https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020

Atmospheric Chemistry and Physics Discussions

Source attribution of Arctic aerosols and associated Arctic warming trend during 1980–2018

Lili Ren[1], Yang Yang[1*], Hailong Wang[2], Rudong Zhang[2], Pinya Wang[1], Hong Liao[1]

[1]Jiangsu Key Laboratory of Atmospheric Environment Monitoring and Pollution Control, Jiangsu Collaborative Innovation Center of Atmospheric Environment and Equipment Technology, School of Environmental Science and Engineering, Nanjing University of Information Science and Technology, Nanjing, Jiangsu, China

[2]Atmospheric Sciences and Global Change Division, Pacific Northwest National Laboratory, Richland, Washington, USA

*Correspondence to yang.yang@nuist.edu.cn

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020

Atmospheric Chemistry and Physics Discussions

Abstract

Observations show that the concentrations of Arctic sulfate and black carbon (BC) aerosols have declined since the early 1980s, which potentially contributed to the recent rapid Arctic warming. In this study, a global aerosol-climate model equipped with an Explicit Aerosol Source Tagging (CAM5-EAST) is applied to quantify the source apportionment of aerosols in the Arctic from sixteen source regions and the role of aerosol variations in the Arctic surface temperature change over the past four decades (1980–2018). The CAM5-EAST simulated surface concentrations of sulfate and BC in the Arctic had a decrease of 43% and 23%, respectively, in 2014–2018 relative to 1980–1984, mainly due to the reduction of emissions from Europe, Russia and Arctic local sources. Increases in emissions from South and East Asia led to positive trends of Arctic sulfate and BC in the upper troposphere. Changes in radiative forcing of sulfate and BC through aerosol-radiation interactions are found to exert a +0.145 K Arctic surface warming during 2014–2018 with respect to 1980–1984, with the largest contribution (61%) by sulfate decrease, especially originating from the mid-latitude regions. The changes in atmospheric BC outside the Arctic produced an Arctic warming of +0.062 K, partially offset by –0.005 K of cooling due to atmospheric BC within the Arctic and –0.041 K related to the weakened snow/ice albedo effect of BC. Through aerosol-cloud interactions, the sulfate reduction gave an Arctic warming of +0.193 K between the first and last five years of 1980–2018, the majority of which is due to the mid-latitude emission change. Our results suggest that changes in aerosols over the mid-latitudes of the Northern Hemisphere have a larger impact on Arctic temperature than other regions

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020


Atmospheric Chemistry and Physics Discussions

43 associated with enhanced poleward heat transport from the aerosol-induced stronger
44 meridional temperature gradient. The combined aerosol effects of sulfate and BC
45 together produce an Arctic surface warming of +0.297 K during 1980–2018, explaining
46 approximately 20% of the observed Arctic warming during the same time period.
47

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020


Atmospheric Chemistry and Physics Discussions

1. Introduction

The Arctic has warmed rapidly since the 1980s, with a 1.5 K increase in the surface air temperature, which is about two to four times faster than the global average (Trenberth et al., 2007; Serreze et al., 2009). The significant rise in air and ground temperatures occurred in phase with dramatic melting of Arctic sea ice and snow, potentially contributing to Arctic amplification (Pithan and Mauritsen, 2014; Zhang et al., 2019). A number of studies have examined possible mechanisms that caused the rapid Arctic warming (Graversen et al., 2008; Screen and Ian, 2010; Screen and Simmonds, 2010; Alexeev et al., 2012; Zhang et al., 2018). Observations and modeling studies suggest that, although anthropogenic long-lived greenhouse gases (GHGs) dominate the radiative forcing of the climate system, variations in black carbon (BC) aerosol and other short-lived air pollutants are a good explanation for the faster Arctic warming (Law and Andreas, 2007; Quinn et al., 2008; Shindell et al., 2008). In particular, Shindell and Faluvegi (2009) indicated that Arctic warming was influenced by the changing aerosol concentrations in the Arctic over the last three decades based on model sensitivity experiments. The aerosols that caused Arctic warming are not only from local emissions. Previous studies have shown that changes in long-range transport of sulfate and BC aerosols from mid-latitude regions have caused strong wintertime warming in the Arctic (e.g., Breider et al., 2014; Fisher et al., 2011; Shindell et al., 2008). In addition, the mid-latitude aerosols can influence Arctic climate through changing poleward heat transport (Navarro et al., 2016).

Observed and modeled seasonal cycles of aerosol concentrations at the remote

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020


Atmospheric Chemistry and Physics Discussions

Arctic surface show a maximum in winter, a phenomenon commonly known as Arctic Haze, and a minimum in summer (Law and Andreas, 2007;Quinn et al., 2007; Eckhardt et al., 2015; Garrett et al., 2010; Sharma et al., 2006). The winter maximum has been attributed to the long-range transport of anthropogenic pollution from the mid-latitudes of the Northern Hemisphere and weak removal in the Arctic (Stohl, 2006; Wang et al., 2014). In contrast, summer aerosol concentrations in the Arctic atmosphere reach a minimum value due to a reduced poleward aerosol transport from the mid-latitudes and efficient wet scavenging processes during the transport (Bourgeois and Bey, 2011; Browse et al., 2012; Garrett et al., 2011). Anthropogenic aerosol species (e.g., sulfate, BC and organic matter) can affect Arctic climate by disturbing the energy balance of the earth system (Yang et al., 2019a). Sulfate aerosols directly scatter solar radiation and indirectly influence cloud processes by serving as cloud condensation nuclei (Yang et al., 2017a; Zamora et al., 2017; Zhao and Garrett, 2015). BC absorbs solar radiation and warms the atmosphere (Bond et al., 2013; Yang et al., 2017b; Lou et al., 2019a), which can increase or decrease cloud cover depending on the vertical distribution of BC relative to clouds (e.g., McFarquhar and Wang, 2006; Lou et al., 2019b). When it deposits on snow and ice, BC can reduce surface albedo and accelerate snow melt (Flanner et al., 2007; Qian et al., 2015). Breider et al. (2017) estimated the aerosol radiative forcing due to aerosol-radiation interactions in the Arctic and found that, averaged over 2005–2010, the top-of-the-atmosphere (TOA) forcing is -0.60 ± 0.02Wm^{-2} for sulfate and +0.44 ± 0.04 Wm^{-2} for BC over the Arctic.

Analysis of long-term changes in sulfate and BC can help to gain a comprehensive

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020


Atmospheric Chemistry and Physics Discussions

92 understanding of their past and present impacts on the Arctic climate. In situ
93 observations of sulfate and BC concentrations in the Arctic (e.g., at Alert, Barrow,
94 Station Nord, and Zeppelin) have shown a declining trend since the 1980s (Gong et al.,
95 2010; Heidam et al., 1999; Hirdman et al., 2010; Quinn et al., 2009; Sharma et al., 2004;
96 Sharma et al., 2006; Sinha et al., 2017; Sirois and Barrie, 1999). Based on the chemical
97 transport model GEOS-Chem simulations, Breider et al. (2017) found that annual
98 sulfate and BC concentrations decreased by 2–3% per year over the Arctic. McConnell
99 et al. (2007) presented a historical BC trend derived from ice-core records, showing
100 that BC concentration had been declining steadily after the peak around 1910.

101 Source attribution analysis of atmospheric aerosols in the Arctic, which can help
102 to understand aerosol trends, is extremely important for air pollution research. There is
103 less local anthropogenic aerosol emission in the Arctic region than in polluted regions
104 of the world. Pollutants in the Arctic are generally from mid-latitude areas by long-
105 distance transport (Fisher et al., 2011; Wang et al., 2014). Recent studies have found
106 that Arctic aerosols mainly originated from Eurasia, Southeast Asia, Siberia and North
107 America (Fisher et al., 2011; Qi et al., 2017; Sharma et al., 2013; Stohl, 2006). The
108 contribution of Eurasia to Arctic sulfate and BC aerosols concentration is dominant in
109 the lower atmosphere, while South and Central Asia contributed the most at high
110 altitudes (e.g., Wang et al., 2014). In general, Northern Europe and Russia, with large
111 industrial emissions, are the main source region of Arctic BC aerosols in spring (Rahn
112 et al., 1977; Rahn, 1981; Raatz and Shaw, 1984; Barrie, 1986; Koch and Hansen, 2005;
113 Sharma et al., 2006; Stohl, 2006). In the past few decades, anthropogenic emissions

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020


Atmospheric Chemistry and Physics Discussions

have changed rapidly, with a decrease in Europe and North America and an increase in South and East Asia. This may have had an important impact on the Arctic aerosols and climate (Breider et al., 2014).

In this study, the global aerosol-climate model CAM5 (Community Atmosphere Model, version 5) equipped with an Explicit Aerosol Source Tagging (CAM5-EAST) is used to examine the attribution of Arctic aerosols to 16 different source regions and the aerosol-related Arctic warming during 1980–2018. We focus on changes in sulfate and BC near-surface concentrations, total column burden, and radiative forcing as well as their impacts on the surface temperature over the Arctic. Modeled and observational sulfate and BC concentrations at remote Arctic stations are compared. CAM5-EAST tagging results are used to quantify the contributions of different sources to the decadal changes in Arctic sulfate and BC surface concentrations and vertical profiles. Based on the Arctic climate sensitivity factors, we estimate the responses of the Arctic surface temperature to the variations in sulfate and BC during the analyzed time periods.

2. Methodology

2.1 Model Description and Experimental Setup

The global aerosol-climate model CAM5, which is the atmospheric component of the earth system model CESM (Community Earth System Model) developed at the National Center for Atmospheric Research (NCAR), is used to simulate Arctic aerosols and climate for years 1980–2018 (after one-year model spin-up). In this model version, mass concentrations of sulfate, BC, primary organic aerosol (POA), second organic aerosol (SOA), mineral dust, and sea salt are predicted with a three-mode modal aerosol

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020


Atmospheric Chemistry and Physics Discussions

module. Aerosol number concentration is also predicted for each mode. Particles in the different size modes are assumed to be externally mixed and internally mixed in the same mode. The optical properties and radiative impact of aerosols are calculated online. The model also includes climate effects of aerosols through aerosol-radiation and aerosol-cloud interactions. In this study, the model is configured to run at a horizontal grid of 1.9° latitude × 2.5° longitude with 30 vertical layers up to 3.6 hPa.

The CAM5 simulation is conducted with prescribed time-varying solar radiation, sea surface temperature, sea ice concentration, GHGs, and emissions of aerosols and their precursor gases. In order to better reproduce the aerosol transport driven by large-scale circulations in the model, the wind field is nudged toward the MERRA-2 (Modern Era Retrospective-Analysis for Research and Applications, Version 2) reanalysis (Rienecker et al., 2011; Gelaro et al., 2017) at a 6-hourly relaxation timscale. Radiative forcing due to aerosol-radiation interactions is calculated as the difference of clear-sky net radiative fluxes at TOA between two separate diagnostic calculations, including and excluding a specific aerosol in the radiative transfer calculation, respectively (Ghan et al., 2012).

2.2 Explicit Aerosol Source Tagging and Source Regions

The Explicit Aerosol Source Tagging (EAST) was implemented in CAM5 to quantify the source-receptor relationships of aerosols in recent studies (Wang et al., 2014;Yang et al., 2017a,b; 2018a,b,c). All physical, chemical and dynamical processes of aerosols for each tagged source region or sector are considered independently and consistently by using additional sets of aerosol variables in CAM5-EAST, which is different from

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020


Atmospheric Chemistry and Physics Discussions

158 the widely used emission sensitivity method that assumes a linear response to emission
159 perturbation or the indirect method of tracing long-lived constituents associated with
160 particular sources. Without such assumption of linear response or constant decaying
161 rate, EAST is more physically accurate than the source attribution methods mentioned
162 above. In this study, sulfate and BC are explicitly tracked throughout the processes from
163 source emissions to deposition in a single model simulation.

164 We focus on the Arctic (66.5°N–90°N) as the receptor region in this study. According
165 to source region definition of the Hemispheric Transport of Air Pollution model
166 experiment phase 2 (HTAP2), sulfate and BC from 16 regions are tagged (Fig. 1):
167 Europe (EUR), North America (NAM), Central America (CAM), South America
168 (SAM), North Africa (NAF), South Africa (SAF), the Middle East (MDE), Southeast
169 Asia (SEA), Central Asia (CAS), South Asia (SAS), East Asia (EAS), Russia-Belarus-
170 Ukraine (RBU, hereafter Russia), Pacific-Australia-New Zealand (PAN), the Arctic
171 (ARC), Antarctic (ANT), and Non-Arctic/Antarctic Ocean (OCN). Note that the OCN
172 tag includes sources from oceans and volcanic eruptions.

