (United States Environment Protection Agency) SO_2 emissions at state level (U.S. Environment Protection Agency, 2016) are used to evaluate the CMIP6 SO_2 emission inventory.

As compared with the IMPROVE data, the model reproduces well the spatial distribution of mean near-surface sulfate concentrations over 2005–2014, with low values in western United States (northwestern + southwestern) and high values in eastern United States (northeastern + southeastern; Figure 2). However, the model overestimates sulfate concentrations by 60–67% in western United States, resulting from a bias in the emission distribution, which was discovered as these results were being analyzed. We find that the CMIP6 SO_2 emissions are much higher in western United States and slightly lower in eastern United States (Figure S3b in the supporting information) as compared to U.S. EPA estimates. This spatial bias is due to the global proxy data sets used to spatially distribute U.S. national emission totals in the CMIP6 emissions data set which likely do not capture lower SO_2 emission intensities in western United States as compared to the east.

Using the tagged model results, we have also estimated sulfate concentrations scaled to match EPA estimated emissions for each U.S. subregion as described in the supporting information (Figures S7 and S8).

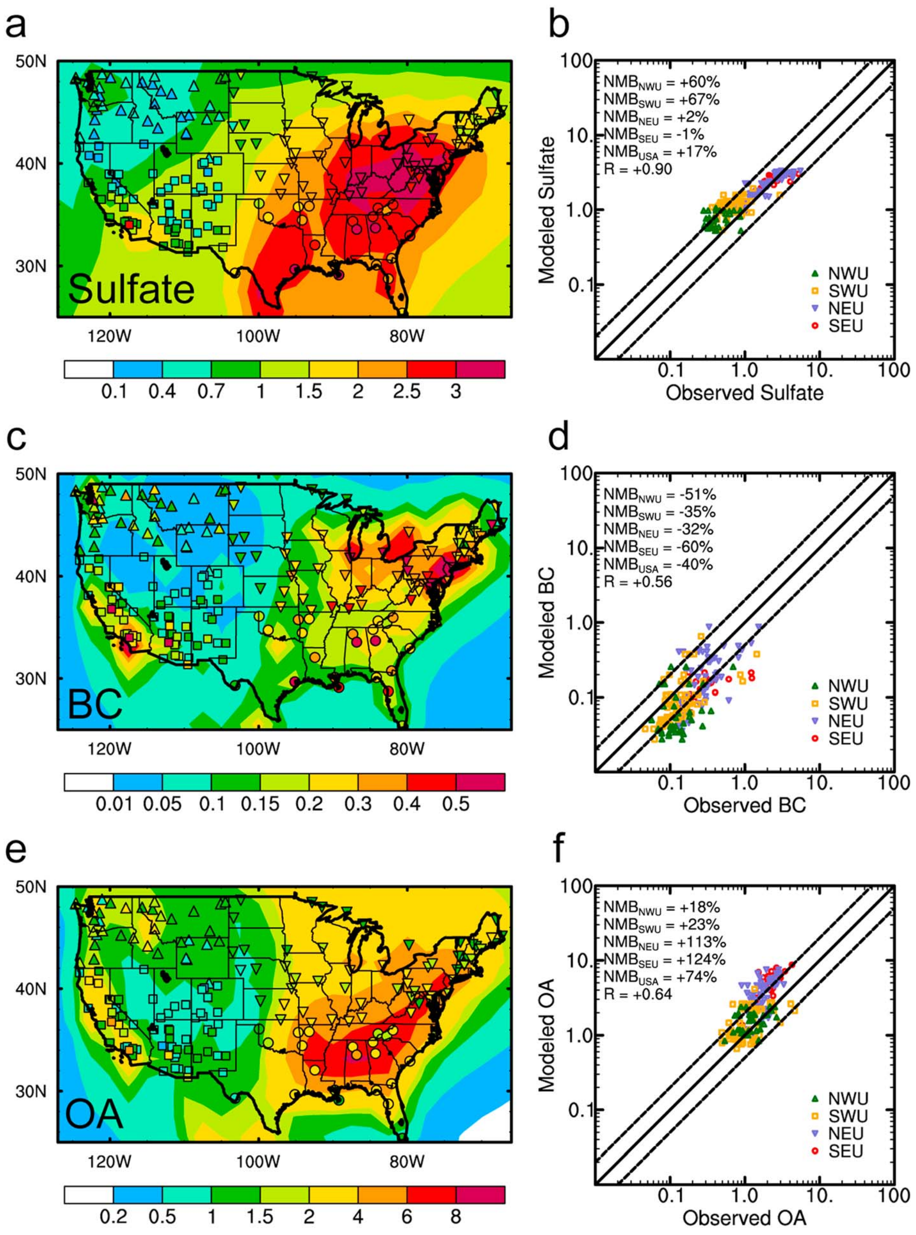

Figure 2. Spatial distribution (left panels) and scatter plots (right panels) between the simulated and observed annual mean near-surface (a, b) sulfate, (c, d) black carbon (BC), and (e, f) organic aerosol (OA, primary OA + second OA) concentrations ($\mu g/m^3$) averaged over years 2005–2014. Observations are from Interagency Monitoring of Protected Visual Environments networks. Solid lines mark the 1:1 ratio, and dashed lines mark the 1:2 and 2:1 ratio. Normalized mean bias (NMB) and correlation coefficient (*R*) between observation and simulation are shown on the right panel.
NMB = 100 % $\times \sum (M_i - O_i)/ \sum O_i$, where M_i and O_i are the modeled and observed values at site *i*, respectively.
NWU = northwestern United States; SWU = southwestern United States; NEU = northeastern United States;
SEU = southeastern United States.

The scaled sulfate surface concentration results match observations much more closely, with an average positive bias of 10% in western United States. This emissions data bias has the largest impact on results from western United States and scaled results for this region will be used below as appropriate.

BC concentrations are underestimated by 40% in the entire United States (32–60% in the subregions), due to either an overly strong wet removal of BC associated with the lack of aging process or the substantial uncertainties in BC emissions (Bond et al., 2013). The model strongly overestimates OA concentrations (by a factor of 2) over eastern United States, mainly contributed by high modeled SOA in summer (Liu et al., 2012).

