**Manuscript # acp-2020-3**

**Responses to Referee #1**

Source attribution of Arctic aerosols is a relevant scientific topic within the scope of ACP. The authors present their data in a structured way and the figures are clear. However, before consider acceptance, I recommend the authors work more on presenting their results in light of related work.

We thank the reviewer for all the insightful comments. Below, please see our point-by-point response (in blue) to the specific comments and suggestions and the changes that have been made to the manuscript, in an effort to take into account all the comments raised here.

1. How does this study contribute to new knowledge in the field? What do you contribute that is different (model/data set/time period)? I would highlight this in the abstract, introduction and conclusion.

Response:

Thanks for the suggestion. Many studies have examined possible mechanisms that can explain the recent Arctic warming, but the quantitative importance of these mechanisms is still on debate. Among these mechanisms, some are related to roles of aerosols in changing the Arctic temperature. Shindell and Faluvegi (2009) found that aerosols may have warmed the Arctic surface due to emission reductions during 1976-2010. Breider et al. (2017) estimated that emission reductions in anthropogenic aerosols during 1980–2010 had contributed to a net warming at the Arctic surface by +0.27 ± 0.04 K using the GEOS-Chem model, which is consistent with our results. However, they did not take into consideration of the radiative forcing from aerosol-cloud interactions and deposition of BC to snow and ice surfaces. Navarro et al. (2016) presented simulations with an Earth system model and showed that the reduction in European $SO_2$ emission over 1980–2005 has caused an Arctic warming by 0.5 K on annual average as a result of the enhanced poleward heat transport, which is larger than our estimates likely due to different emissions and models used here and in Navarro et al. (2016).

Different from the emission perturbation method that was often used in previous studies, 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 affecting changes in the Arctic surface temperature from 1980 to 2018. All aerosol radiative impacts are considered including aerosol-radiation and aerosol-cloud interactions, as well as black carbon deposition on snow and ice-covered surfaces. We quantitatively showed that the combined total effects of sulfate and BC produced an Arctic surface warming of +0.297 K, explaining approximately 20% of the observed Arctic warming. We have now highlighted these in the various components of the manuscript.

2. Your conclusions are not new (but it is still very important to test what others have done!), but I would then add, 'as also shown in…etc etc. For instance, it have been shown in other studies that the declining emissions in Europe and the collapse of the Soviet Union are the main reasons why we see declining trends in the Arctic and that emissions from Asia contribute to higher level aerosols in the high-Arctic.

Response:

Thanks for the suggestion. We have now included such context for our conclusions as follows: "Previous studies also pointed out that, in April, BC showed a high concentration in the mid-troposphere of the Arctic, mainly due to the effect of Asian anthropogenic aerosols that are transported to the Arctic through warm conveyor belt (Wang et al., 2011). Evidence from aircraft and ground-based measurements showed that eastern and southern Asia source regions contributed the most to the BC concentration in the Arctic mid-troposphere, while northern Asia dominated the contribution to the Arctic surface BC (Abbatt et al., 2019)." And "Similar to our findings, Breider et al. (2017) found that the simulated decrease in aerosol optical depth in the Arctic from 1980 to 2010 was driven by a strong decrease in aerosol loading at lower altitudes due to the emission changes in West Eurasia, Russia and North America and an increase in aerosols at higher altitudes resulting from the changes in emissions in regions such as South Asia and East Asia."

3. I would also compare your numbers with other studies. Do they differ from other studies or do they support other findings? If different; try to explain why.

Response:

Thanks for the suggestion. The atmospheric 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, which is similar to the values of 0.1~0.6 $Wm^{-2}$ estimated in previous studies (Koch and Hansen, 2005; Flanner et al., 2009; AMAP, 2011; Bond et al., 2011; Samset et al., 2014; Wang et al., 2014).

Shindell et al. (2008) studied the sensitivity of simulated Arctic aerosol concentrations to emissions perturbations in 2001 and found that European emissions contributed to Arctic sulfate concentrations near the surface and at 500 hPa by 73% and 51%, respectively. East Asia has the largest contribution at 250 hPa, reaching 36%, which is consistent with our results. Based on simulations of a chemical transport model, Fisher et al. (2011) concluded that West Asia emissions dominated wintertime Arctic sulfate concentration, with contributions between 30% and 45%.

Using the GEOS-Chem model, Breider et al. (2017) estimated that emission reductions in anthropogenic aerosols during 1980–2010 had contributed to a net warming at the Arctic surface by +0.27 ± 0.04 K, which is consistent with our results. However, they did not take into consideration radiative forcing from aerosol-cloud interactions and deposition of BC to snow or ice surfaces. Navarro et al. (2016) presented simulations with an Earth system model and showed that the reduction in Europe $SO_2$ emission over 1980–2005 has caused the Arctic warms by 0.5 K on annual average as a result of the enhanced poleward heat transport, which is larger than our estimates likely due to different emissions and models used here and in Navarro et al. (2016).