173 ## 2.3 Aerosol and Precursor Emissions

174 In order to simulate the long-term temporal variations in aerosols, historical
175 anthropogenic (Hoesly et al., 2018) and biomass combustion (van Marle et al., 2017)
176 emissions of aerosols and precursor gases during 1980–2014 are used in the simulation
177 following the CMIP6 (Coupled Model Intercomparison Project Phase 6) protocol. For
178 the most recent years (2015-2018), yearly interpolated emissions from the SSP2-4.5
179 scenario, which is the modest scenario compared to other SSPs and is widely utilized

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020


Atmospheric Chemistry and Physics Discussions

in many MIPs (O'Neill et al., 2016) are used. Figures 1 and 2 show the spatial distribution and time series of annual anthropogenic SO_2 and BC emissions, respectively, during 1980–2018, from the 16 source regions. The global total anthropogenic SO_2 and BC emission rates, averaged over 1980–2018, are 118.4 Tg yr^{-1} and 8.1 Tg yr^{-1}, respectively. SO_2 emissions are relatively high in East Asia (23.6 Tg yr^{-1}), Europe (15.8 Tg yr^{-1}) and North America (15.4 Tg yr^{-1}), while BC emissions show a high mean value in East Asia (1.8 Tg yr^{-1}), South Africa (1.6 Tg yr^{-1}) and South Asia (0.9 Tg yr^{-1}). Comparing 2014–2018 to 1980–1984, global anthropogenic SO_2 emission was reduced by 32.2 Tg yr^{-1} (24.8% relative to 1980–1984). The largest decreases took place in Europe (83.0%), North America (80.7%) and Russia (74.8%). In East Asia, emissions of anthropogenic SO_2 were increased by a factor of 2.7 from 1980 to 2014, followed by a decreasing trend after 2014 due to stricter air pollution regulations. The global anthropogenic BC emission increased from 6.5 Tg yr^{-1} in 1980 to a peak of 9.6 Tg yr^{-1} in 2014, followed by a slow decline, with an overall increase of 42% between the first and last five years of 1980–2018. Regionally, compared to 1980–1984, averaged BC emissions in 2014–2018 in Europe, Russia and the Arctic decreased by 45.2%, 44.1% and 38.3%, respectively, while BC emissions in East Asia and South Asia almost increased by a factor of 2. Within the Arctic, SO_2 and BC emissions decreased by 5.8% and 38.3%, respectively.

2.4 Model Evaluation

To assess the ability of the model to simulate Arctic sulfate and BC, Figs. 3 and 4 compare simulated near-surface concentrations of sulfate and BC, respectively, in

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020


Atmospheric Chemistry and Physics Discussions

spring and summer during 1980–2018 with observations at five Arctic stations: Alert (82°N, 62°W), Station Nord (81°N,16°W), Barrow (71°N,156°W), Ny-Alesund (78°N,11°E) and Kevo (69°N,27°E). The observations are derived from European Monitoring and Evaluation Programme and World Data Centre for Aerosols database (http://ebas.nilu.no) and Breider et al. (2017).

Overall, the sulfate and BC concentrations in spring is higher than those in summer, mainly due to lower removal rate and more efficient transport (Stohl, 2006). The model underestimates aerosol concentrations in spring, likely due to biases in simulated precipitation and aerosol wet removal during the transport to high latitudes. All sites show that sulfate concentrations decrease during the analyzed time period and BC decreases at specific sites, which can be explained by the reduction of non-local emissions as illustrated by the source attribution. Compared to the observed values, the model can reasonably simulate the time variations of sulfate and BC in the Arctic but the magnitude at some of the sites is largely underestimated. The Kevo site, which is close to Western Eurasia, is the only site that has both sulfate and BC data for more than 30 years. At this site, the simulated sulfate and BC in spring (summer) decreased at a rate of -3.18% (-1.92%) and -2.89% (-1.74%) per year, which is similar to -4.37% (-3.26%) and -3.01% (-2.82%) per year from observations. The Alert site has 33-year sulfate observations, where the simulated sulfate concentrations declined at a rate of -2.08% (-2.00%) per year, consistent with the observed decreasing trends of -2.89% (-0.47%) per year.

Observational data are very limited in the Arctic, especially the long-term

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020


Atmospheric Chemistry and Physics Discussions

observations. The available BC measurements are equivalent elemental carbon (EBC), which is usually obtained by converting the light absorbed by the particles accumulated on the ground instrument filter into the BC concentration. The uncertainty in optical properties of BC makes this conversion challenging. Other light absorbing substances, such as dust and organic carbon, also affect the BC measurements, so EBC would tend to be higher than the actual BC concentration. Researchers found that BC observations could be biased by 30% to 200% (Sharma et al., 2017; Sinha et al., 2017) due to the inclusion of other light absorption components in the atmosphere. Shindell et al. (2008) and Koch et al. (2009) found great differences between the current models and observations of Arctic BC and sulfate through multi-model comparation studies, including incorrect seasonality and order of magnitude biases. Given the large apparent discrepancies in BC for all models, it is difficult to determine the relative authenticity of the models using currently available data (Shindell et al., 2008).

3. Source Apportionment of Aerosols in the Arctic

The near-surface concentrations of sulfate and BC over the Arctic can be quantitatively attributed to both Arctic local emissions and remote sources outside the Arctic through the source tagging in CAM5-EAST. The absolute and relative source contributions are shown in Fig. 5. Arctic local emissions and sources near the Arctic (e.g., Europe and Russia) are the main contributors to the near-surface concentrations of Arctic sulfate and BC. Relative to 1980–1984, the simulated annual sulfate concentration over the Arctic has a decrease of 42.8% in 2014–2018 (Table 1). Sulfate concentration shows a considerable decreasing trend from 1980 to 2000 and then slows

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020


Atmospheric Chemistry and Physics Discussions

down after 2000. The decrease in sulfate during this time period primarily results from the reduction in emissions from Europe and Russia, which contributes to -18.6% and -18.8% of the decline of the Arctic sulfate concentrations, respectively. The change in emissions from Central Asia and North America, respectively, explains -1.6% and -3.4% of the reduced concentration.

Simulated Arctic BC concentration also shows a considerable decline before 2000, but a slight rise after 2000. Overall, the average concentration of BC in the Arctic had a decrease of 22.98% in 2014–2018 relative to 1980–1984, mainly due to the reductions in emissions originating from the Arctic and Russia, which lead to 9.32% and 14.91% of the concentration decrease (Table 1). Sources in Europe, North America, and East Asia account for the changes in Arctic near-surface BC concentration in range of ±1–3%. The remaining source regions have no substantial impact on the BC concentration in the Arctic (total contribution less than 2%) due to weak emission strength or long transport pathways. Since the Arctic sulfate and BC aerosol concentrations contributed by non-local sources have been reducing, the fractional contribution of Arctic local source increased from 33.6% and 53.4% to 55.1% and 57.3%, respectively. To further reduce present-day or future aerosols in the Arctic, efforts can be made to control the local sources in the Arctic.

Aerosols are often transported across continents in the free troposphere rather than near the surface, resulting in a higher relative contribution of non-local sources to the aerosol concentration at higher altitudes than near the surface. Figure 6 shows the vertical profiles of absolute and relative contributions of major source regions to sulfate

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020


Atmospheric Chemistry and Physics Discussions

and BC concentrations in the Arctic. Different source regions have very distinct vertical distributions of their contributions. The largest contribution of East and South Asia emissions is at 8-10 km, accounting for about two thirds of the Arctic aerosol concentrations at this height, which is consistent with results from other models (e.g., Shindell et al., 2008). Emissions from the Arctic and Russia account for the majority of Arctic sulfate and BC concentrations below 2 km and 6 km, respectively. The contributions of emissions from Europe and North America are mainly located below 10 km. As the source emissions vary with time, the vertical aerosol concentrations contributed by individual sources also change.

The changes in source contributions to the annual mean vertical profile of sulfate and BC concentrations over the Arctic between 2014–2018 and 1980–1984 are shown in Fig. 7. Due to the effective emission reduction, the contribution from Europe and Russia to the Arctic sulfate below 6 km was each decreased by nearly 0.1 μg m^{-3} in 2014–2018, compared to 1980–1984. North America contribution also had a slight decline below 2 km. Contributions from South and East Asia increased at the upper troposphere between 10–15 km, which is consistent with the increase in emissions over these regions, leading to a combined increase in sulfate concentration of up to 0.1 μg m^{-3} at the upper levels of the Arctic. The BC concentration below 2 km contributed by Arctic and Russia emissions each had a decrease of up to 2 ng m^{-3}, which dominated the decrease of BC concentration in the Arctic lower atmosphere. As with sulfate, BC concentrations contributed by East Asia and South Asia show an increasing trend in the high altitudes, mainly due to increased emissions in these two regions, offsetting the decrease in

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020


Atmospheric Chemistry and Physics Discussions

column burden owing to the reduced concentration in the lower altitudes.

Linear trends of the annual near-surface concentrations and column burden of sulfate and BC from 1980 to 2018 are shown in Fig. 8 and the individual source contributions to these trends are summarized in Table 2. During 1980–2018, simulated Arctic near-surface concentration and column burden of sulfate decreased by 20% and 13 % per decade, respectively. Due to the air pollution regulations in Europe and dissolution of the former Soviet Union, reductions in emissions from Europe and Russia led to decreasing trends of 7–10% per decade in the near-surface concentration and column burden of sulfate, having the largest contributions to sulfate trends among all tagged source regions. In addition, the change in North America emissions contributed to a 2–4% per decade decreasing trend in the Arctic sulfate concentration and burden, which is related to its emission control since 1980s. South and East Asia together contributed to an increase of total Arctic sulfate burden at a rate of 8% per decade, associated with the emission rise during this time period. The near-surface concentration of Arctic BC has a decreasing trend of 12% per decade during 1980–2018, mostly driven by the decreases in contributions from Russia and Arctic local emissions (6% per decade each). For BC column burden, the decreasing trends contributed by the reductions in emissions from Russia and Europe are offset by the increasing trends caused by emission increases in East and South Asia, resulting in an insignificant change of total BC burden during 1980–2018.

4. Aerosol radiative forcing and associated Arctic warming

Both sulfate and BC influence the Arctic climate through perturbing atmospheric and

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020


Atmospheric Chemistry and Physics Discussions

surface radiation balance. The spatial distribution of the climatological mean TOA radiative forcing due to aerosol-radiation interactions (RF_{ari}) of sulfate and BC during 1980–2018 is shown in Fig. 9. The Arctic sulfate exerts a negative RF_{ari} primarily by scattering incoming solar radiation back into the space, with the forcing in range of -0.4~0 Wm^{-2}, while BC can absorb solar radiation in the atmosphere and leads to a positive RF_{ari} of 0.1~0.4 Wm^{-2} in the Arctic. In the high and mid-latitudes of the Northern Hemisphere, the RF_{ari} of sulfate over Europe and Russia is in the range of -1.0~-0.4 Wm^{-2}. Sulfate RF_{ari} over North America varies from -0.2 Wm^{-2} to -1.0 Wm^{-2}. The negative RF_{ari} of sulfate over East Asia is more than -1.0 Wm^{-2}, mainly due to the high sulfate concentrations. BC over Europe, Russia and Central Asia exerts a positive RF_{ari} of 0.4~1 Wm^{-2}. The BC RF_{ari} over East Asia reaches a high value over 1.0 Wm^{-2}.