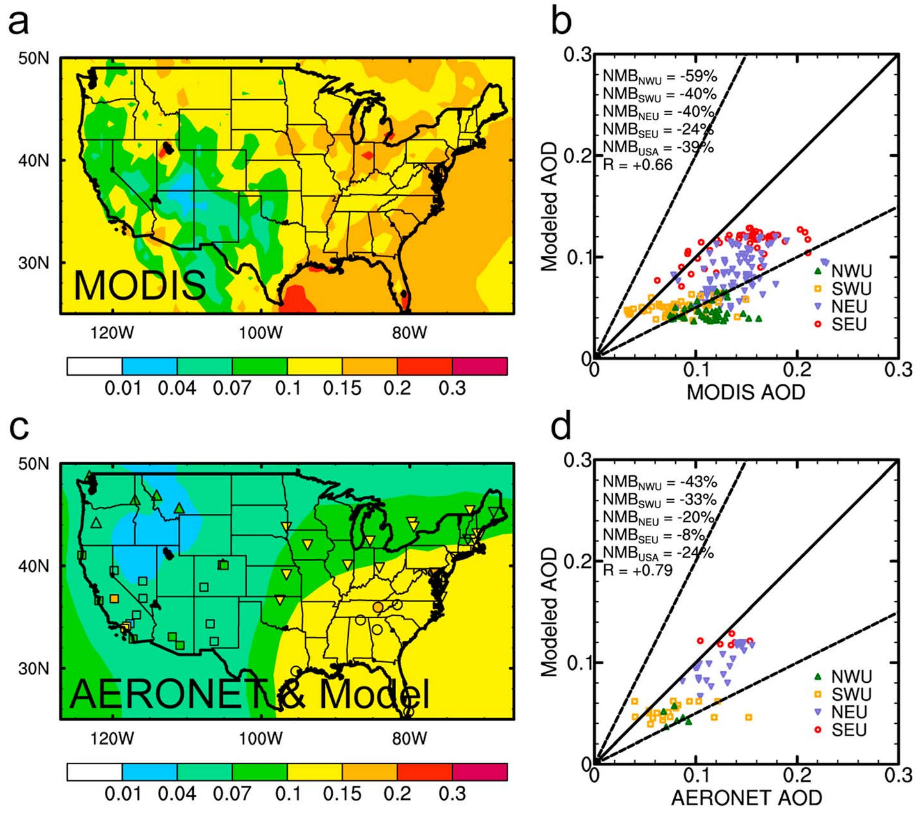

Figure 3. Spatial distribution of (a) Moderate Resolution Imaging Spectroradiometer (MODIS) aerosol optical depth (AOD) and (b) simulated together with Aerosol Robotic Network (AERONET) AOD, as well as (c, d) scatter plots between the simulated and observed values averaged over years 2005–2014. Solid lines mark the 1:1 ratio, and dashed lines mark the 1:2 and 2:1 ratios. Normalized mean bias (NMB) and correlation coefficient (*R*) between observation and simulation are shown on the right panel. NMB = 100 % $\times \sum(M_i - O_i)/\sum O_i$, where M_i and O_i are the modeled and observed values at site *i*, respectively. NWU = northwestern United States; SWU = southwestern United States; NEU = northeastern United States; SEU = southeastern United States.

Compared to the MODIS and AERONET data, the model underestimates the mean AOD in the United States by 24–39%, especially in western United States (Figure 3). The low bias in AOD likely comes from missing nitrate and ammonium aerosols in the model, which are particularly important in west and midwest agricultural regions of the United States (NARSTO, 2004). Underestimation of dust and/or POA from open burning could also contribute to bias. We will further discuss the biases and their possible impacts on results in the following section.

3. Source Attribution of Aerosols in the United States

Figures 4a and 4b show the spatial distributions of annual mean total near-surface sulfate-BC-POA concentrations attributed to U.S. and non-U.S. emissions, respectively, averaged over 2005–2014. Sulfate-BC-POA concentrations are dominated by U.S. domestic emissions in both western and eastern United States. In northwestern and southwestern United States, domestic emissions contribute 0.5–2.0 μg/m^3 to sulfate-BC-POA concentrations. Contributions from non-U.S. emissions are 0.2–0.5 μg/m^3, 20–40% of the total. Contributions by domestic emissions to sulfate-BC-POA concentrations in eastern United States are in the range of 1–5 μg/m^3, higher than those in western United States due to higher local emissions. Non-U.S. emissions account for 0.5–1 μg/m^3 of the mean concentration in eastern United States, which is about twice as much in absolute values as in western United States, but only 10–30% of the total. In contrast, total TOA sulfate-BC-POA DRF in western United States has similar contributions, in the range of −0.1 to −0.4 W/m^2, from both U.S. and non-U.S. emissions (see Figures 4c and 4d). In eastern United States, domestic emissions are responsible for −0.2 to −1.0 W/m^2 of the total DRF, larger than the foreign contributions of −0.1 to

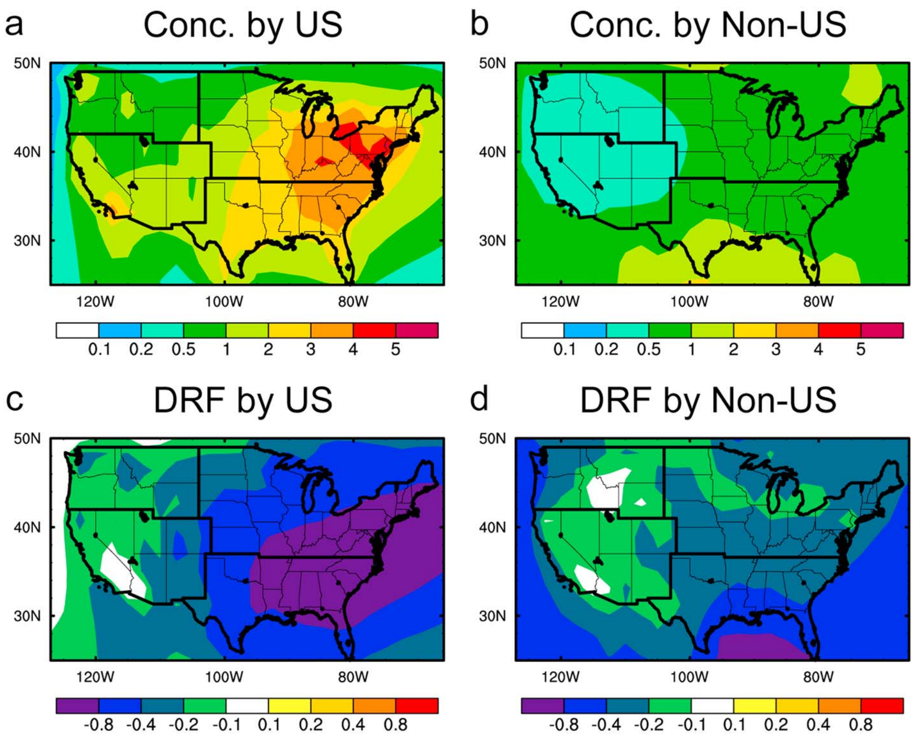

Figure 4. Annual mean (a, b) near-surface concentrations (μg/m^3) of sulfate-black carbon-primary organic aerosol and (c, d) their total direct radiative forcing (DRF) (W/m^2) contributed by U.S. and non-U.S. emissions, averaged over 2005–2014.