We have included these comparisons in the manuscript.

4. The authors use sensitivity factors to estimate the temperature response to the declining trends. This method needs to be explained in Methods along with uncertainties.

Response:

Thanks for the suggestion. 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). The change in equilibrium temperature response is defined as $\Delta T = \sum_{j=LAT} \lambda_j * \Delta RF_j$ . Δ represents the difference of the annual mean of a variable for a specific year compared to the average during 1980–1984 in this study. RF is radiative forcing due to aerosol-radiation or aerosol-cloud interactions associated with sulfate or black carbon. LAT represents latitudinal bands over the Arctic (60°N–90°N), Northern Hemisphere mid-latitudes (28°N–60°N), tropics (28°S–28°N) and Southern Hemisphere (90°S–28°S). Many studies used these climate sensitivity factors to estimate the Arctic temperature responses using RF calculated from different models (e.g., Sand et al., 2016). However, we note that, since the λ values were calculated with a different climate model (NASA-GISS), the estimated Arctic equilibrium temperature response based on these factors could be biased.

5. Can you please add a description in Methods on how BC and sulfate are treated in the model? Aging, mixing etc.

Response:

Thanks for the suggestion. We have now added a description as follows. Mass and number concentrations of sulfate particles are predicted for the three lognormal modes (i.e., Aitken, accumulation, and coarse modes) of the three-mode modal aerosol module (Liu et al., 2012) in CAM5. Aerosols are internally mixed in the same aerosol mode and then externally mixed between modes. Within each mode, sulfate is internally mixed with primary/secondary organic matter, BC, mineral dust, and/or sea salt. BC is mixed with other aerosol species (e.g., sulfate, POA, SOA, sea salt, and dust) in the accumulation mode immediately after being emitted into the atmosphere without considering explicit aging processes.

Specific comments by line number:

Title: You are only looking at BC and SO4, so I would change 'aerosols' to reflect that + specify surface warming, and not just warming.

Thanks for the suggestion. We have now modified the title to "Source attribution of Arctic black carbon and sulfate aerosols and associated Arctic surface warming during 1980–2018."

L23: Wouldn't a decrease in BC, at least hypothetically, lead to a cooling?

That's likely true for the Arctic local BC alone. To avoid confusion, this sentence has been revised as follows: "Observations show that the concentrations of Arctic sulfate and black carbon (BC) aerosols have declined since the early 1980s. Previous studies have reported that reducing sulfate aerosols potentially contributed to the recent rapid Arctic warming."

L32: You need to specify that you have calculated the surface temperature response using sensitivity factors (and not by running a climate model).

We have now added "By using climate sensitivity factors, …".

L42: What other regions do you refer to here? Most aerosols are emitted NH mid lats?

The other regions refer to latitudinal bands: Arctic (ARC, 60°N–90°N), tropics (TRO, 28°S–28°N) and Southern Hemisphere (SHM, 90°S–28°S). Aerosols over any region can influence Arctic surface temperature through changing radiative fluxes or poleward heat transport based on the climate sensitivity factors. The mid-latitude region of the Northern Hemisphere is close to the Arctic and changes in aerosols over this region affect Arctic temperature through enhancing poleward heat transport. This warming effect is stronger than impacts of aerosols over other latitudinal bands.

L140: What kind of aerosol-cloud interaction are included in the model?

Aerosols interact with stratiform clouds through two-moment microphysics, in which the nucleation of stratiform cloud droplets is based on the scheme of Abdul-Razzak and Ghan et al. (2000). Although aerosols have no microphysical impact on convective clouds, the ambient temperature and convection can be affected by BC-induced atmospheric heating. We have added this description in the Methodology section.

L197: Where in the Arctic are those emissions mostly from? I would assume northern Russia?

Time series (1980–2018) of absolute and relative contributions of emissions from major source regions to the simulated annual mean near-surface sulfate and BC concentrations averaged over the Arctic is shown in Figure 5. It's correct that source regions near the Arctic (e.g., Europe and Russia) are the main contributors to the near-surface concentrations of Arctic sulfate and BC.

L216: Could you be more specific on where the Kevo site is located besides close to western Eurasia?

We have now revised the sentence to reflect this: "The Kevo site (69°N, 27°E), which is close to Western Eurasia, is the only site that has both sulfate and BC data for more than 30 years."