Previous studies have suggested that Arctic climate responds not only to Arctic local forcings but also to forcings outside the Arctic due to the meridional energy transport change (Navarro et al., 2016). To estimate the relative roles of recent aerosol trends in the Arctic warming, we look into the temporal variation of annual mean RF_{ari} of sulfate and BC in different latitude bands during 1980–2018 (Fig. 10). Within the Arctic (66.5°N–90°N), the negative RF_{ari} of sulfate decreases from -0.21 Wm^{-2} in 1980–1984 to a moderate value of -0.10 Wm^{-2} in 2014–2018, indicating a warming effect in the Arctic from the local sulfate change. Over the mid-latitudes (28°N–66.5°N), the sulfate RF_{ari} decreases from -0.87 Wm^{-2} to -0.53 Wm^{-2} between the first and last five years of 1980–2018, while the magnitude of the sulfate RF_{ari} in the tropical region (28°S–28°N) increases from -0.52 Wm^{-2} to -0.60 Wm^{-2}. The positive BC RF_{ari} increases from 0.55

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020


Atmospheric Chemistry and Physics Discussions

334 Wm^{-2} to 0.74 Wm^{-2} in the mid-latitudes and from 0.51 Wm^{-2} to 0.76 Wm^{-2} in the tropics,
335 while the BC RF_{ari} over the Arctic has no obvious change during this time period.
336 Systematic assessment of the impact of aerosols on Arctic warming since 1980s
337 requires quantifying the Arctic temperature responses to changes in radiative forcing of
338 different aerosol species over different regions. Here, we apply Arctic climate
339 sensitivity factors, defined as the Arctic temperature response per unit radiation, for
340 each short-lived climate forcers over the Arctic, mid-latitudes of the Northern
341 Hemisphere, tropics and Southern Hemisphere from Sand et al. (2016) and Shindell
342 and Faluvegi (2009) to calculate the recent Arctic surface temperature change related
343 to the variations in sulfate and BC radiative forcings over the different latitude bands
344 during 1980–2018 (Fig. 11 and Table 3). This method has been widely adopted to
345 examine the Arctic temperature response to aerosol forcings (e.g., Breider et al., 2017;
346 Flanner, 2013; Sand et al., 2016; Shindell and Faluvegi, 2009; Yang et al., 2018c).
347 It is estimated that, between 1980–1984 and 2014–2018, changes in total RF_{ari} of
348 sulfate and BC produce a surface warming of +0.145 K over the Arctic, with +0.088 K
349 (61%) contributed by the sulfate forcing change and the remaining explained by the BC
350 forcing change. The sulfate-related Arctic warming is mainly due to the decrease in
351 sulfate in mid-latitudes that enhances the temperature gradient between the mid-
352 latitudes and Arctic, resulting in a strengthened meridional heat transport and, therefore,
353 the Arctic warming of +0.059 K. The change in Arctic local RF_{ari} of sulfate provides
354 +0.035 K of the surface warming, while the forcing change in the tropics has a
355 negligible influence on the Arctic temperature change. The Arctic temperature

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020


Atmospheric Chemistry and Physics Discussions

356 responses to increases in BC RF_{ari} over the mid-latitudes and tropics are +0.029 K and
357 +0.031 K, respectively, related to the enhanced poleward heat transport from the
358 warming radiative impact in the mid-latitudes, while changes in the Arctic BC RF_{ari}
359 only exert a weak cooling of -0.005 K. Overall, the RF_{ari} change over the mid-latitudes
360 provides the strongest warming effect (+0.088K) to the Arctic compared to other
361 latitude bands, owing to the aerosol-induced increase in the poleward heat transport.

362 While the results above focus on the effects of aerosol-radiation interactions, the
363 aerosol-cloud interactions (RF_{aci}) and BC snow/ice albedo effects can also influence
364 Arctic climate. Sulfate RF_{aci} is estimated by scaling sulfate RF_{ari} based on the ratio of
365 sulfate RF_{aci} and RF_{ari} over different latitudes from Sand et al. (2016). Within the Arctic,
366 the magnitude of negative TOA RF_{aci} of sulfate decreases from -0.48 Wm^{-2} in 1980–
367 1984 to -0.23 Wm^{-2} in 2014–2018, indicating a warming effect due to the local sulfate
368 change. Over the mid-latitudes, the sulfate RF_{aci} decreases from -2.46 Wm^{-2} to -1.49
369 Wm^{-2} between the first and last five years of 1980–2018, while the magnitude of the
370 sulfate RF_{aci} in the tropical region increases from -1.78 Wm^{-2} to -2.08 Wm^{-2}. The
371 positive RF due to BC in snow/ice decreases from 0.34 Wm^{-2} in 1980–1984 to 0.29
372 Wm^{-2} in 2014–2018 over the Arctic, while that over the mid-latitudes increases from
373 0.19 Wm^{-2} to 0.23 Wm^{-2}.

374 Based on the Arctic climate sensitivities, impacts of changes in radiative forcing due
375 to aerosol-cloud interactions of sulfate are also estimated. The sulfate RF_{aci} provides an
376 Arctic warming of +0.193 K between 1980–1984 and 2014–2018, with +0.165 K
377 contributed by the RF_{aci} change over the mid-latitudes and +0.078 K resulting from the

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020


Atmospheric Chemistry and Physics Discussions

Arctic RF_{aci} change. It should be noted that aerosol-cloud interactions at high latitude regions are complicated and highly uncertain in climate models. The temperature changes presented here only provide a very rough estimate. BC in snow/ice reduces surface albedo and increases snow/ice melt (Flanner et al., 2007; Qian et al., 2015). Due to the decrease in Arctic BC concentration and depostion, BC concentration in the Arcitc snow has been decreasing (e.g., Zhang et al., 2019). The weakened BC snow/ice albedo effect leads to an Arctic cooling of –0.061 K, while the mid-latitude BC in snow/ice causes an Arctic warming of +0.019 K. The total BC snow/ice albedo effects result in an Arctic surface temperature change of –0.041 K during 1980–2018, partially offsetting the solar absorbing effect of BC in the atmosphere. Combining all the effects, we estimate that between 1980 and 2018, sulfate and BC contribute a total of +0.297 K to the Arctic surface temperature change, approximately 20% of the observed Arctic warming during this period.

5. Conclusions and discussion

The Arctic has warmed rapidly since the 1980s, with the surface air temperature increasing by 1.5 K. Many studies have examined possible mechanisms that caused the Arctic warming, but many are still on debate. In this study, we use a global aerosol-climate model equipped with the Explicit Aerosol Source Tagging module (CAM5-EAST) to quantify the source attribution of aerosols in the Arctic and the aerosol-related Arctic warming during 1980–2018. The model can reasonably simulate the spatial distribution and temporal variation of the Arctic near-surface sulfate and BC concentrations compared with several site measurements, while it underestimates the

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020


Atmospheric Chemistry and Physics Discussions

magnitude of sulfate and BC to some extent.

Compared to 1980–1984, the simulated annual average of sulfate and BC concentrations over the Arctic in 2014–2018 had a decrease of 42.8% and 23.0%, respectively. The decrease in emissions from Europe and Russia contributed -18.6% and -18.8% of the near-surface sulfate concentration decrease (out of -42.8%) and the reduction in Arctic local emissions and emission from Russia led to -9.3% and -14.9% of the BC concentration reduction (out of -23.0%), respectively. In 2014–2018, increases in emissions from South and East Asia together contributed to an increase of sulfate and BC concentrations up to 0.1 μg m^{-3} and 2 ng m^{-3}, respectively, at the upper troposphere, compared to 1980–1984. The contribution of Europe and Russia emissions to the Arctic sulfate concentration each had a decrease of about 0.1 μg m^{-3} under 6 km. Below 2 km, the BC concentration contributed by emissions from Arctic and Russia each had a decrease of up to 2 ng m^{-3}. Simulated sulfate near-surface concentration and column burden had a decreasing trend of 20% per decade and 13% per decade, respectively, in the Arctic during 1980–2018, mainly driven by the reductions in emissions from Europe and Russia, both of which led to decreasing trends at a rate of 7–10% per decade. Due to the decreases in contributions from Russia and Arctic local emissions (6% per decade each), the near-surface concentration of Arctic BC presents a decreasing trend of 12% per decade during 1980–2018.

Aerosols within and outside the Arctic can influence the Arctic climate through changing the radiative balance. The magnitude of negative TOA RF_{ari} of sulfate over the Arctic decreased from -0.21 Wm^{-2} in 1980–1984 to -0.10 Wm^{-2} in 2014–2018. Over

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020


Atmospheric Chemistry and Physics Discussions

the mid-latitudes, the sulfate RF_{ari} magnitude decreased from -0.87 Wm^{-2} to -0.53 Wm^{-2}, while the sulfate RF_{ari} over the tropics increased from -0.52 Wm^{-2} to -0.60 Wm^{-2}. The positive BC RF_{ari} in the mid-latitudes and tropics increased from 0.55 Wm^{-2} and 0.51 Wm^{-2} to 0.74 Wm^{-2} and 0.76 Wm^{-2}, respectively, while that over the Arctic had no significant change during this time period.

By applying Arctic climate sensitivity factors obtained from the literature to the variations in aerosol radiative forcing, the aerosol-induced Arctic surface temperature change is estimated in this study. During 1980–2018, through aerosol-radiation interactions, sulfate and BC together produced a +0.145 K warming to the Arctic, +0.088 K (61%) of which is contributed by sulfate. The decrease in sulfate in mid-latitudes led to an increase in Arctic temperature of +0.059 K, whereas the Arctic local sulfate provided +0.035 K of the surface warming. The Arctic temperature responses to changes in atmospheric BC over the mid-latitudes and tropics are +0.029 K and +0.031 K, respectively, while changes BC in the Arctic atmosphere only exert a weak cooling of -0.005 K. Through aerosol-cloud interactions, sulfate exerted an Arctic warming of +0.193 K during 1980–2018, with +0.165 K contributed by the forcing change over the mid-latitudes and +0.078 K due to the forcing change over the Arctic. Therefore, changes in aerosols over the mid-latitudes had the largest impact on the Arctic temperature than other regions during 1980–2018 through enhancing meridional temperature gradient and therefore poleward heat transport, followed by changes in Arctic local aerosol forcings. Due to the decrease in Arctic BC concentration, the weakened BC snow/ice albedo effect led to an Arctic cooling of –0.061 K, partially

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020


Atmospheric Chemistry and Physics Discussions

offset by Arctic warming of +0.019 K induced by the BC snow/ice albedo effect over the mid-latitudes. Combining all aerosol effects, sulfate and BC together produced a total of +0.297 K in the Arctic surface temperature change during 1980–2018, explaining approximately 20% of the observed Arctic warming during this period.

There are a few sources of uncertainty in the results presented in this study. As discussed above, the model underestimates the near-surface sulfate and BC concentrations over the Arctic, probably due to an overly aerosol wet removal during the long-range transport (e.g., Wang et al., 2013), uncertainties in aerosol emissions, and biases in observations. Here we only discussed the effects of sulfate and BC on the Arctic surface temperature without considering other aerosol species, due to large uncertainties in the simulation of second organic aerosols and the lack of other aerosol treatments (e.g., nitrate) in current model version. These may lead to biases of the aerosol climate effects in this study. In addition, we estimated the temperature response of the Arctic to the aerosol-induced TOA radiative forcing change based on the climate sensitivity factors derived from the literature. For more accurate estimation of the aerosol-related Arctic warming, the coupled model configuration with free running simulations should be conducted in the future. The RF_{ari} calculation follows Ghan et al. (2012), which falls into the definition of effective RF_{ari} (ERF_{ari}), while the climate sensitivity factors were calculated based on the stratospherically adjusted radiative forcing. Considering that the assessment for adjusted RF_{ari} (-0.35 ± 0.5 W m^{-2}) is slightly lower than that for ERF_{ari} (-0.45 ± 0.5 W m^{-2}) (Boucher et al., 2013), the temperature response could be relatively smaller than estimated here. The relatively

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020


Atmospheric Chemistry and Physics Discussions

low model resolution may not capture the complexity of the Arctic terrain (Yang et al., 2018c), which also induces uncertainties to the simulated aerosols in the Arctic. High resolution or regionally refined model is more desirable if resources allow. Given that assumed injection heights of anthropogenic emissions in models are uncertain, the ability to simulated surface aerosol concentrations and vertical distribution in models could also be compromised (Yang et al., 2019b). In this study, we did not discuss the effects of meteorological parameters on the long-term aerosol simulation mainly because the decadal aerosol variation is dominated by changes in anthropogenic emissions rather than meteorology (Yang et al., 2016).

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020


Atmospheric Chemistry and Physics Discussions

475 ***Data availability.***

476 The CAM5 model is available at http://www.cesm.ucar.edu/models/cesm1.2/ (last
477 access: 8 December 2019). Our CAM5-EAST model code and results can be made
478 available through the National Energy Research Scientific Computing Center (NERSC)
479 servers upon request.

480

481 ***Competing interests.***

482 The authors declare that they have no conflict of interest.

483

484 ***Author contribution***.

485 YY and HW designed the research; YY performed the model simulations; LR analyzed
486 the data. All the authors discussed the results and wrote the paper.