−0.4 W/m^2. These indicate that although U.S. domestic emissions dominate aerosol concentrations and DRF in the United States, emissions beyond the domestic domain can be important, especially to aerosol DRF.

Figure 5 presents the relative contributions of emissions from major source regions to near-surface concentrations and column burdens of aerosols in the United States. (Table S1 summarizes these values, and Figure S1 shows the absolute contributions.) In northwestern United States, 36% and 15% (21% and 6% for scaled results, see below) of the near-surface sulfate concentration comes from local and southwestern U.S. emissions, followed by 15% (22%) from East Asia and 9% (13%) from Canada (WCA + ECA). In southwestern United States, local emissions account for 48% (19%) of sulfate concentrations, while East Asia contributes 12% (19%). The percentages in parentheses indicate estimates scaled to account for the spatial bias in the emissions data set (see supporting information), which point to a larger influence from non-U.S. sources.

In northeastern United States, local emissions and emissions from southeastern United States explain 59% and 14%, respectively, of the sulfate concentration. In southeastern United States, the near-surface sulfate is mainly attributed to local emissions (43%) and emissions from northeastern United States (20%). Emissions from other regions, especially from shipping and natural emissions near the coasts, contribute 11–13% of the near-surface sulfate over northwestern, southwestern, and southeastern United States. BC and POA have larger local contributions to their near-surface concentrations (63–84%), because wet scavenging by precipitation more efficiently removes these primary aerosols as compared to gaseous SO_2 (precursor gas of sulfate aerosol), making distant sources less efficient. Emissions from Canada (WCA + ECA) account for 13–22% of the near-surface POA in northwestern and northeastern United States. Emissions from Central America contribute 11–15% to the sulfate, BC, and POA in southeastern United States. Averaged over 2005–2014, sources within the United States contribute 56–80% of the near-surface sulfate and 73–92% of the BC and POA in the United States. These results suggest that air quality in a specific region of the United States is largely controlled by local emissions with contributions from other U.S. source regions. Canada and Central America emissions can also have an impact through transboundary transport. East Asia emissions influence air quality in western United States through long-range transpacific transport.

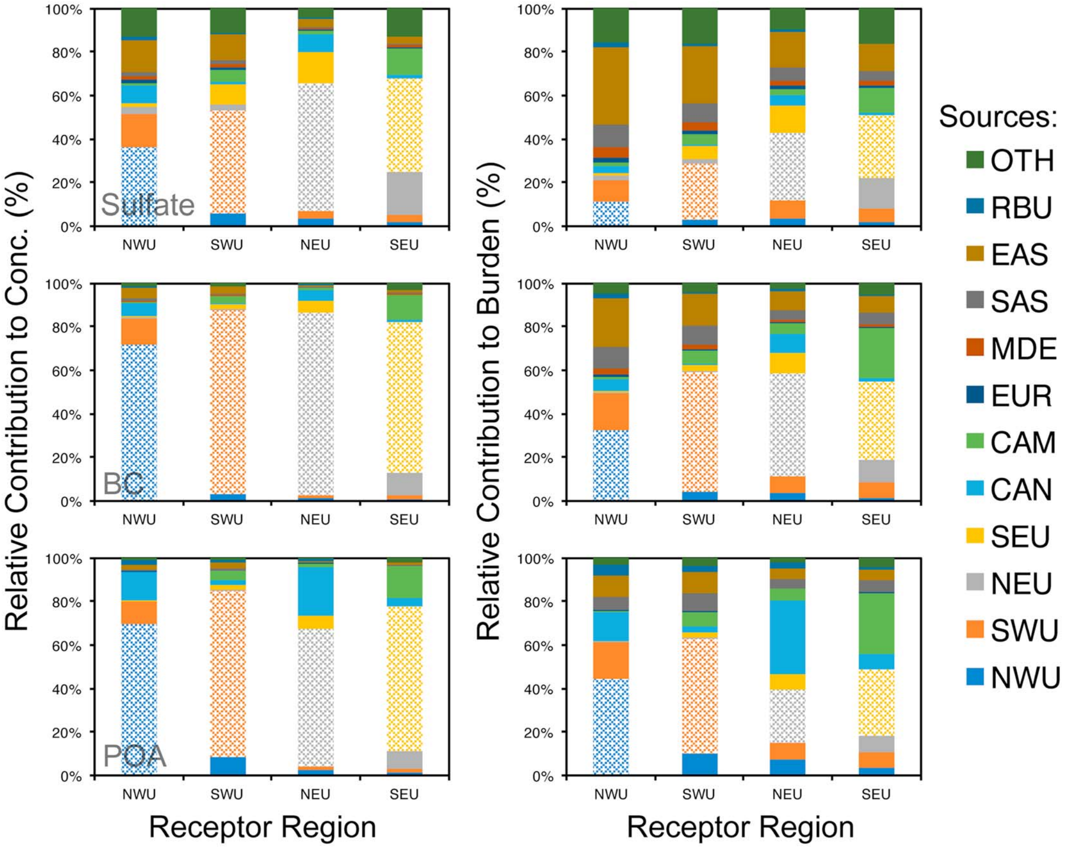

Figure 5. Relative contributions (%) to near-surface concentrations (left panels) and column burdens (right panels) of sulfate (top row), black carbon (BC; middle row), and primary organic aerosol (POA; bottom row) averaged over northwestern, southwestern, northeastern, and southeastern United States from emission of the major tagged source regions (NWU, SWU, NEU, SEU, CAN (western and eastern Canada), CAM, EUR, MDE, SAS, EAS, and RBU) and other regions (OTH, including SAM, NAF, SAF, SEA, CAS, PAN, ARC, ANT, and OCN), averaged over years 2005–2014. Patterned bars represent local emission contributions. OTH = other regions; RBU = Russia/Belarus/Ukraine; EAS = East Asia; SAS = South Asia; MDE = Middle East; EUR = Europe; CAM = Central America; CAN = Canada; NWU = northwestern United States; SWU = southwestern United States; NEU = northeastern United States; SEU = southeastern United States.

The long-range transport of aerosols is more efficient in the free atmosphere, especially in the upper troposphere, where there are stronger winds, lower temperatures, and less precipitation (hence longer lifetime), than in the boundary layer. This explains the larger relative contributions from nonlocal emissions to column burdens of aerosols than to the near-surface concentrations (Table S1). Because of this, the fractional contribution of local sources to column burden is smaller than their fractional contribution to near-surface concentration for all three types of aerosol. In northwestern United States, only 10% of the column burden of sulfate is from local sources, whereas East Asia and South Asia are responsible for 35% and 11% of the sulfate burden, respectively. East Asia accounts for 26%, 17%, and 12% of the sulfate burdens in southwestern, northeastern, and southeastern United States, respectively. Besides sulfate, 15–22% of BC and 10% of the POA burden in western United States comes from East Asia emissions. Canada and Central America also contribute substantially to the BC and POA column burden over eastern United States. Nonlocal contributions are even larger when our estimates are scaled to account for western U.S. SO_2 emissions bias.