L217: Can you split these two sentences; one for bc and one for sulfate so it is easier to follow?

Following the suggestion, we have split the sentences for BC and sulfate: "At this site, the simulated sulfate in spring and summer decreased at a rate of -3.18% and -1.92% per year, respectively, which are similar to -4.37% and -3.26% per year from observations. The decreasing rates of BC in spring and summer were -2.89% and -1.74%, respectively, that are also consistent with the observed values of - 3.01% and - 2.82%."

L257: Could you remind us which regions those are?

The remaining source regions are 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), Pacific-Australia-New Zealand (PAN), Antarctic (ANT), and Non-Arctic/Antarctic Ocean (OCN). We have now included such information in the revised text.

L280: this is the first time you report concentrations in ug/m3 decrease and not % decrease. Can you add the total concentration number as well, so we can relate the number?

Following the suggestion, we have now revised the text as follows:

"Relative to the average of 0.447 μg/m$^3$ during 1980–1984, the simulated annual sulfate concentration over the Arctic has a decrease of 42.8% (0.191 μg/m$^3$) in 2014–2018 (Table 1). Sulfate concentration shows a considerable decreasing trend from 1980 to 2000, which then slows 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% (0.083 μg/m$^3$) and 18.8% (0.084 μg/m$^3$) of the decline of the Arctic sulfate concentrations, respectively. The change in emissions from Central Asia and North America, respectively, explains 1.6% (0.007 μg/m$^3$) and 3.4% (0.015 μg/m$^3$) 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% (3.7 ng/m$^3$ relative to the 1980–1984 average of 16.1 ng/m$^3$) in 2014–2018, mainly due to the reductions in emissions originating from the Arctic and Russia, which lead to 9.32% (1.5 ng/m$^3$) and 14.91% (2.4 ng/m$^3$) of the decrease (Table 1)."

L313: 'during'? How is this calculated? First and last 5 years?

We have revised it to "averaged over 1980–2018".

L329: What is a moderate value?

We have revised the text as follows: "Within the Arctic (60°N–90°N), the magnitude of sulfate $RF_{ari}$ decreases from -0.21 Wm$^{-2}$ in 1980–1984 to -0.10 Wm$^{-2}$ in 2014–2018, indicating a warming effect in the Arctic from the local sulfate change."

L332: this is the first time you mention the tropical region?

Yes. To estimate the relative roles of regional aerosol trends in affecting the Arctic warming, we looked into the temporal variation of annual mean radiative forcing of sulfate and BC in different latitudinal bands during 1980–2018. The four latitudinal bands considered in this study are Arctic (60°N–90°N), Northern Hemisphere mid-latitudes (28°N–60°N), tropics (28°S–28°N) and Southern Hemisphere (90°S–28°S).

L348: I would decrease the number of significant figures for these temperature response numbers, as the uncertainties are much higher.

We agree with the reviewer that the uncertainties associated with these numbers are likely high, but the number of digits after the decimal point is kept same for all the numbers here for consistency.

L394: Can you list these references you refer to here?

Many studies have examined possible mechanisms that can explain the recent Arctic warming, but the quantitative importance of these mechanisms is still on debate (e.g., Breider et al., 2017; Navarro et al. 2016).

L400: 'to some extent' seem vague.

We have revised the text as follows: "Considering that the model underestimates the magnitude of sulfate and BC concentrations, the estimated impact on Arctic temperature from sulfate and BC could be even larger if the model were able to accurately reproduces the measurements in the Arctic."

L408: Increase compared to what?

We have revised it to "Compared to the annual mean concentrations during 1980–1984".

Figure 1: it is hard to see the letters/dots representing the observation sites. Could another plot be made in this figure, zooming in on the Arctic (90-60N) and only showing the stations for example?

Following the suggestion, we have now revised the Figure 1 to zoom in to the Arctic for a better display of the observational sites. Please see below.