487

488 ***Acknowledgments.***

489 This research was support by the National Natural Science Foundation of China under
490 grant 41975159, Jiangsu Specially Appointed Professor Project and the U.S.
491 Department of Energy (DOE), Office of Science, Biological and Environmental
492 Research as part of the Earth and Environmental System Modeling program. The
493 Pacific Northwest National Laboratory is operated for DOE by Battelle Memorial
494 Institute under contract DE-AC05-76RLO1830. The National Energy Research
495 Scientific Computing Center (NERSC) provided computational support.

496

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020

Atmospheric Chemistry and Physics Discussions

References

Alexeev, V. A., Esau, I., Polyakov, I. V., Byam, S. J., and Sorokina, S.: Vertical structure of recent Arctic warming from observed data and reanalysis products, Clim. Change, 111, 215-239, https://doi.org/10.1007/s10584-011-0192-8, 2012.

Bond, T. C., Doherty, S. J., Fahey, D. W., Forster, P. M., Berntsen, T., DeAngelo, B. J., Flanner, M. G., Ghan, S., Kärcher, B., Koch, D., Kinne, S., Kondo, Y., Quinn, P. K., Sarofim, M. C., Schultz, M. G., Schulz, M., Venkataraman, C., Zhang, H., Zhang, S., Bellouin, N., Guttikunda, S. K., Hopke, P. K., Jacobson, M. Z., Kaiser, J. W., Klimont, Z., Lohmann, U., Schwarz, J. P., Shindell, D., Storelvmo, T., Warren, S. G., and Zender, C. S.:Bounding the role of black carbon in the climate system: A scientific assessment, J. Geophys. Res. Atmos., 118, 5380–5552, https://doi.org/10.1002/jgrd.50171, 2013.

Boucher, O., Randall, D., Artaxo, P., Bretherton, C., Feingold, G., Forster, P., Kerminen, V.-M., Kondo, Y., Liao, H., Lohmann, U., Rasch, P., Satheesh, S. K., Sherwood, S., Stevens, B., and Zhang, X. Y.: Clouds and Aerosols, in: Climate Change 2013: The Physical Science Basis, Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, edited by: Stocker, T. F., Qin, D., Plattner, G.-K., Tignor, M., Allen, S. K., Boschung, J., Nauels, A., Xia, Y., Bex, V., and Midgley, P. M., Cambridge University Press, Cambridge, UK and New York, NY, USA, 571–658, https://doi.org/10.1017/CBO9781107415324.016, 2013.

Bourgeois, Q., and Bey, I.: Pollution transport efficiency toward the Arctic: Sensitivity to aerosol scavenging and source regions, J. Geophys. Res. Atmos., 116, D08213, https://doi.org/10.1029/2010JD015096, 2011.

Breider, T. J., Mickley, L. J., Jacob, D. J., Wang, Q., Fisher, J. A., Chang, R. Y. W., and Alexander, B.: Annual distributions and sources of Arctic aerosol components, aerosol optical depth, and aerosol absorption, J. Geophys. Res. Atmos., 119, 4107–4124, https://doi.org/10.1002/2013JD020996, 2014.

Breider, T. J., Mickley, L. J., Jacob, D. J., Ge, C.,Wang, J., Sulprizio Payer, M., Croft, B., Ridley, D. A., McConnell, J. R., Sharma, S., Husain, L., Dutkiewicz, V. A., Eleftheriadis, K., Skov, H., and Hopke, P. K.: Multidecadal trends in aerosol radiative forcing over the Arctic: Contribution of changes in anthropogenic aerosol to Arctic warming since 1980, J. Geophys. Res. Atmos., 122, 3573–3594, https://doi.org/10.1002/2016JD025321, 2017.

Browse, J., Carslaw, K., Arnold, S., Pringle, K., and Boucher, O.: The scavenging processes controlling the seasonal cycle in Arctic sulphate and black carbon aerosol, Atmos. Chem. Phys., 12, 6775-6798, https://doi.org/10.5194/acp-12-6775-2012, 2012.

Dutkiewicz, V. A., DeJulio, A. M., Ahmed, T., Laing, J., Hopke, P. K., Skeie, R. B., Viisanen, Y., Paatero, J., and Husain, L.: Forty-seven years of weekly atmospheric black carbon measurements in the Finnish Arctic: Decrease in black carbon with declining emissions, J. Geophys. Res. Atmos., 119, 7667-7683, https://doi.org/10.1002/2014JD021790, 2014.

Eckhardt, S., Quennehen, B., Olivié, D. J. L., Berntsen, T. K., Cherian, R., Christensen, J. H., Collins, W., Crepinsek, S., Daskalakis, N., Flanner, M., Herber, A., Heyes, C., Hodnebrog, Ø., Huang, L., Kanakidou, M., Klimont, Z., Langner, J., Law, K. S., Lund, M. T., Mahmood, R., Massling, A., Myriokefalitakis, S., Nielsen, I. E., Nøjgaard, J. K., Quaas, J., Quinn, P. K., Raut, J.-C., Rumbold, S. T., Schulz, M., Sharma, S., Skeie, R. B., Skov, H., Uttal, T., von Salzen, K., and

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020

Stohl, A.: Current model capabilities for simulating black carbon and sulfate concentrations in the Arctic atmosphere: a multi-model evaluation using a comprehensive measurement data set, Atmos. Chem. Phys., 15, 9413–9433, https://doi.org/10.5194/acp-15-9413-2015, 2015.

Eleftheriadis, K., Vratolis, S., and Nyeki, S.: Aerosol black carbon in the European Arctic: measurements at Zeppelin station, Ny-Ålesund, Svalbard from 1998–2007, Geophys. Res. Lett., 36, L02809, https://doi.org/10.1029/2008GL035741, 2009.

Fisher, J. A., Jacob, D. J., Wang, Q., Bahreini, R., Carouge, C. C., Cubison, M. J., Dibb, J. E., Diehl, T., Jimenez, J. L., Leibensperger, E. M., Lu, Z., Meinders, M. B. J., Pye, H.O. T., Quinn, P. K., Sharma, S., Streets, D. G., van Donkelaar, A., and Yantosca, R. M.: Sources, distribution, and acidity of sulfate–ammonium aerosol in the Arctic in winter–spring, Atmos. Environ., 45, 7301–7318, https://doi.org/10.1016/j.atmosenv.2011.08.030, 2011.

Flanner, M. G., Zender, C. S., Randerson, J. T., and Rasch, P. J.: Present-day climate forcing and response from black carbon in snow, J. Geophys. Res., 112, D11202, https://doi.org/10.1029/2006JD008003, 2007.

Flanner, M. G.: Arctic climate sensitivity to local black carbon, J. Geophys. Res. Atmos., 118, 1840-1851, https://doi.org/10.1002/jgrd.50176, 2013.

Garrett, T. J., Zhao, C., and Novelli, P.: Assessing the relative contributions of transport efficiency and scavenging to seasonal variability in Arctic aerosol, Tellus B, 62, 190–196, https://doi.org/10.1111/j.1600-0889.2010.00453.x, 2010.

Garrett, T. J., Brattström, S., Sharma, S., Worthy, D. E., and Novelli, P.: The role of scavenging in the seasonal transport of black carbon and sulfate to the Arctic, Geophys. Res. Lett., 38, L16805, https://doi.org/10.1029/2011GL048221, 2011.

Gelaro, R., McCarty, W., Suárez, M. J., Todling, R., Molod, A., Takacs, L., Randles, C. A., Darmenov, A., Bosilovich, M. G., Reichle, R., Wargan, K., Coy, L., Cullather, R., Draper, C., Akella, S., Buchard, V., Conaty, A., da Silva, A. M., Gu, W., Kim, G.-K., Koster, R., Lucchesi, R., Merkova, D., Nielsen, J. E., Partyka, G., Pawson, S., Putman, W., Rienecker, M., Schubert, S. D., Sienkiewicz, M., and Zhao, B.: The Modern-Era Retrospective Analysis for Research and 476 Applications, Version 2 (MERRA-2), J. Climate, 30, 5419–5454, https://doi.org/10.1175/JCLI-D-16-0758.1, 2017.

Ghan, S. J., Liu, X., Easter, R. C., Zaveri, R., Rasch, P. J., Yoon, J. H., and Eaton, B.: Toward a minimal representation of aerosols in climate models: Comparative decomposition of aerosol direct, semidirect, and indirect radiative forcing, J. Climate, 25, 6461–6476, https://doi.org/10.1175/JCLI-D-11-00650.1, 2012.

Gong, S. L., Zhao, T. L., Sharma, S., Toom-Sauntry, D., Lavoué, D., Zhang, X. B., Leaitch,W. R., and Barrie, L. A.: Identification of trends and interannual variability of sulfate and black carbon in the Canadian High Arctic: 1981–2007, J. Geophys. Res., 115, D07305, https://doi.org/10.1029/2009JD012943, 2010.

Graversen, R. G., Mauritsen, T., Tjernström, M., Källén, E., and Svensson, G.: Vertical structure of recent Arctic warming, Nature, 451, 53-56, https://doi.org/10.1038/nature06502, 2008.

Heidam, N. Z., Wåhlin, P., and Christensen, J. H.: Tropospheric gases and aerosols in northeast Greenland, J. Atmos. Sci., 56, 261-278, https://doi.org/10.1175/1520-0469(1999)056<0261:TGAAIN>2.0.CO;2, 1999.

Hirdman, D., Burkhart, J. F., Sodemann, H., Eckhardt, S., Jefferson, A., Quinn, P. K., Sharma, S., Ström, J., and Stohl, A.: Long-term trends of black carbon and sulphate aerosol in the Arctic:

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020

changes in atmospheric transport and source region emissions, Atmos. Chem. Phys., 10, 9351-9368, https://doi.org/10.5194/acp-10-9351-2010, 2010.

Hoesly, R. M., Smith, S. J., Feng, L., Klimont, Z., Janssens-Maenhout, G., Pitkanen, T., Seibert, J. J., Vu, L., Andres, R. J., Bolt, R. M., Bond, T. C., Dawidowski, L., Kholod, N., Kurokawa, J.-I., Li, M., Liu, L., Lu, Z., Moura, M. C. P., O'Rourke, P. R., and Zhang, Q.: Historical (1750–2014) anthropogenic emissions of reactive gases and aerosols from the Community Emissions Data System (CEDS), Geosci. Model Dev., 11, 369–408, https://doi.org/10.5194/gmd-11-369-2018, 2018.

Koch, D., and Hansen, J.: Distant origins of Arctic black carbon: a Goddard Institute for Space Studies ModelE experiment, J. Geophys. Res., 110, D04204, https://doi.org/10.1029/2004JD005296, 2005.

Koch, D., Schulz, M., Kinne, S., McNaughton, C., Spackman, J. R., Balkanski, Y., Bauer, S., Berntsen, T., Bond, T. C., Boucher, O., Chin, M., Clarke, A., De Luca, N., Dentener, F., Diehl, T., Dubovik, O., Easter, R., Fahey, D. W., Feichter, J., Fillmore, D., Freitag, S., Ghan, S., Ginoux, P., Gong, S., Horowitz, L., Iversen, T., Kirkevåg, A., Klimont, Z., Kondo, Y., Krol, M., Liu, X., Miller, R., Montanaro, V., Moteki, N., Myhre, G., Penner, J. E., Perlwitz, J., Pitari, G., Reddy, S., Sahu, L., Sakamoto, H., Schuster, G., Schwarz, J. P., Seland, Ø., Stier, P., Takegawa, N., Takemura, T., Textor, C., van Aardenne, J. A., and Zhao, Y.: Evaluation of black carbon estimations in global aerosol models, Atmos. Chem. Phys., 9, 9001–9026, https://doi.org/10.5194/acp-9-9001-2009, 2009.

Law, K. S., and Stohl, A.: Arctic air pollution: Origins and impacts, Science, 315, 1537-1540, https://doi.org/10.1126/science, 2007.

Lou, S., Yang, Y., Wang, H., Lu, J., Smith, S. J., Liu, F., and Rasch P. J.: Black carbon increases frequency of extreme ENSO events, J. Climate, 32, 8323–8333, https://doi.org/10.1175/JCLI-D-19-0549.1, 2019a.

Lou, S., Yang, Y., Wang, H., Smith, S. J., Qian, Y., and Rasch, P. J., Black carbon amplifies haze over the North China Plain by weakening the East Asian winter monsoon, Geophys. Res. Lett., 46, 452–460, https://doi.org/10.1029/2018GL080941, 2019b.