Over 2005–2014, the large contributions to the free tropospheric aerosol lead to a stronger DRF from non-U.S. emissions than from U.S. emissions for both sulfate and the sum of BC and POA, especially in western United States (Figure S2). These suggest a significant role for non-U.S. emissions in multidecadal trends of aerosol, and DRF in particular, as discussed in the next section.

4. Long-Term Trends of Source Contributions

Between 1980 and 2014, due to air pollution regulations, SO_2 emissions from the continental United States decreased by 80% (from 12.3 to 2.5 Tg S/year), while BC and POA emissions were estimated to have

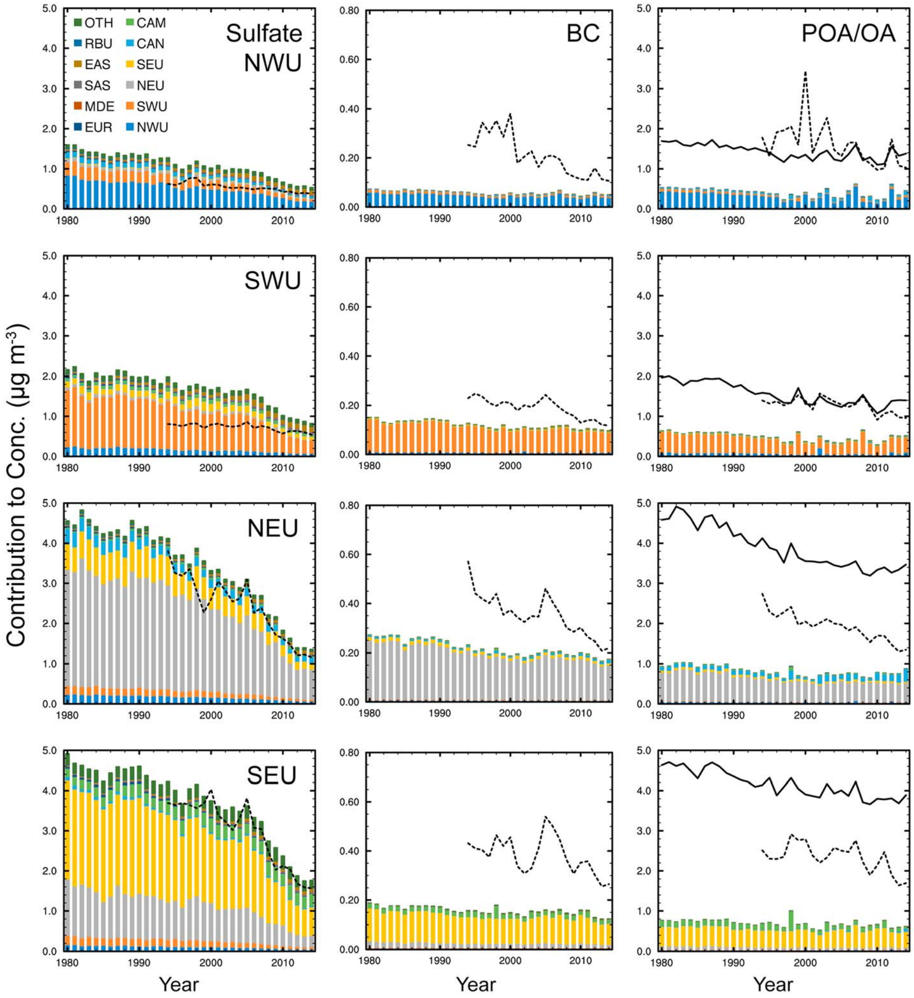

Figure 6. Time series (1980–2014) of simulated annual mean near-surface sulfate (left column), black carbon (BC; middle column), and primary organic aerosol (POA; right column) concentrations (μg/m^3) averaged over northwestern, southwestern, northeastern, and southeastern United States (from top to bottom) contributed by emissions from major source emissions (bars). Dashed lines represent sulfate, BC and OA (POA + SOA) concentrations observed from the IMPROVE networks over 1994–2014. Solid lines in right panels represent simulated OA concentrations for comparison. OTH = other regions; RBU = Russia/Belarus/Ukraine; EAS = East Asia; SAS = South Asia; MDE = Middle East; EUR = Europe; CAM = Central America; CAN = Canada; NWU = northwestern United States; SWU = southwestern United States; NEU = northeastern United States; SEU = southeastern United States.

decreased by about 40% (from 0.39 to 0.23 Tg C/year) and 30% (from 1.40 to 0.99 Tg C/year), respectively. All four U.S. subregions show decreasing trends of aerosol and precursor emissions during this time period (Figure S3), especially eastern United States. Over the same time, emissions from Europe and Russia also decreased, whereas East Asia and South Asia experienced increases (Figure S4). These changes in U.S. and non-U.S. emissions have led to changes in the source contributions of aerosol concentrations, column burden, and DRF in the United States.

Figure 6 shows time series (1980–2014) of the near-surface aerosol concentrations over the four subregions of the United States contributed by major source regions. Near-surface sulfate concentrations decreased substantially over time in all four subregions of the United States, especially in eastern United States where sulfate concentrations decreased from 4–5 to 1–2 μg/m^3. U.S. BC and POA concentrations are also decreasing, but at a slower rate than sulfate. These aerosol concentration decreases are mostly due to decreases in local emissions, with contributions also from emission decreases in other U.S. source regions (e.g., southeastern U.S. influence on sulfate in northeastern United States). Total near-surface sulfate-BC-POA concentrations (Figure S5), used here to represent the aerosol impact on air quality, also indicate that local emissions and

Table 1
Differences in Contributions From U.S., non-U.S., and East Asia Emissions to Near-Surface Concentrations and DRF at the TOA Over Four Subregions of the United States Between 1980–1984 and 2010–2014