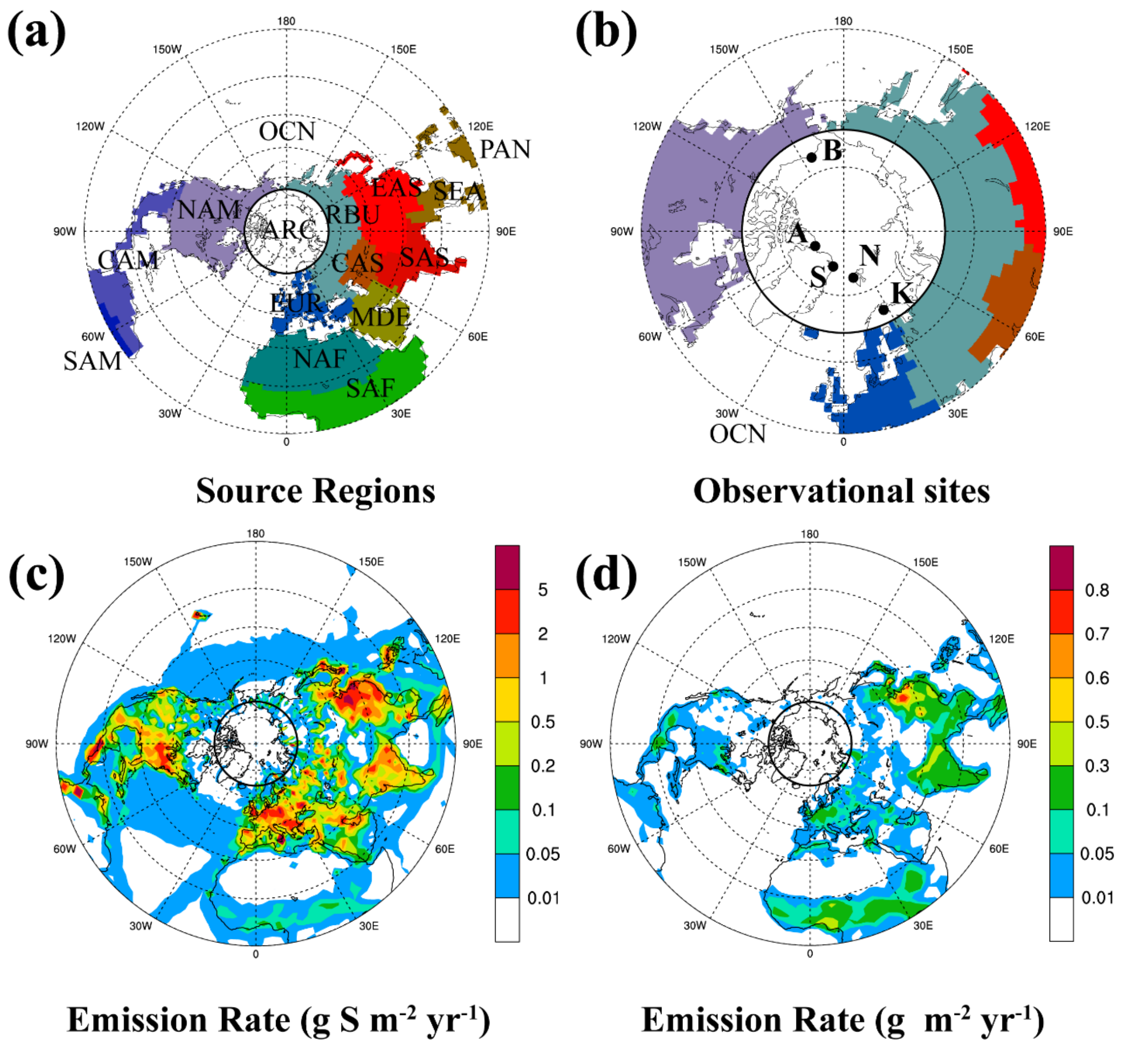


**Figure 1.** (a) Sixteen anthropogenic source regions (Europe (EUR), North America (NAM), Central America (CAM), South America (SAM), North Africa (NAF), South Africa (SAF), the Middle East (MDE), Southeast Asia (SEA), Central Asia (CAS), South Asia (SAS), East Asia (EAS), Russia-Belarus-Ukraine (RBU), Pacific-Australia-New Zealand (PAN), the Arctic (ARC), Antarctic (ANT), and Non-Arctic/Antarctic Ocean (OCN)). Dots in (b) mark 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). Spatial distribution of annual mean (c) $SO_2$ (g S $m^{-2}$ $yr^{-1}$) and (d) BC (g C $m^{-2}$ $yr^{-1}$) emissions averaged over 1980-2018. The thick black circles mark the Arctic (66.5°N–90°N).

Reference:

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**Manuscript # acp-2020-3**

**Responses to Referee #2**

This study uses source apportionment method to study the changes Arctic BC and Sulfate concentration, and the contributions from worldwide 16 other regions. They also performed sensitivity analysis to discuss the contribution of Arctic warming from the different source regions.
In general, I think the paper has an interesting theme. However, the method is not well presented, and the discussion is not well structured neither. The paper heavily focusses on the model results, and was not strong to make adequate discussions on why the simulated results happen.
We thank the reviewer for all the insightful comments. Below, please see our point-by-point response (in blue) to the specific comments and suggestions and the changes that have been made to the manuscript, in an effort to take into account all the comments raised here.