McConnell, J. R., Edwards, R., Kok, G. L., Flanner, M. G., Zender, C. S., Saltzman, E. S., Banta, J. R., Pasteris, D. R., Carter, M. M., and Kahl, J. D. W.: 20th-century industrial black carbon emissions altered arctic climate forcing, Science, 317, 1381–1384, https://doi.org/10.1126/science.1144856, 2007.

Mcfarquhar, G. M., and Wang, H.: Effects of aerosols on trade wind cumuli over the Indian Ocean: Model simulations, Q. J. R. Meteorol. Soc., 132, 821-843, https://doi.org/10.1256/qj.04.179, 2006.

Navarro, J. C. A., Varma, V., Riipinen, I., Seland, Ø., Kirkevåg, A., Struthers, H., Iversen, T., Hansson, H.-C., and Ekman, A. M. L.: Amplification of Arctic warming by past air pollution reductions in Europe, Nat. Geosci., 9, 277–281, https://doi.org/10.1038/ngeo2673, 2016.

O'Neill, N. T., Baibakov, K., Hesaraki, S., Ivanescu, L., Martin, R. V., Perro, C., Chaubey, J. P., Herber, A., and Duck, T. J.: Temporal and spectral cloud screening of polar winter aerosol optical depth (AOD): impact of homogeneous and inhomogeneous clouds and crystal layers on climatological-scale AODs, Atmos. Chem. Phys., 16, 12753-12765, https://doi.org/10.5194/acp-16-12753-2016, 2016.

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020

Pithan, F. and Mauritsen, T.: Arctic amplification dominated by temperature feedbacks in contemporary climate models, Nat. Geosci., 7, 181–184, https://doi.org/10.1038/ngeo2071, 2014.

Polissar, A. V., Hopke, P. K., and Harris, J. M.: Source regions for atmospheric aerosol measured at Barrow, Alaska, Environ. Sci. Technol., 35, 4214-4226, https://doi.org/10.1021/es0107529, 2001.

Qi, L., Li, Q., Henze, D. K., Tseng, H.-L., and He, C.: Sources of springtime surface black carbon in the Arctic: an adjoint analysis for April 2008, Atmos. Chem. Phys., 17, 9697–9716, https://doi.org/10.5194/acp-17-9697-2017, 2017.

Qian, Y., Yasunari, T. J., Doherty, S. J., Flanner, M. G., Lau, W. K., Ming, J., Wang, H., Wang, M., Warren, S. G., and Zhang, R.: Light-absorbing particles in snow and ice: Measurement and modeling of climatic and hydrological impact, Adv. Atmos. Sci., 32, 64-91, https://doi.org/10.1007/s00376-014-0010-0, 2015.

Quaas, J., Ming, Y., Menon, S., Takemura, T., Wang, M., Penner, J. E., Gettelman, A., Lohmann, U., Bellouin, N., and Boucher, O.: Aerosol indirect effects–general circulation model intercomparison and evaluation with satellite data, Atmos. Chem. Phys., 9, 8697-8717, https://doi.org/10.5194/acp-9-8697-2009, 2009.

Quinn, P., Shaw, G., Andrews, E., Dutton, E., Ruoho-Airola, T., and Gong, S.: Arctic haze: current trends and knowledge gaps, Tellus B, 59, 99-114, https://doi.org/10.1111/j.1600-0889.2006.00236.x, 2007.

Quinn, P. K., Bates, T. S., Baum, E., Doubleday, N., Fiore, A. M., Flanner, M., Fridlind, A., Garrett, T. J., Koch, D., Menon, S., Shindell, D., Stohl, A., and Warren, S. G.: Short-lived pollutants in the Arctic: their climate impact and possible mitigation strategies, Atmos. Chem. Phys., 8, 1723–1735, https://doi.org/10.5194/acp-8-1723-2008, 2008.

Quinn, P. K., Bates, T. S., Schulz, K., and Shaw, G. E.: Decadal trends in aerosol chemical composition at Barrow, Alaska: 1976–2008, Atmos. Chem. Phys., 9, 8883–8888, https://doi.org/10.5194/acp-9-8883-2009, 2009.

Raatz, W. E., and Shaw, G. E.: Long-range tropospheric transport of pollution aerosols into the Alaskan Arctic, J. Clim. Appl. Meteor., 23, 1052-1064, https://doi.org/10.1175/1520-0450, 1984.

Rahn, K. A., Borys, R. D., and Shaw, G. E.: The Asian source of Arctic haze bands, Nature, 268, 713-715, https://doi.org/10.1038/268713a0, 1977.

Rahn, K. A.: Relative importances of North America and Eurasia as sources of Arctic aerosol, Atmos. Environ., 15, 1447-1455, https://doi.org/10.1016/0004-6981(81)90351-6, 1981.

Sand, M., Berntsen, T., Von Salzen, K., Flanner, M., Langner, J., and Victor, D.: Response of Arctic temperature to changes in emissions of short-lived climate forcers, Nat. Clim. Change, 6, 286-289, https://doi.org/10.1038/nclimate2880, 2016.

Screen, J. A., and Simmonds, I.: Increasing fall-winter energy loss from the Arctic Ocean and its role in Arctic temperature amplification, Geophys. Res. Lett., 37, L16707, https://doi.org/10.1029/2010GL044136, 2010a.

Screen, J. A., and Simmonds, I.: The central role of diminishing sea ice in recent Arctic temperature amplification, Nature, 464, 1334-1337, https://doi.org/10.1038/nature09051, 2010b.

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020

Atmospheric Chemistry and Physics Discussions

669 Serreze, M. C., Barrett, A. P., Stroeve, J. C., Kindig, D. N., and Holland, M. M.: The emergence of
670 surface-based Arctic amplification, The Cryosphere, 3, 11–19, https://doi.org/10.5194/tc-3-11-
671 2009, 2009.
672 Sharma, S., Lavoué, D., Cachier, H., Barrie, L. A., and Gong, S. L.: Long-term trends of the black
673 carbon concentrations in the Canadian Arctic, J. Geophys. Res. Atmos., 109, D15203,
674 https://doi.org/10.1029/2003JD004331, 2004.
675 Sharma, S., Andrews, E., Barrie, L., Ogren, J., and Lavoué, D.: Variations and sources of the
676 equivalent black carbon in the high Arctic revealed by long-term observations at Alert and
677 Barrow: 1989–2003, J. Geophys. Res., 111, D14208, https://doi.org/10.1029/2005JD006581,
678 2006.
679 Sharma, S., Ishizawa, M., Chan, D., Lavoué, D., Andrews, E., Eleftheriadis, K., and Maksyutov, S.:
680 16-year simulation of Arctic black carbon: Transport, source contribution, and sensitivity
681 analysis on deposition, J. Geophys. Res. Atmos., 118, 943-964,
682 https://doi.org/10.1029/2012JD017774, 2013.
683 Sharma, S., Leaitch, W. R., Huang, L., Veber, D., Kolonjari, F., Zhang, W., Hanna, S. J., Bertram,
684 A. K., and Ogren, J. A.: An Evaluation of three methods for measuring black carbon at Alert,
685 Canada, Atmos. Chem. Phys., 17, 15225–15243, https://doi.org/10.5194/acp-2017-339, 2017.
686 Shindell, D. T., Chin, M., Dentener, F., Doherty, R. M., Faluvegi, G., Fiore, A. M., Hess, P., Koch,
687 D. M., MacKenzie, I. A., Sanderson, M. G., Schultz, M. G., Schulz, M., Stevenson, D. S., Teich,
688 H., Textor, C., Wild, O., Bergmann, D. J., Bey, I., Bian, H., Cuvelier, C., Duncan, B. N.,
689 Folberth, G., Horowitz, L.W., Jonson, J., Kaminski, J.W., Marmer, E., Park, R., Pringle, K. J.,
690 Schroeder, S., Szopa, S., Takemura, T., Zeng, G., Keating, T. J., and Zuber, A.: A multi-model
691 assessment of pollution transport to the Arctic, Atmos. Chem. Phys., 8, 5353–5372,
692 https://doi.org/10.5194/acp-8-5353-2008, 2008.
693 Shindell, D., and Faluvegi, G.: Climate response to regional radiative forcing during the twentieth
694 century, Nat. Geosci., 2, 294-300, https://doi.org/10.1038/ngeo473, 2009.
695 Sinha, P. R., Kondo, Y., Koike, M., Ogren, J. A., Jefferson, A., Barrett, T. E., Sheesley, R. J., Ohata,
696 S., Moteki, N., Coe, H., Liu, D., Irwin, M., Tunved, P., Quinn, P. K. and Zhao, Y.: Evaluation
697 of ground-based black carbon measurements by filter-based photometers at two Arctic sites, J.
698 Geophys. Res. Atmos., 122, 3544–3572, https://doi.org/10.1002/2016JD025843, 2017.
699 Sirois, A., and Barrie, L. A.: Arctic lower tropospheric aerosol trends and composition at Alert,
700 Canada: 1980–1995, J. Geophys. Res. Atmos., 104, 11599-11618,
701 https://doi.org/10.1029/1999JD900077, 1999.
702 Stohl, A.: Characteristics of atmospheric transport into the Arctic troposphere, J. Geophys. Res., 111,
703 D11306, https://doi.org/10.1029/2005JD006888, 2006.
704 Trenberth, K., Jones, P., Ambenje, P., Bojariu, R., Easterling, D., Klein Tank, A., Parker, D.,
705 Rahimzadeh, F., Renwick, J., and Rusticucci, M.: Observations: surface and atmospheric
706 climate change. chap. 3 of Climate Change: The Physical Science Basis, in Contribution of
707 Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate
708 Change, edited by S. Solomon et al., 235–336, Cambridge Univ. Press, Cambridge, U. K., and
709 New York, 2007.
710 Twomey, S.: Pollution and the planetary albedo, Atmos. Environ., 8, 1251-1256,
711 https://doi.org/10.1016/0004-6981(74)90004-3, 1974.

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020

712 Wang, H., Easter, R. C., Rasch, P. J., Wang, M., Liu, X., Ghan, S. J., Qian, Y., Yoon, J.-H., Ma, P.-
713 L., and Vinoj, V.: Sensitivity of remote aerosol distributions to representation ofcloud–aerosol
714 interactions in a global climate model, Geosci. Model Dev., 6, 765-782,
715 https://doi.org/10.5194/gmd-6-765-2013, 2013.
716 Wang, H., Rasch, P. J., Easter, R. C., Singh, B., Zhang, R., Ma, P. L., Qian, Y., Ghan, S. J., and
717 Beagley, N.: Using an explicit emission tagging method in global modeling of source-receptor
718 relationships for black carbon in the Arctic: Variations, sources, and transport pathways, J.
719 Geophys. Res., 119, 12,888-12,909, https://doi.org/10.1002/2014JD022297, 2014.
720 Yang, Y., Liao, H., and Lou, S.: Increase in winter haze over eastern China in recent decades: Roles
721 of variations in meteorological parameters and anthropogenic emissions, J. Geophys. Res., 121,
722 13, 050-13, 065, https://doi.org/10.1002/2016JD025136, 2016.
723 Yang, Y., Wang, H., Smith, S. J., Easter, R., Ma, P.-L., Qian, Y., Yu, H., Li, C., and Rasch, P. J.:
724 Global source attribution of sulfate concentration and direct and indirect radiative forcing,
725 Atmos. Chem. Phys., 17, 8903–8922, https://doi.org/10.5194/acp-17-8903-2017, 2017a.
726 Yang, Y., Wang, H., Smith, S. J., Ma, P.-L., and Rasch, P. J.: Source attribution of black carbon and
727 its direct radiative forcing in China, Atmos. Chem. Phys., 17, 4319–4336,
728 https://doi.org/10.5194/acp-17-4319-2017, 2017b.
729 Yang, Y., Wang, H., Smith, S. J., Easter, R. C., and Rasch, P. J.: Sulfate aerosol in the Arctic: Source
730 attribution and radiative forcing, J. Geophys. Res., 123, 1899-1918,
731 https://doi.org/10.1002/2017JD027298, 2018a.
732 Yang, Y., Wang, H., Smith, S. J., Zhang, R., Lou, S., Qian, Y., Ma, P.-L., and Rasch, P. J.: Recent
733 intensification of winter haze in China linked to foreign emissions and meteorology, Sci. Rep.,
734 8, 2107, https://doi.org/10.1038/s41598-018-20437-7, 2018b.
735 Yang, Y., Wang, H., Smith, S. J., Zhang, R., Lou, S., Yu, H., Li, C., and Rasch, P. J.: Source
736 apportionments of aerosols and their direct radiative forcing and long-term trends over
737 continental United States, Earth's Future, 6, 793-808, https://doi.org/10.1029/2018EF000859,
738 2018c.
739 Yang, Y., Smith, S. J., Wang, H., Lou, S., and Rasch, P. J.: Impact of anthropogenic emission
740 injection height uncertainty on global sulfur dioxide and aerosol distribution, J. Geophys.
741 Res.,124, 4812-4826, https://doi.org/10.1029/2018JD030001, 2019a.
742 Yang, Y., Smith, S. J., Wang, H., Mills, C. M., and Rasch, P. J.: Variability, timescales, and
743 nonlinearity in climate responses to black carbon emissions, Atmos. Chem. Phys., 19, 2405-
744 2420, https://doi.org/10.5194/acp-19-2405-2019, 2019b.
745 Zamora, L. M., Kahn, R. A., Eckhardt, S., McComiskey, A., Sawamura, P., Moore, R., and Stohl,
746 A.: Aerosol indirect effects on the nighttime Arctic Ocean surface from thin, predominantly
747 liquid clouds, Atmos. Chem. Phys., 17, 7311-7332, https://doi.org/10.5194/acp-17-7311-2017,
748 2017.
749 Zhang, R., Wang, H., Fu, Q., Pendergrass, A. G., Wang, M., Yang, Y., Ma P.-L., and Rasch, P. J.:
750 Local radiative feedbacks over the Arctic based on observed short-term climate variations,
751 Geophys. Res. Lett., 45, 5761–5770, https://doi.org/10.1029/2018GL077852, 2018.
752 Zhang, R., Wang, H., Fu, Q., Rasch, P. J., and Wang, X.: Unraveling driving forces explaining
753 significant reduction in satellite-inferred Arctic surface albedo since the 1980s, P. Natl. Acad.
754 Sci. USA, 116, 23947-23953, https://doi.org/10.1073/pnas.1915258116, 2019.