	NWU			SWU		
	Sulfate	BC + POA	All	Sulfate	BC + POA	All
Differences in contribution to concentrations (μg/m^3)						
U.S.	−0.87 ± 0.07	−0.20 ± 0.09	−1.07 ± 0.11	−1.22 ± 0.11	−0.22 ± 0.07	−1.44 ± 0.11
Non-U.S.	−0.04 ± 0.02	0.04 ± 0.02	−0.01 ± 0.03	0.06 ± 0.02	0.02 ± 0.00	0.08 ± 0.02
EAS	0.07 ± 0.01	0.00 ± 0.00	0.07 ± 0.01	0.08 ± 0.01	0.01 ± 0.00	0.09 ± 0.01
Difference in contribution to DRF (W/m^2)						
U.S.	0.48 ± 0.05	0.00 ± 0.03	0.48 ± 0.03	0.53 ± 0.05	−0.06 ± 0.03	0.46 ± 0.03
Non-U.S.	−0.12 ± 0.02	0.01 ± 0.05	−0.10 ± 0.04	−0.16 ± 0.02	0.04 ± 0.04	−0.12 ± 0.04
EAS	−0.16 ± 0.01	0.03 ± 0.01	−0.13 ± 0.01	−0.13 ± 0.01	0.03 ± 0.00	−0.10 ± 0.01
	NEU			**SEU**		
	Sulfate	BC + POA	All	Sulfate	BC + POA	All
Differences in contribution to concentrations (μg/m^3)						
U.S.	−2.86 ± 0.18	−0.41 ± 0.04	−3.28 ± 0.21	−2.76 ± 0.20	−0.14 ± 0.05	−2.90 ± 0.21
Non-U.S.	−0.18 ± 0.02	0.12 ± 0.03	−0.06 ± 0.05	0.08 ± 0.05	−0.02 ± 0.03	0.07 ± 0.07
EAS	0.04 ± 0.01	0.00 ± 0.00	0.05 ± 0.01	0.05 ± 0.01	0.00 ± 0.00	0.06 ± 0.01
Difference in contribution to DRF (W/m^2)						
U.S.	1.52 ± 0.08	−0.11 ± 0.02	1.41 ± 0.07	1.39 ± 0.10	−0.07 ± 0.02	1.32 ± 0.09
Non-U.S.	−0.06 ± 0.02	0.05 ± 0.02	−0.02 ± 0.02	−0.17 ± 0.03	0.04 ± 0.03	−0.13 ± 0.04
EAS	−0.12 ± 0.01	0.03 ± 0.00	−0.09 ± 0.01	−0.11 ± 0.01	0.02 ± 0.00	−0.09 ± 0.01

Note. Standard deviation for the interannual temporal variation is also provided. EAS = East Asia; NWU = northwestern United States; SWU = southwestern United States; DRF = direct radiative forcing; NEU = northeastern United States; SEU = southeastern United States; BC = black carbon; POA = primary organic aerosol.

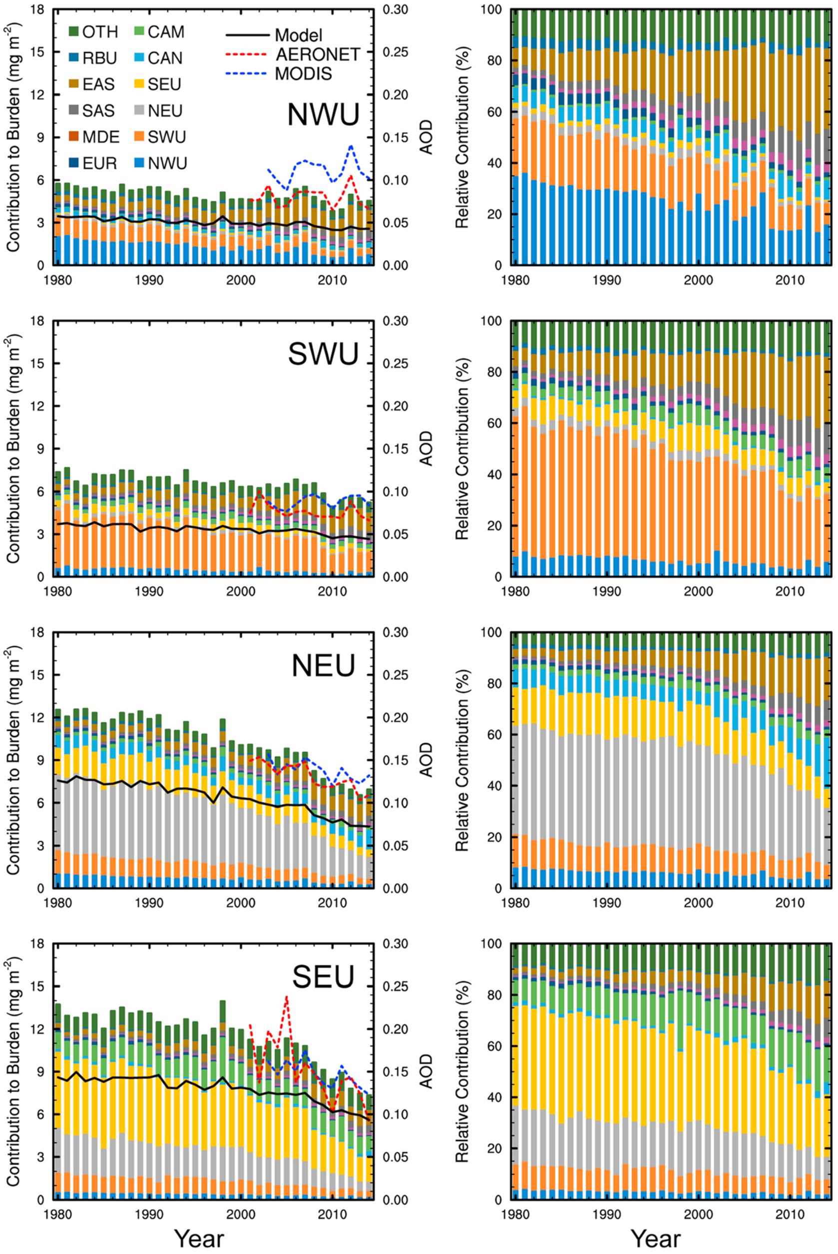

Figure 7. Time series (1980–2014) of simulated annual mean column burdens of sulfate-black carbon-primary organic aerosol averaged over northwestern, southwestern, northeastern, and southeastern United States (from top to bottom) contributed by emissions from major source emissions (mg/m^2, bars in left panels) as well as their relative contributions (%, right panels). Solid and dashed lines represent simulated and observed aerosol optical depth (AOD) from Aerosol Robotic Network (AERONET) networks over 2001–2014 (red) and Moderate Resolution Imaging Spectroradiometer (MODIS) data (blue) over 2003–2014. OTH = other regions; RBU = Russia/Belarus/Ukraine; EAS = East Asia; SAS = South Asia; MDE = Middle East; EUR = Europe; CAM = Central America; CAN = Canada; NWU = northwestern United States; SWU = southwestern United States; NEU = northeastern United States; SEU = southeastern United States.

emissions from other source regions within the United States dominate the long-term variation of air quality in the United States. However, due to the lower contributions from U.S. domestic sources, emissions from East Asia have become more important to U.S. air quality in more recent years.