Main comment:
I suggest the authors reorganize the abstract from L32-43: think about the order of discussing the sulfate/BC radiative forcing changes, local vs long-range transport, temperature changes from aerosol-direct and indirect effects.
Response:
Following the suggestion, we have now revised this part of the abstract as follows: "Within the Arctic, sulfate reductions caused a TOA warming of 0.11 and 0.25 W $m^{-2}$, respectively, through aerosol-radiation and aerosol-cloud interactions. While the changes in Arctic atmospheric BC has little impact on local radiative forcing, the decrease of BC in snow/ice led to a net cooling of 0.05 W $m^{-2}$. By applying climate sensitivity factors for different latitudinal bands, global changes in sulfate and BC during 2014–2018 (with respect to 1980–1984) exerted a +0.088 K and 0.057 K Arctic surface warming, respectively, through aerosol-radiation interactions. Through aerosol-cloud interactions, the sulfate reduction gave an Arctic warming of +0.193 K between the two time periods. The weakened BC effect on snow/ice albedo led to an Arctic surface cooling of –0.041 K. The changes in atmospheric sulfate and BC outside the Arctic totally produced an Arctic warming of +0.25 K, the majority of which is due to the mid-latitude changes in radiative forcing. 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 through enhanced poleward heat transport. The combined total effects of sulfate and BC produced an Arctic surface warming of +0.297 K, explaining approximately 20% of the observed Arctic warming since the early 1980s."

It has been known that there are very large discrepancies for the emissions in China from MEIC emission inventory and CMIP6 (Paulot et al., 2018). Comment how this discrepancy could affect the main results.

Reference: Paulot, F., Paynter, D., Ginoux, P., Naik, V., and Horowitz, L. W.: Changes in the aerosol direct radiative forcing from 2001 to 2015: observational constraints and regional mechanisms, Atmos. Chem. Phys., 18, 13265–13281, https://doi.org/10.5194/acp-18-13265-2018, 2018.

Response:

Thanks for bringing up this issue. In our simulations of 1980–2018, we used both the CMIP6 historical emissions for 1980-2014 and emission scenario (SSP2-4.5) interpolated 2015-2018. Over China, the decline of aerosols emissions since 2011 is not well represented in the CMIP6 historical anthropogenic emissions, compared to the MEIC emission inventory (Paulot et al., 2018). Emissions of $SO_2$ and BC from China in SSP2-4.5 show declines since 2014, which is consistent with MEIC emissions. However, the decrease of CMIP6 $SO_2$ and BC emissions over China by 39% and 0.5%, respectively, in year 2017 compared to 2010 is less than the corresponding magnitude, 62% and 27%, in MEIC emission inventory. We have now included this point in the discussion section as follows: "Previous studies have reported large discrepancies of aerosol and precursors emissions in China between MEIC (Multi-resolution Emission Inventory for China) and CMIP6 emission inventories (e.g., Paulot et al., 2018). The CMIP6 emissions dataset shows similar decreasing trends in anthropogenic $SO_2$ and BC emissions over China since 2011 as in the MEIC inventory (Fig. S3). However, the decrease of CMIP6 anthropogenic $SO_2$ and BC emissions by 39% and 0.5%, respectively, in 2017 compared to 2010 is less than the corresponding magnitude of 62% and 27% in MEIC (Zheng et al., 2018). It indicates that the increase in aerosol contribution from East Asia during the recent decade and its impact on Arctic surface temperature could be overestimated in this study."