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020

Atmospheric Chemistry and Physics Discussions

755 Zhao, C., and Garrett, T. J.: Effects of Arctic haze on surface cloud radiative forcing, Geophys. Res.
756 Lett., 42, 557-564, https://doi.org/10.1002/2014GL062015, 2015.

757

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020

Atmospheric Chemistry and Physics Discussions

758 **Table 1.** Contributions of emissions from major source regions to the simulated
759 annual mean near-surface sulfate and BC concentrations (μg m^{-3}) averaged over the
760 Arctic in 1980–1984 and 2014–2018, as well as the percentage differences (%)
761 between 1980–1984 and 2014–2018 relative to 1980–1984.
762

	Sulfate Conc.		
	1980-1984	**2014-2018**	**Last 5 -First 5**
Sum	0.447	0.256	-42.83%
ARC	0.15	0.141	-2.02%
EUR	0.097	0.014	-18.61%
NAM	0.022	0.007	-3.36%
CAS	0.013	0.006	-1.57%
RBU	0.129	0.045	-18.83%
OCN	0.029	0.032	0.67%
OTH	0.006	0.01	0.90%
	BC Conc.		
	1980-1984	**2014-2018**	**Last 5 -First 5**
Sum	0.0161	0.0124	-22.98%
ARC	0.0086	0.0071	-9.32%
EUR	0.0011	0.0006	-3.11%
NAM	0.0004	0.0009	3.11%
EAS	0.0002	0.0003	0.62%
RBU	0.0056	0.0032	-14.91%
OTH	0.0002	0.0003	0.62%

763

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020

Atmospheric Chemistry and Physics Discussions

764 **Table 2.** Trends in annual mean sulfate and BC concentrations (% per decade) in
765 surface air and in the column contributed by 16 anthropogenic source regions during
766 1980–2018 relative to the 39-year averages.
767

Region	Sulfate Conc.	Sulfate Burden	BC Conc.	BC Burden
Sum	-19.83%	-13.18%	-11.93%	3.98%
EUR	-8.42%	-10.30%	-1.61%	-2.26%
NAM	-1.52%	-3.90%	0.96%	1.45%
CAM	0.00%	0.05%	0.00%	-0.01%
SAM	0.00%	-0.03%	0.00%	0.01%
NAF	0.02%	0.12%	0.05%	0.51%
SAF	0.00%	-0.02%	0.00%	0.18%
MDE	0.09%	0.85%	0.04%	0.79%
SEA	0.00%	0.11%	0.00%	0.09%
CAS	-0.72%	-1.01%	-0.04%	-0.05%
SAS	0.06%	3.49%	0.04%	1.97%
EAS	0.45%	4.24%	0.43%	5.90%
RBU	-8.54%	-6.64%	-6.12%	-3.74%
PAN	0.00%	0.00%	0.00%	0.00%
ARC	-1.38%	-0.20%	-5.96%	-1.01%
ANT	0.00%	0.00%	0.00%	0.00%
OCN	0.14%	0.08%	0.27%	0.16%

768

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020

Atmospheric Chemistry and Physics Discussions

Table 3. Estimated annual mean of the response in Arctic surface temperatures (K) to the change in TOA radiative forcing due to aerosol-radiation interactions (RF_{ari}) of sulfate and BC, aerosol-cloud interactions (RF_{aci}) of sulfate and radiative forcing (RF) due to BC in snow/ice (W m^{-2}) in each latitude band. The Arctic equilibrium temperature response is estimated using Arctic climate sensitivity factors (λ, K W^{-1} m^2), defined as the change in Arctic surface temperature per unit RF, for different latitudinal bands from Sand et al. (2016) and Shindell and Faluvegi (2009).

Forcing location	**Arctic equilibrium surface temperature response (K)***			
	Sulfate RF_{ari}	**Sulfate RF_{aci}**	**BC RF_{ari}**	**BC snow/ice**
60°N - 90°N	0.035	0.078	-0.005	-0.061
28°N - 60°N	0.059	0.165	0.029	0.019
28°S - 28°N	-0.001	-0.048	0.031	0.000
90°S - 28°S	-0.005	-0.002	0.002	0.000
SUM	0.088	0.193	0.057	-0.041

*The λ are 0.31, 0.17, 0.16, 0.06 for sulfate RF_{ari} and RF_{aci}; -0.08, 0.15, 0.31, 0.06 for BC RF_{ari}; 1.06, 0.45, 0.93, 0.18 for RF due to BC in snow/ice, according to the order given by forcing locations in the table. Sulfate RF_{aci} is not archived in this study and is roughly estimated here by scaling sulfate RF_{ari} based on the ratio of sulfate RF_{aci} and RF_{ari} over different latitudes from Sand et al. (2016).

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020

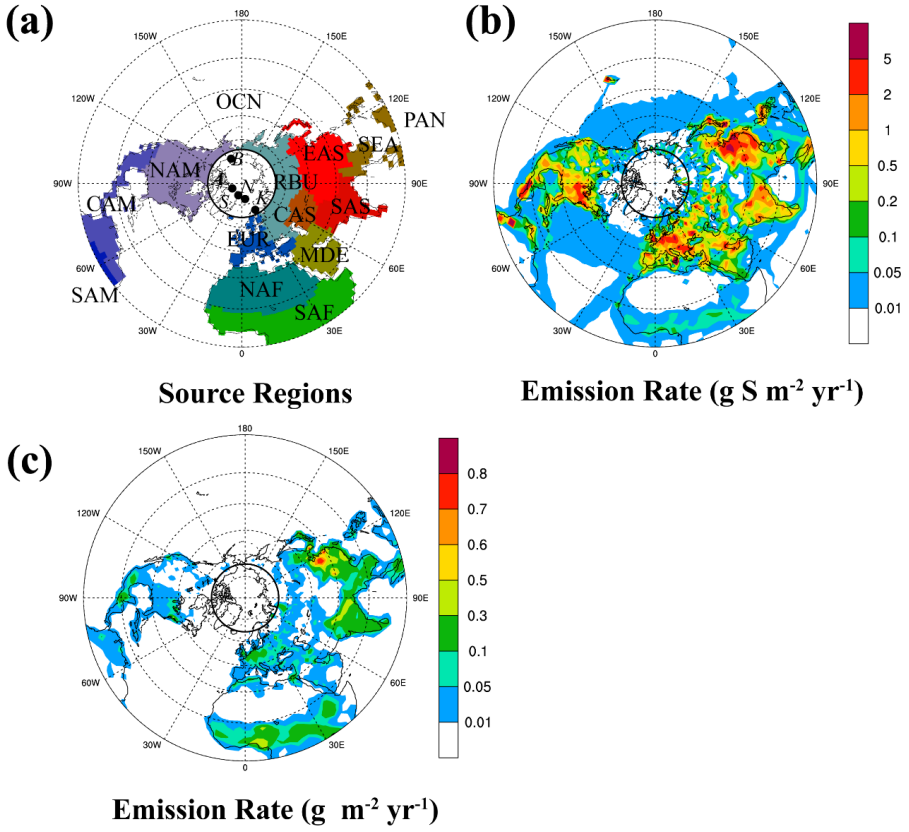


Figure 1. (a) Sixteen anthropogenic source regions (Europe (EUR), North America (NAM), Central America (CAM), South America (SAM), North Africa (NAF), South Africa (SAF), the Middle East (MDE), Southeast Asia (SEA), Central Asia (CAS), South Asia (SAS), East Asia (EAS), Russia-Belarus-Ukraine (RBU), Pacific-Australia-New Zealand (PAN), the Arctic (ARC), Antarctic (ANT), and Non-Arctic/Antarctic Ocean (OCN)). Spatial distribution of annual mean (b) SO_2 (g S m^{-2} yr^{-1}) and (c) BC (g C m^{-2} yr^{-1}) emissions averaged over 1980-2018. The thick black circle represents the Arctic (66.5°N–90°N). Dots in (a) are observational sites at Alert ("A", 82°N, 62°W), Station Nord ("S", 81°N, 16°W), Barrow ("B", 71°N, 156°W), Ny-Alesund ("N", 78°N, 11°E) and Kevo ("K", 69°N, 27°E).

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020

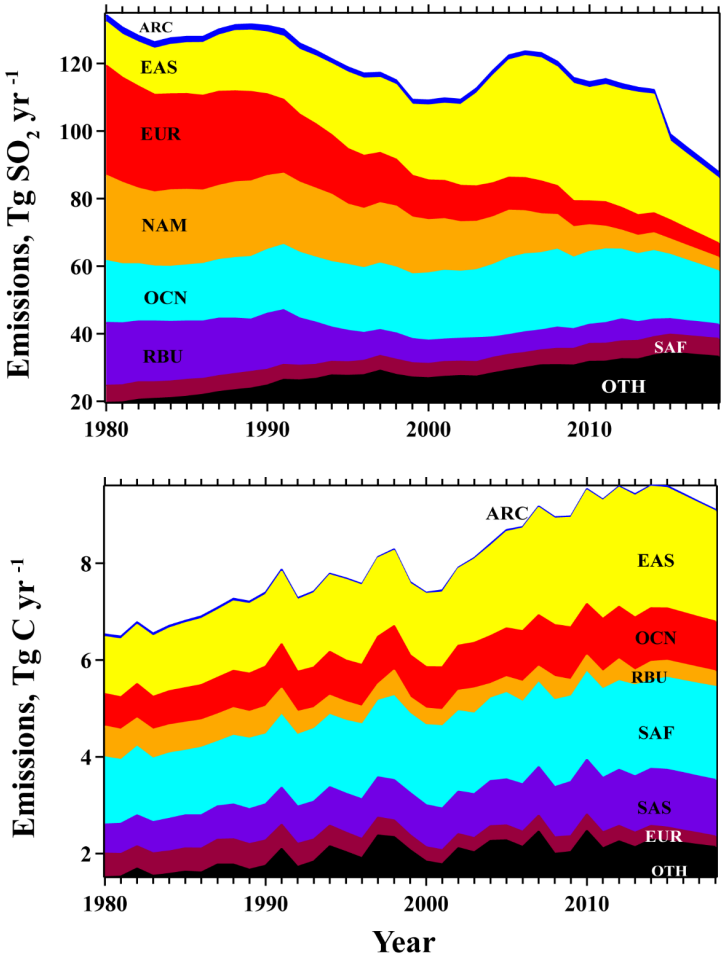
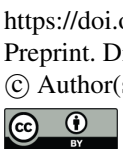


Figure 2. Time series of global total anthropogenic emissions of (top) SO_2 (Tg SO_2 yr^{-1}) and (bottom) BC (Tg C yr^{-1}), classified by key anthropogenic source regions.