Compared to 1980–1984, the mean near-surface sulfate in 2010–2014 over northwestern and southwestern United States are lower by −0.87 (±0.07) and −1.22 (±0.11) μg/m^3, respectively, due to U.S. emission control

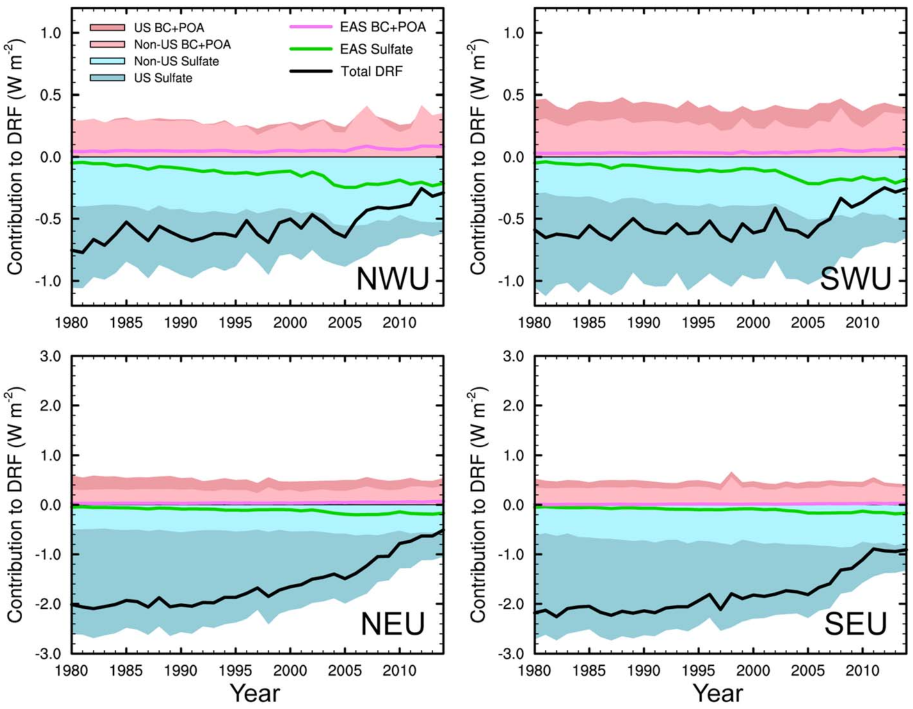

Figure 8. Stacked area chart for the time series (1980–2014) of simulated annual mean direct radiative forcing (DRF; W/m^2) of black carbon (BC) + primary organic aerosol (POA) and sulfate contributed by U.S. (dark pink and dark blue area) and non-U.S. sources (light pink and light blue area) over the four subregions of the United States. Purple and green lines represent DRF of BC + POA and sulfate, respectively, contributed by emissions from East Asia for comparison with non-U.S. contribution. Black lines represent the net DRF, considering both BC + POA and sulfate aerosols as well as sources from both U.S. and non-U.S. source emissions. NWU = northwestern United States; SWU = southwestern United States; NEU = northeastern United States; SEU = southeastern United States.

programs, whereas the changes in contributions from non-U.S. emissions are −0.04 (±0.02) and 0.06 (±0.02) μg/m^3 (Table 1). East Asia contributes 0.07 (±0.01) and 0.08 (±0.01) μg/m^3 more to the sulfate concentrations in northwestern and southwestern United States in 2010–2014 than in 1980–1984, respectively, which was offset by decreases in contributions from Canada, Europe, and Russia (Table S2). In northeastern and southeastern United States, the near-surface sulfate contributed by U.S. emissions decreases from 1980–1984 to 2010–2014 by −2.86 (±0.18) and −2.76 (±0.20) μg/m^3, respectively, while Canada, Central America, Europe, and East Asia also exert a small contribution to the total differences. With BC and POA included, the total near-surface sulfate-BC-POA concentrations decrease by −1.08 (±0.12)/−1.36 (±0.12) μg/m^3 in northwestern/southwestern United States and −3.34 (±0.20)/−2.83 (±0.25) μg/m^3 in northeastern/southeastern United States. The increases in East Asia emissions offset the decrease in sulfate-BC-POA concentration, caused by the domestic emission reductions, by 6–7% in western United States and 2% in eastern United States. As a result, reductions in U.S. domestic aerosol and precursor emissions have efficiently decreased near-surface concentrations of aerosols and improved air quality in the United States. Although non-U.S. emissions can influence decadal changes in surface aerosol concentrations, their influence is much smaller than that of U.S. domestic emission changes over this period.

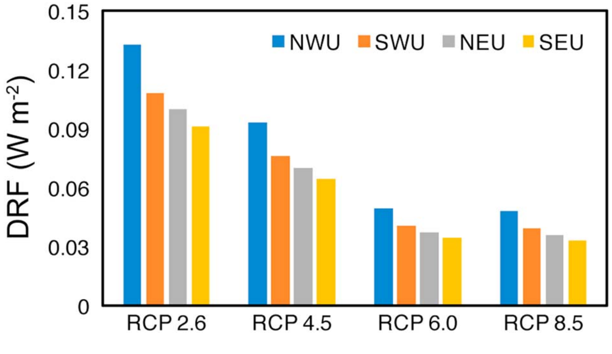

Figure 9. Sulfate direct radiative forcing (DRF) in the United States contributed by emissions from East Asia in 2030, relative to 2010, under the different Representative Concentration Pathways (RCPs). The DRF here is calculated by scaling the forcing contributed by East Asian SO_2 emissions (see Table 1) with the ratio of historical changes (comparing 2010–2014 to 1980–1984) and future changes (comparing 2030 to 2010) in East Asian SO_2 emissions. The future changes are calculated based on the 2010 Coupled Model Intercomparison Project Phase 6 East Asian SO_2 emissions and the percentage reduction in SO_2 emissions in East Asia under each of the RCP scenarios (−51.3%, −36.0%, −19.2%, and −18.5% for RCP 2.6, RCP 4.5, RCP 6.0, and RCP 8.5, respectively). NWU = northwestern United States; SWU = southwestern United States; NEU = northeastern United States; SEU = southeastern United States.