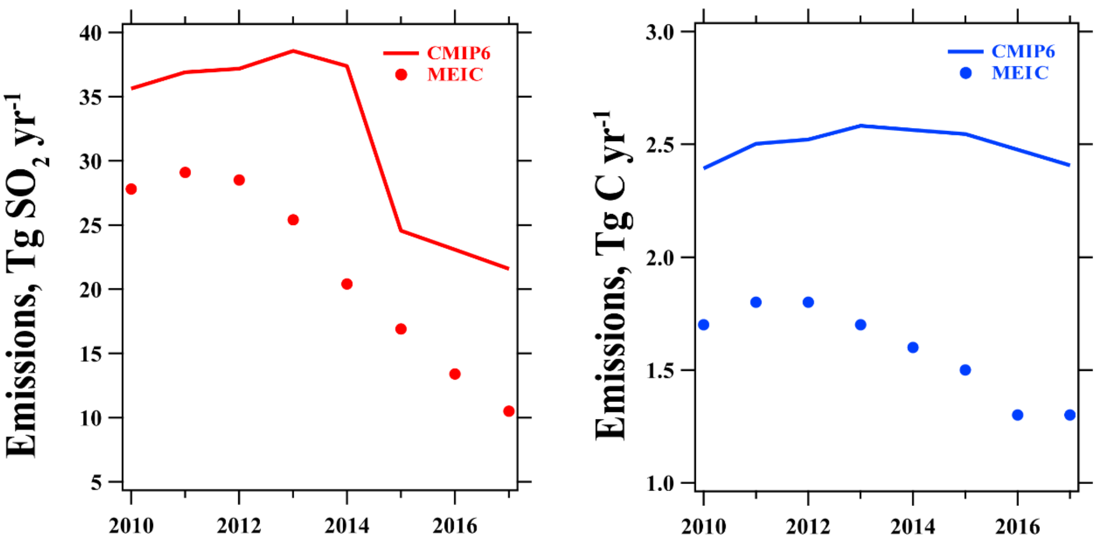

Figure S3. Annual anthropogenic emissions of $SO_2$ and BC in China from CMIP6 (solid lines) and MEIC (dotted lines).

Beginning from section 3, when the authors discuss the trends analysis, I did not find anywhere how the authors performed the trend analysis, as well as the significance test. Those are very basic concepts when we discuss trend analysis. A few example: line 245-line 248; line 251-252, and Table 2, Fig. 8.

Response:

Thanks for the suggestion. We have now included statistical test results in Table 2 and Figure 8. All trend values mentioned in that paragraph are statistically significant at the 95% confidence level. We have added this sentence to the manuscript.

Line 269-270: when the authors discuss the "largest contribution of East and South Asia", does the authors mean East and South Asia contributes most at this altitude compared with other regions, or this altitude is where East and South Asia contributes most for their contributions at different altitudes? As a matter of fact, I think these several paragraphs are terribly written (line 264-290). Keep in mind that, when you talk about the contribution, you are comparing between different source regions as well as the altitudes. I highly suggest the authors reorganize these several paragraphs.

Response:

We have now revised these paragraphs as follows to avoid the confusion: "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 and BC concentrations in the Arctic. Different source regions have very distinct vertical distributions of their contributions. Below 1 km, Arctic local emissions account for the majority of Arctic sulfate and BC concentrations. For BC and sulfate located between 1 km and 5 km, emissions from Russia are the major sources. Above 8 km, East Asia and South Asia are the major source regions of the Arctic aerosol concentrations, which is consistent with results using other models (e.g., Shindell et al., 2008). Arctic and Russia have their maximum absolute contributions at 0.2 km and 1.4 km, respectively. Europe and North America have their maximum absolute contributions around 2 km. The contribution of East Asia and South Asia increases with the increase of altitude, reaching their maximum contribution values at 8 km and 11 km, respectively.

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. Below 6 km, due to the effective emission reduction, the contribution from Europe and Russia to the Arctic sulfate 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. Between 10–15 km, contributions from South Asia and East Asia increased at the upper troposphere, 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 troposphere 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. Similar to sulfate, BC concentrations contributed by East Asia and South Asia increased in the high altitudes, mainly due to increased emissions in these two regions, offsetting the decrease in column burden owing to the reduced loading in the lower atmosphere."

Editorial comments: Line 35: explain what "61%" is compared to.

Response:

It is a comparison between 1980–1984 and 2014–2018. We have now clarified it in the text.

Line 38: the snow/ice albedo effect from BC refers to local or other source regions?

Response:

Here, the snow/ice albedo effect from BC refers to both local and other source regions. We have followed the suggestion in main comments to reorganize the abstract to avoid confusion as such.

Line 98: add from which year for the 2-3% changes.

Response:

Following the suggestion, we have now revised the sentence to "Based on the chemical transport model (GEOS-Chem) simulations, Breider et al. (2017) found that annual sulfate and BC concentrations decreased by 2–3% per year over the Arctic during 1980-2010."

Line 122: change "observational" to "observation"

Response:

Following the suggestion, we have now revised the sentence to "Sulfate and BC concentrations from the CAM5-EAST model and observations at remote Arctic stations are compared."

Line 153: EAST was already defined.

Response:

Deleted.

Line 181-182: Technically, neither Fig 1 nor Fig 2 showed the emission changes from "1980-2010" "from the 16 source regions".

Response:

Figure 1 shows the spatial distribution of annual mean $SO_2$ and BC emissions averaged over 1980-2018 from the 16 source regions and Figure 2 shows time series of annual anthropogenic $SO_2$ and BC emissions of major tagged source regions and other regions (OTH, including ANT,CAM, CAS, MDE, NAF, PAN, SAM,SEA, and SAF/NAM). In order to better see the time series of annual emissions of other regions (OTH) individually, we have added the time series these emissions in the supplementary materials (Fig. S1), which is also shown below.