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020



Atmospheric Chemistry and Physics Discussions

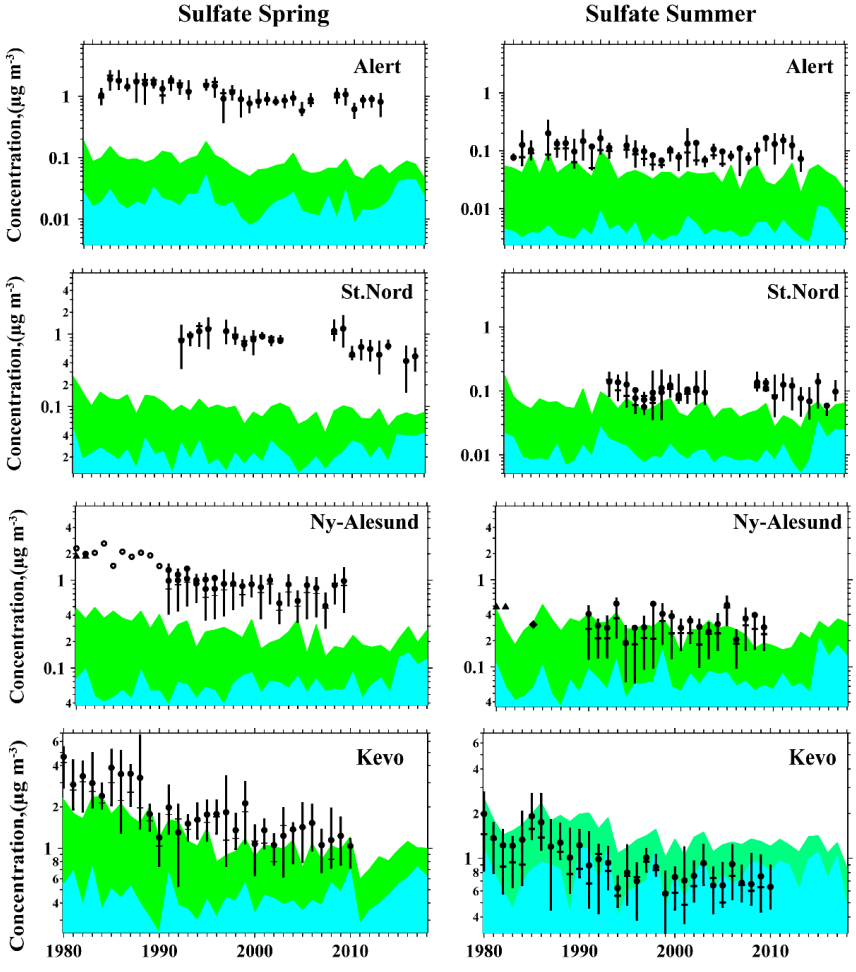


Figure 3. Surface concentrations of sulfate aerosols (μg m^{-3}) in spring (March–May) and summer (June–August) at four locations (Alert, Station Nord, Ny-Alesund, Kevo) in the Arctic during 1980–2018. Seasonal means are denoted by solid black circles, medians as short horizontal bars, and the 25th to 75th percentile ranges as vertical bars. Stacked contours represent the Arctic (blue) and non-Arctic anthropogenic source region (green) contributions to the modeled concentrations.

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020

Atmospheric Chemistry and Physics Discussions

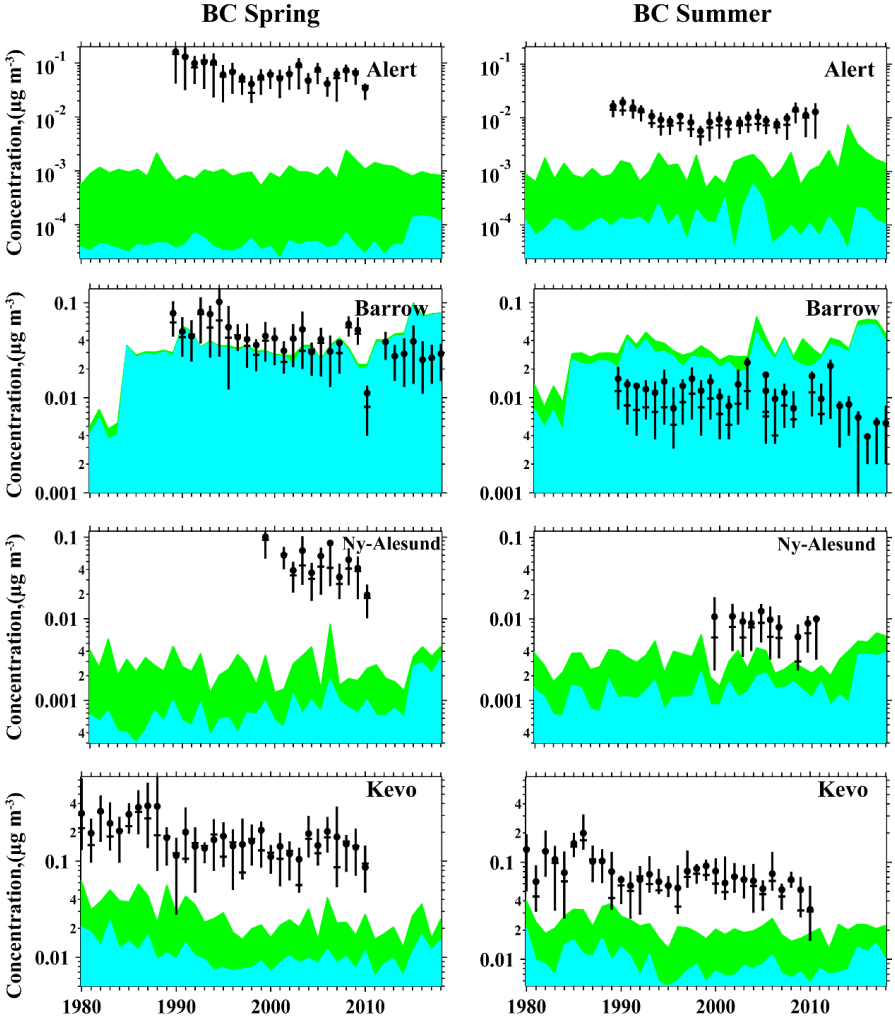


Figure 4. Same as Figure 3, but for surface BC (μg m^{-3}) at four (Alert, Barrow, Ny-Alesund, Kevo) Arctic sites.

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020

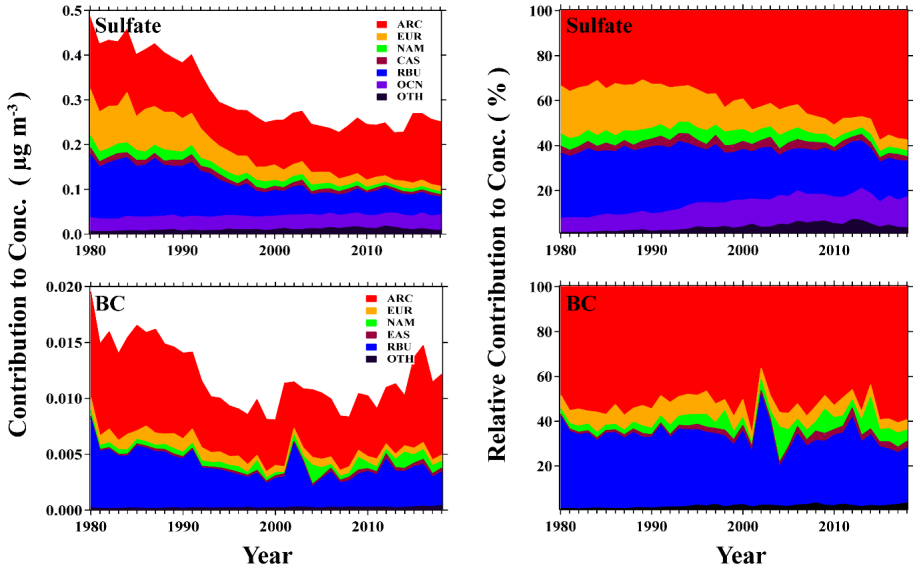


Figure 5. Time series (1980–2018) of absolute (left, μg m^{-3}) and relative (right, %) contributions of emissions from major source regions to the simulated annual mean near-surface sulfate and BC concentrations averaged over the Arctic.

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020

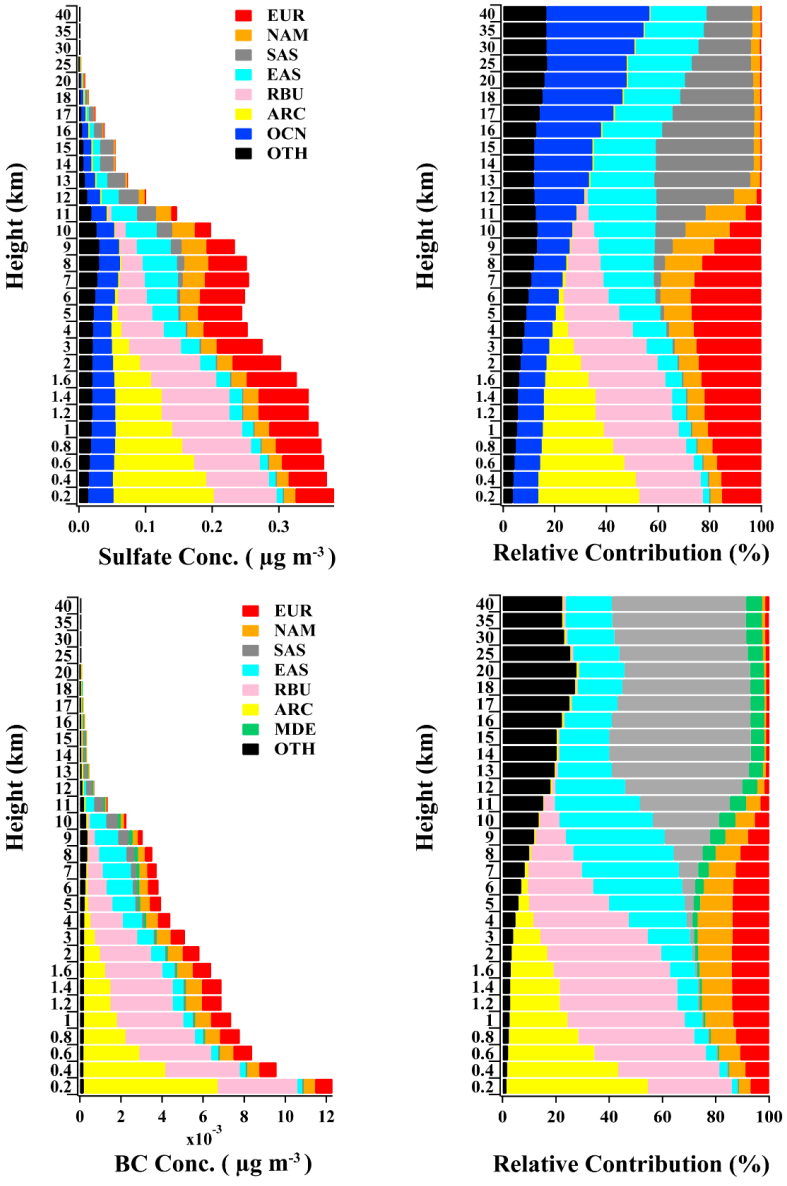


Figure 6. Annual mean vertical profile of sulfate (top) and BC (bottom) concentrations (μg m^{-3}) over the Arctic contributed by the tagged source regions (left) and their relative contributions (right, %) during 1980–2018. Sources with annual burden contributions less than 5% are combined and shown as OTH.