Column burdens of sulfate-BC-POA in western United States do not show the same rate of decrease as the near-surface concentration (from 5–8 mg/m^2 in 1980 to 4–6 mg/m^2 in 2014, for a 28% net decrease as compared to the 70% decrease of surface concentration; Figure 7). Although the column burden from domestic emissions decreases during this time period, non-U.S. contributions (e.g., from East Asia and South Asia)

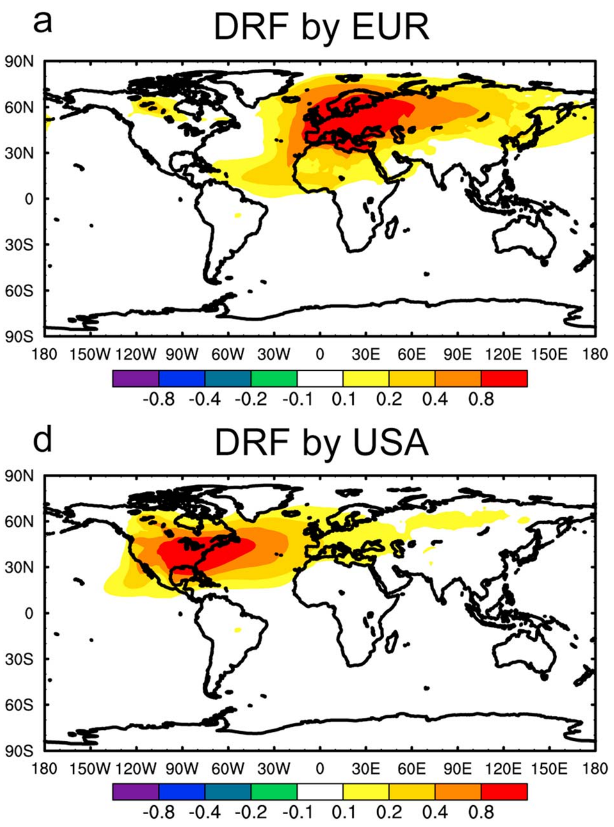

Figure 10. Changes in annual mean sulfate direct radiative forcing (DRF; W/m^2) contributed by emissions from (a) Europe (EUR) and (b) United States between 1980–1984 and 2010–2014.

increased, which partly offset the domestic decreases. Considering the model overestimates the domestic decrease due to bias in western U.S. SO_2 emissions, the non-U.S. contributions are even more important to the column burden and forcing trends than indicated here. In eastern United States, due to the strong reduction in local emissions, the column burden decreases more significantly, from 12–14 mg/m^2 in 1980 to 7–8 mg/m^2 in 2014. Sources from East Asia have become increasingly important to the column burden of aerosols and thus the aerosol DRF.

Between 1980 and 2014, the negative sulfate DRF at the TOA in United States from domestic emissions decreased substantially (Figure 8), which, all else being equal, would have contributed to a regional warming. However, the increased column burden by non-U.S. emissions had an opposite impact (i.e., strengthening the negative sulfate DRF). The DRF of BC and POA together do not have a strong trend compared to that of sulfate, in part due to the cancellation between the warming effect from BC and cooling effect from POA and the lower rate of decrease in domestic emissions of BC and POA (30–40%) compared to that of SO_2 (80%). Note that, because BC and POA are sometimes co-emitted, for the computational efficiency, we did not separate the DRF between BC and POA in this study. Adding the opposite trends in DRF from U.S. and non-U.S. emissions, the net effect is a slight decrease in the magnitude of DRF in both western and eastern United States. The increases in the contribution from East Asia are similar to those from all non-U.S. emissions, suggesting that East Asia dominates the trend of non-U.S. contributions to the DRF over the United States during 1980–2014.

Decreases in U.S. emissions between 1980–1984 and 2010–2014 lead to a warming effect of +0.48 (±0.03)/+0.46 (±0.03) W/m^2 in northwestern/southwestern United States and + 1.41 (±0.07)/+1.32 (±0.09) W/m^2 in northeastern/southeastern United States (Table 1). However, the warming effect is partly offset by a cooling effect from changes in non-U.S. emissions. The increase in emissions from East Asia leads to a DRF of −0.13 (±0.01)/−0.10 (±0.01) W/m^2 in northwestern/southwestern United States and −0.09 (±0.01)/−0.09 (±0.01) W/m^2 in northeastern/southeastern United States, mitigating the warming effect from decreases in U.S. emissions by 22–27% in western United States and 6–7% in eastern United States. Given the local SO_2 overestimate in western United States, we conclude that most of the cooling from U.S. SO_2 reductions in western United States may have been offset by increased contributions from Asian emissions (supporting information). Although increase in emissions from South Asia and decrease from Europe also lead to changes in DRF in the United States (Table S3), their contributions are small compared to that from East Asia.

5. Conclusions and Discussion

This is the first study, to our knowledge, that examines the net effects and relative roles of multidecadal U.S. pollution-control programs and changes in emissions in other regions of the world on aerosol concentrations and TOA DRF over the continental United States for the three major anthropogenic species (i.e., sulfate, BC, and POA) in a global aerosol-climate model. We found that, during 1980–2014, reductions in U.S. domestic aerosol and precursor emissions have efficiently decreased near-surface concentrations of aerosols and improved air quality in the United States. While, non-U.S. emissions generally have a modest impact on surface concentrations, we estimate that 15–20% of the improvement in surface sulfate concentrations in western United States has been offset by increasing contributions from East Asia (supporting information).

East Asia has a much larger contribution to aerosols in the free troposphere over the United States, where they have offset much of the warming forcing in western United States that would otherwise have been associated with reductions in U.S. domestic emissions. As U.S. domestic emissions continue to decrease due to current polices (Shi et al., 2017), the relative impact of foreign emissions may well continue to increase. It is, therefore, important for future projections to consider the potential impact of foreign emissions on radiative forcing, but also on surface concentrations, particularly in western United States.

There are a few sources of uncertainties in these results. As discussed above and in the supporting information the overestimation of western U.S. SO_2 emissions in the data used here means that the influence of SO_2 transported from Asia is relatively more important than indicated in our model results.

The model underestimates near-surface BC over the entire continental United States, probably due to an overly strong wet removal of BC associated with the lack of aging process and/or uncertainties in BC emissions. For OA, the model reproduces well the near-surface concentrations in western United States but strongly overestimates its concentration in eastern United States, especially in summer (Figure S9), probably due to large uncertainties in modeled SOA (Liu et al., 2012). Although the model has biases in the magnitude of total sulfate-BC-OA concentrations, it simulates fairly well the decreasing trends in all four subregions of the United States (Figures S5 and S7). Due to large uncertainties in the simulation of SOA in global models as well as the consideration of computational efficiency, only the near-surface sulfate-BC-POA concentration is used to represent air quality in this study. Transport efficiency, wet and dry removal, and oxidation efficiency for aerosols are parameterized in global models, which can also lead to biases.