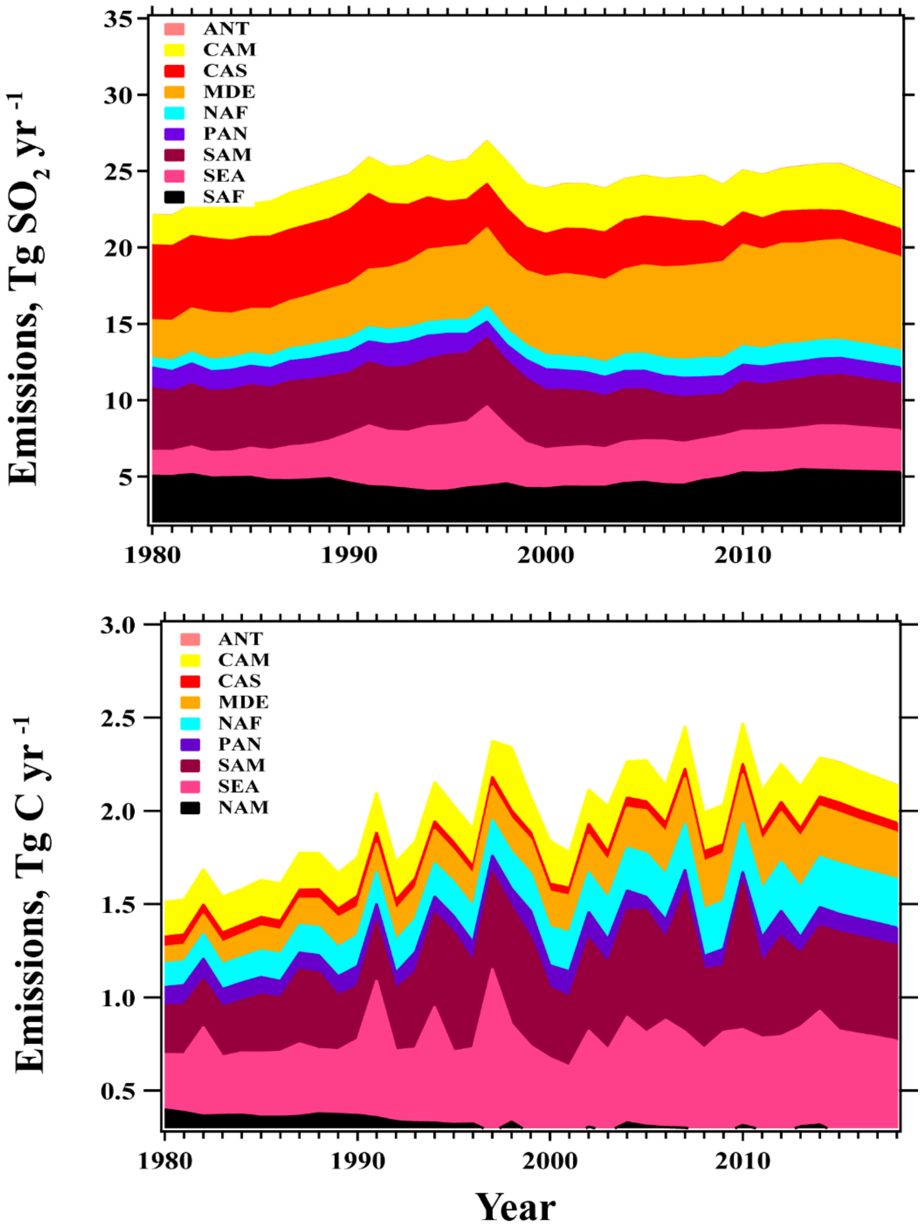


Figure S1. Time series of annual anthropogenic (top) $SO_2$ (Tg $SO_2$ $yr^{-1}$) and (bottom) BC (Tg C $yr^{-1}$) emissions from other regions of Fig. 2 individually.

Line 209-210: Please put reference or show precipitation/wet deposition plots to confirm the theory.

Response:

We have now revised the sentence to "According to previous CAM5 studies on aerosol wet removal and long-range transport, the model underestimates aerosol concentrations in spring, likely due to biases in parameterizations of convective transport and wet scavenging of aerosols (Bond et al., 2013; Liu et al., 2011; Wang et al., 2013; Qian et al., 2014; Yang et al., 2018a)."

Line 212-213: I thought local BC reduction by 38% in Artic are pretty high. So are you sure the BC concentration changes are dominated by the emission changes from other source region? Meanwhile, I got different conclusion from Fig. 5 as the ARC clearly dominated the total BC changes.