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020

Atmospheric Chemistry and Physics Discussions

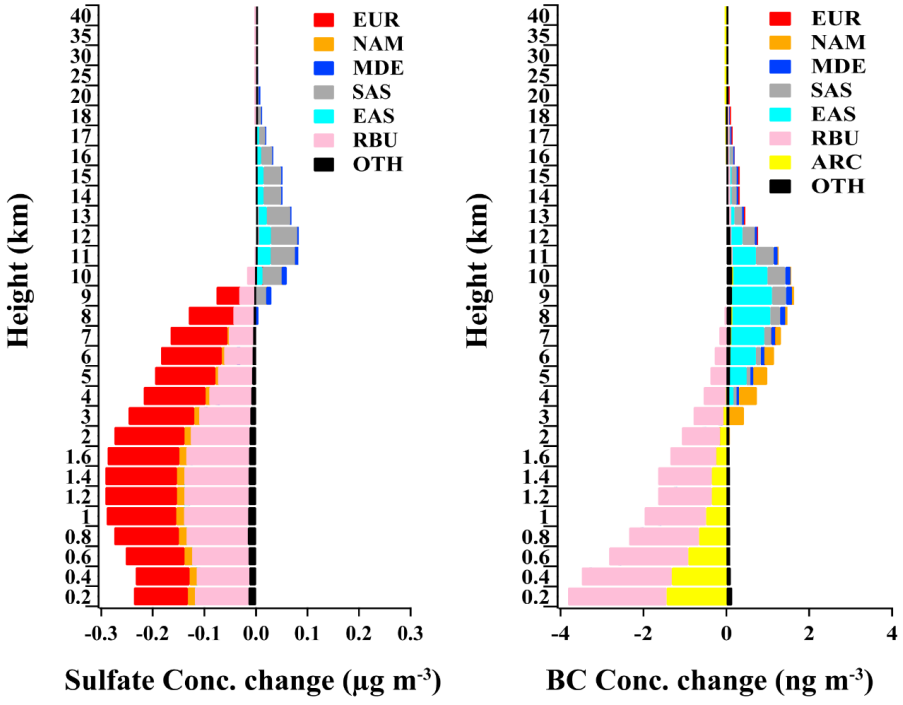


Figure 7. Changes in annual mean vertical profile of sulfate (μg m^{-3}, left) and BC (ng m^{-3}, right) concentrations over the Arctic contributed by the tagged source regions between 1980–1984 and 2014–2018.

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020

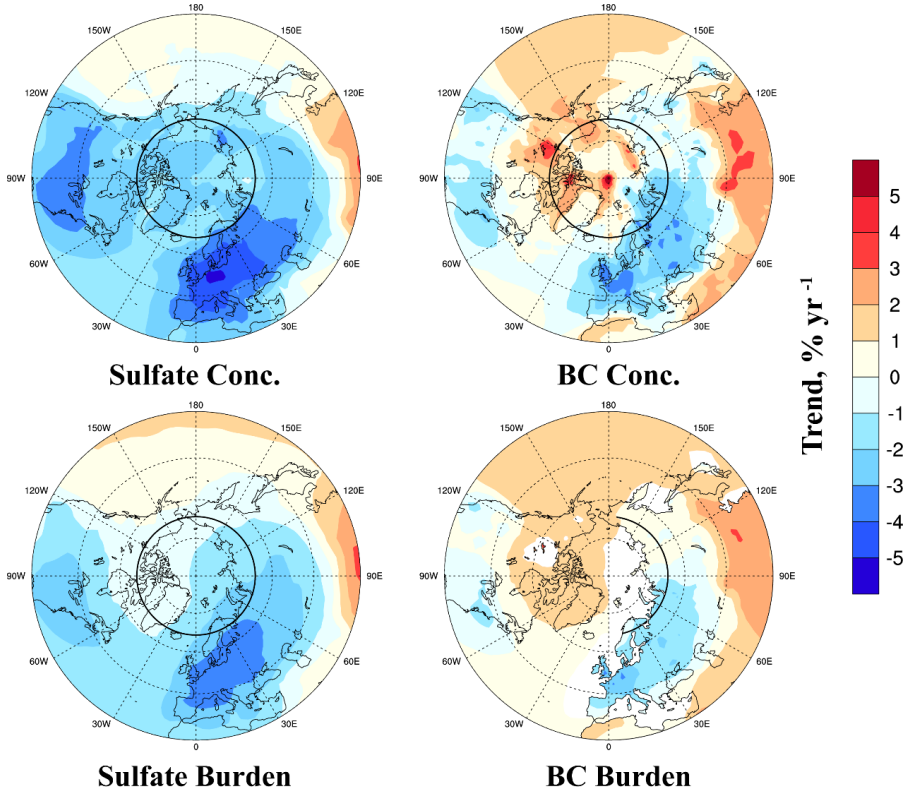


Figure 8. Spatial distribution of linear trends in annual mean sulfate (left) and BC (right) concentrations (% yr^{-1}) near the surface (top) and column burden (bottom) relative to the 39-year averages.

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020

Atmospheric Chemistry and Physics Discussions

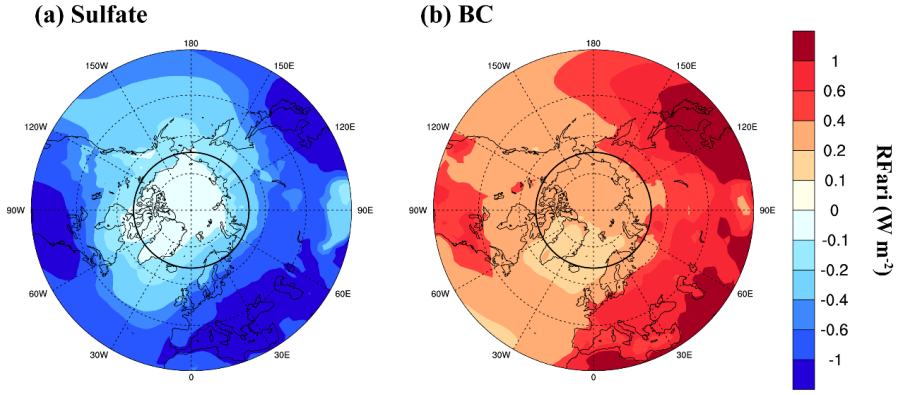


Figure 9. Spatial distribution of annual mean radiative forcing due to aerosol-radiation interactions (RF_{ari}) of (a) sulfate and (b) BC (W m^{-2}) at the TOA averaged over 1980–2018.

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020

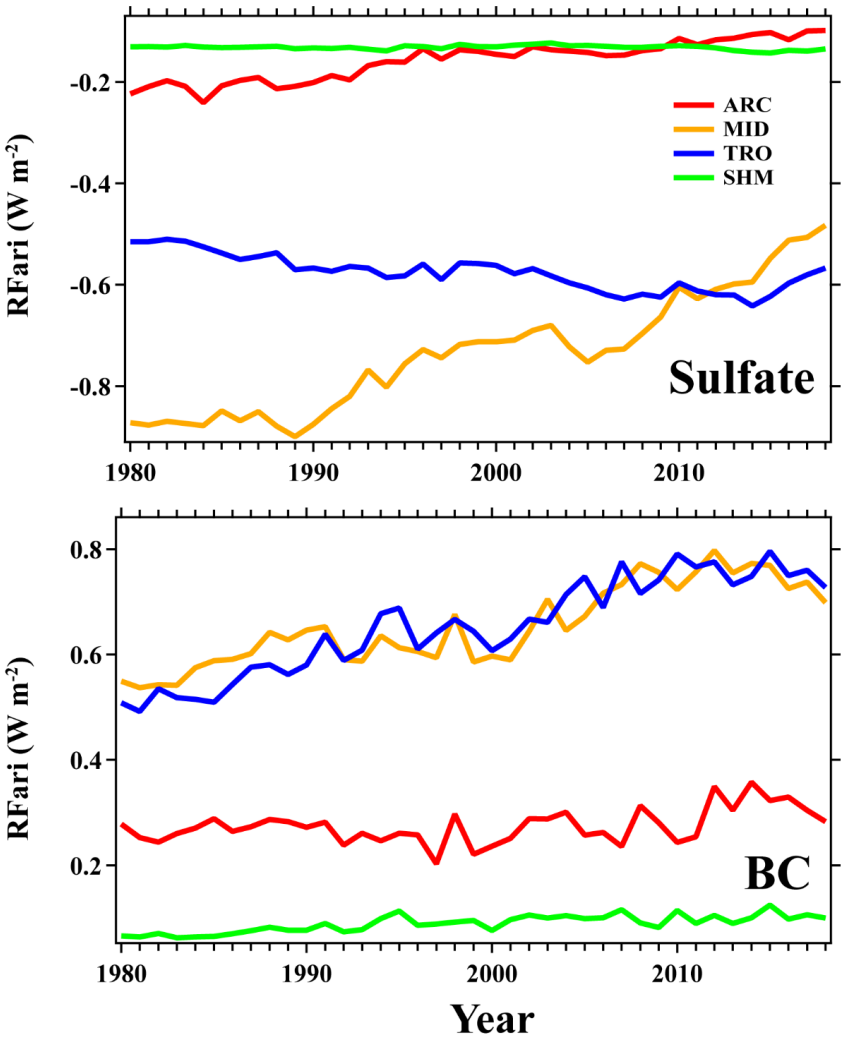
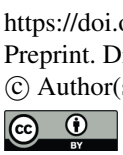


Figure 10. Time series (1980–2018) of annual radiative forcing due to aerosol-radiation interactions (RF_{ari}, W m^{-2}) of sulfate and BC over the Arctic (ARC, 66.5°N–90°N), Northern Hemisphere mid-latitudes (MID, 28°N–66.5°N), tropics (TRO, 28°S–28°N) and Southern Hemisphere (SHM, 90°S–28°S).

https://doi.org/10.5194/acp-2020-3
Preprint. Discussion started: 5 February 2020



Atmospheric Chemistry and Physics Discussions

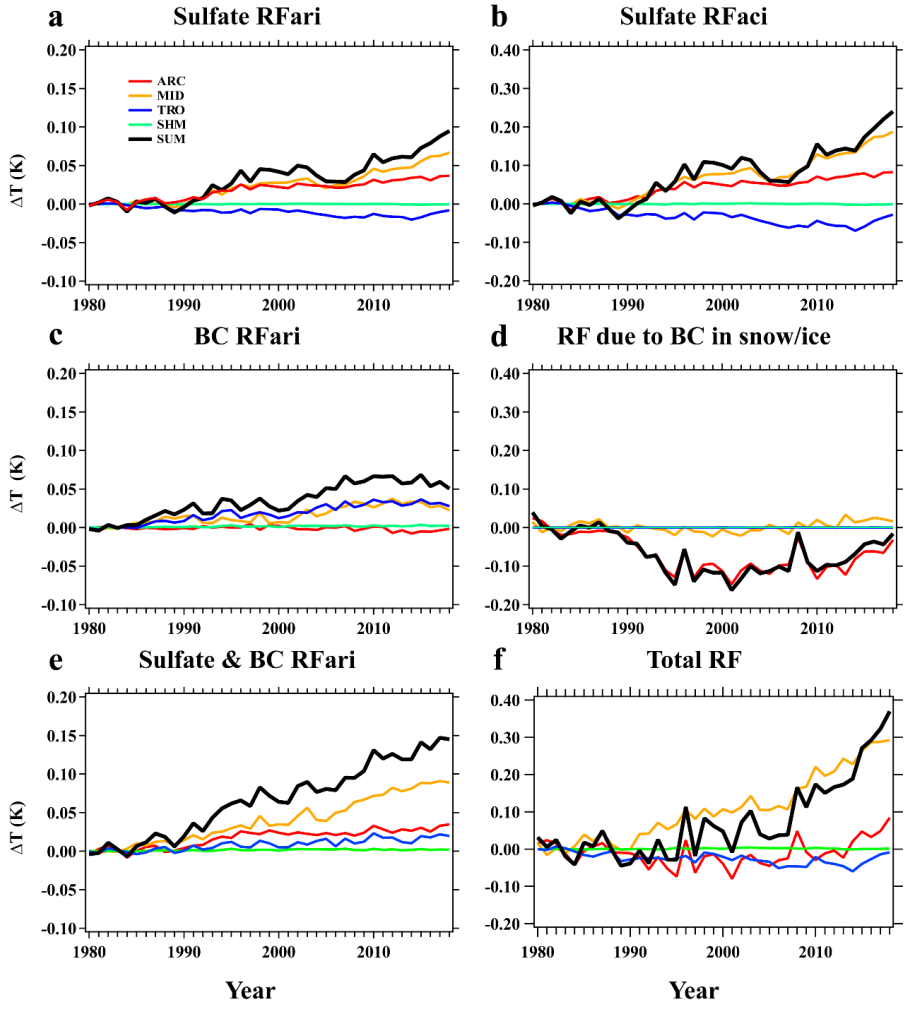


Figure 11. Time series (1980–2018) of the estimated response in surface temperatures (K) to the change in radiative forcing due to the aerosol-radiation interactions (RF_{ari}) of (a) sulfate, (c) BC, and (e) sum of sulfate and BC RF_{ari}, (b) radiative forcing due to aerosol-cloud interactions (RF_{aci}) of sulfate, (d) radiative forcing (RF) due to BC in snow/ice, (f) sum of all RF in each latitude bands and the sum of them (SUM).