Recent studies indicate that the transpacific transport of sulfate from East Asia contributes to 30–50% of the total nonlocal near-surface sulfate concentrations in western United States and 10–30% in eastern United States (Heald et al., 2006; Liu et al., 2008; Park et al., 2004), which are similar to the percentages in our study of 34% and 10–18% in western and eastern United States respectively, averaged over 2010–2014. In western United States, 15–22% of the column burden of BC is due to emissions from East Asia, also in accord with 10–30% in previous studies (Wang et al., 2014).

The influence of SOA, which was not diagnosed here, could change the net DRF of BC plus OA as compared to BC plus POA forcing considered here. However, this study focused on decadal-scale changes. Decreasing sulfate dominates AOD changes during 1980–2014 in the United States, while POA and SOA only have very small AOD changes (Figure S6). Therefore, including SOA in the DRF calculation for carbonaceous aerosols would not substantially change the overall results, which are driven largely by decreases in sulfate. Note that a large amount of SOA precursor gases come from natural emissions, which do not vary annually in these simulations, leading to small interannual variability of SOA in the model.

Compared to AERONET measurements and MODIS satellite data, the model underestimates AOD in all four subregions of the United States (Figure 7). The model simulation does not include nitrate and ammonium aerosols, which are particularly important in west and midwest agricultural regions of the United States, which will lead to a low bias in simulated particulate matter concentration and AOD. There is possibly even an increase in U.S. ammonia emissions (Warner et al., 2017). Due to regulatory controls on the emissions of gas-phase precursors, nitrate has been reported to decrease since 2000 in California (Pusede et al., 2016). As a scattering aerosol, reductions in nitrate aerosol would produce additional positive DRF. Therefore, missing nitrate in the model may have caused an underestimation of the magnitude of decreasing trends in total aerosol concentration and local warming, which, however, is not expected to change our conclusion that increases in emissions from East Asia partly mitigated the warming effect induced by reductions in U.S. emissions in western United States. The possible underestimation of dust and/or POA from open burning could also contribute to the AOD bias. The successful simulation of trends in the near-surface concentrations and AOD in the model provides a foundation for source attributions of trends in aerosols and their DRF. However, the low bias (by a factor of 2) in model simulated sulfate concentrations over East Asia (Yang, Wang, Smith, Easter, et al., 2017), along with the indication of relatively low contribution by East Asia to the nonlocal sulfate concentrations in western United States (34% compared to 30–50% from previous studies discussed above), can lead to a low bias in the contributions by East Asia to AOD and DRF in western United States. Given also the overestimation of SO_2 emissions in western United States in CMIP6 emissions data set, the cooling effect of changes in non-U.S. emissions on radiation may have overwhelmed warming effects over western United States due to U.S. emission decreases. In addition, the coarse-resolution model simulation may not capture aerosol concentrations where small-scale topography is complex (Yang et al., 2018), but this is less likely to influence results at the continent-scale level (Liu et al., 2016; Ma et al., 2015).

In this study, only three aerosol modes were used in the aerosol module. All species in the same mode therefore have the same mean diameter and standard deviation. However, aerosols from Asian outflow reaching

United States are more aged than local freshly emitted particles. The size of these aged aerosols can be quite different from freshly emitted aerosols. The difference in aerosol size can lead to different effects in light scattering and absorption and thus affect the source attribution of radiative forcing, which warrants further studies with more sophisticated aerosol representations.

In the MAM3 aerosol representation of CAM5, BC aging process is neglected by assuming the immediate internal mixing of BC with other aerosol species in the accumulation mode. This assumed internal mixing for BC tends to produce stronger light absorption or higher positive forcing than external mixing (Jacobson, 2001). Thus, the internal mixing assumption may cause an overestimation of the positive DRF of BC where BC and other species might actually be externally mixed. This assumption could also lead to an overestimation of wet removal of BC and an underestimation of BC concentrations (Figures 2c and 2d), which might cancel out part of the enhanced absorption by internal mixing.

While the work here focused on the aerosol direct effect, the indirect effect of aerosols on clouds also impacts regional energy balance. Based on simulations from a recent study (Yang, Wang, Smith, Easter, et al., 2017) with 20% sulfur emission reductions over North America and globally, North America emissions only account for about one third of the sulfate indirect radiative forcing (IRF) over the United States for 2010–2014, while nonlocal emissions contribute the remaining two thirds. It suggests a strong contribution to IRF over the United States and trends since 1980 by non-U.S. emissions, which would contribute to an even larger cooling trend due to non-U.S. emissions over recent decades.

Due to the U.S. emission control programs, SO_2 emissions have decreased dramatically during the recent decades. Total SO_2 emission, from U.S. EPA estimates, decreased from 0.63 Tg S/year in 1990 to 0.14 Tg S/year in 2016 over western United States and from 10.88 to 1.18 Tg S/year in the same time period over eastern United States (Figure S3). There is limited room to further reduce U.S. SO_2 emissions in the future, and, therefore, a limited extent to which radiative forcing over the United States will be enhanced by future reductions in U.S. domestic SO_2 emissions. Due to increasing health concerns, China has enacted policies to reduce SO_2 emissions (Lu et al., 2010), which have at least stabilized and may have begun to decrease. Under the Representative Concentration Pathways (RCP) scenarios (van Vuuren et al., 2011), anthropogenic SO_2 emissions from East Asia are projected to decrease by about 20% (RCP 6.0 and RCP 8.5) to 50% (RCP 2.6) by 2030 relative to the year-2010 level. Based on the CMIP6 SO_2 emissions in 2010 and the relative reductions among the RCP scenarios for 2010–2030, we can estimate East Asian SO_2 emissions in 2030 under a range of future emission pathways. Reductions in SO_2 emissions from East Asia are projected to produce a positive direct sulfate forcing over the United States between 2010 and 2030 (Figure 9). Under the RCP 2.6, East Asia SO_2 emission reductions lead to a 0.09–0.13 W/m^2 positive forcing warming in the United States, decreasing to 0.03–0.05 W/m^2 under the RCP 6.0 and 8.5 scenarios.

Averaged over 2010–2014, sulfate and BC (Yang et al., 2018) produce DRF of -1.04 and 0.47 W/m^2, respectively, over the United States. A simultaneous reduction in light-absorbing BC aerosol would mitigate a portion of the positive forcing from projected future reductions in SO_2 emissions in the United States and East Asia.

We have focused on the effect of non-U.S. emissions on decadal changes in DRF over the United States. A decrease in U.S. emissions will also contribute to local warming in other regions. In Europe, for example, sulfate DRF has decreased substantially, leading to a warming effect over Europe (Figure 10a) of 1.64 (±0.07) W/m^2 due to local SO_2 emission reductions in 2010–2014 as compared to 1980–1984. Meanwhile, the reductions in U.S. SO_2 emissions over this period results in an additional forcing of 0.16 (±0.01) W/m^2 over Europe, which enhances forcing by 10% (Figure 10b).

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