Response:

The decrease here refers to the BC concentration change at the Arctic sites (especially Kevo) that are strongly influenced by non-local sources. Kevo is close to western Eurasia. The drop in BC after 1988 at Kevo is attributed to the emission reduction resulting from the economic contraction in former Soviet states and eastern bloc countries at that time. The concentration change shown in Fig. 5 is the average over the entire Arctic (66.5°N–90°N).

Line 256: how did the "+/- 1-3%" come from? It looks like uncertainty range to me.

Response:

We have now revised it to "Sources in Europe, North America, and East Asia account for less than 4% of the changes in Arctic near-surface BC concentration."

Line 262-263: the authors conclude to reduce local sources in the Arctic to control the sulfate and BC. Can the authors give some specific suggestions on the sectors which the local source should be reduced?

Response:

The $SO_2$ and BC emissions from individual sectors in the Arctic are shown below. The industry and energy sectors account for the majority of local sources in the Arctic (Fig. S4). Although this is not the focus of our research, reducing the emissions of industry and energy sectors may be effective for the reduction of local sulfate and BC concentrations in the Arctic. We have now added this analysis in the manuscript.

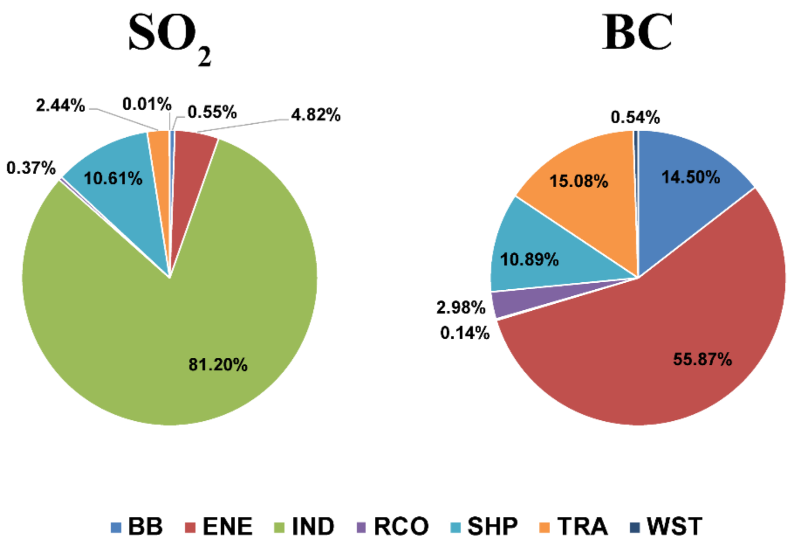


Figure S4. Annual mean of $SO_2$ and BC emissions from individual sectors in the Arctic during 2010-2014. BB: biomass burning, ENE: energy, IND: industry, RCO: residential, SHP: international shipping, TRA: transportation, WST: waste treatment.

Line 272-273: If I am reading the plots right, I think Europe also has largest contribution for both sulfate and BC below 2km, compared with Arctic? That being said, I still can not figure out what the authors refer to when they say “largest contribution”.

Response:

We have now revised this part of the text substantially, taking into consideration of this comment. Please see our response to the fourth main comment.

Line 287-288: Again not clear how to comprehensive the “increasing trend” contributed by East Asia and South Asia. Also, the authors have a theory why East Asia and South Asia are larger high altitudes, any references or evidences? Please explain.

Response:

We have now revised this sentence to explain: “Similar to sulfate, BC concentrations contributed by East Asia and South Asia increased in the high altitudes (Breider et al. 2017, Fisher et al., 2011; Qi et al., 2017; Sharma et al., 2013; Stohl, 2006), mainly due to increased emissions in these two regions, offsetting the decrease in column burden owing to the reduced loading in the lower atmosphere.”

Line 317-322 Here is redundant to discuss the radiative forcing changes in other NH regions since this is not the focus of this paper. Remember the paper’s interest is on the transport of other source region on the reception region (Arctic).

Response:

Although we focus on the Arctic local changes, aerosols over other regions outside the Arctic can also affect the Arctic climate through changing poleward heat transport, which is also an important factor to consider for the Arctic temperature change in the following part of the manuscript.

Line 329-330: The authors previously showed that the sulfate concentration changes over Arctic are dominated by other source regions than local. So why the authors conclude the local sulfate change for the radiative forcing increases?

Response:

Here the local change refers to the Arctic as a receptor rather than a source region. It is compared with the impact of remote changes. We apply Arctic climate sensitivity factors for sulfate and BC over the Arctic, mid-latitudes of the Northern Hemisphere, tropics and Southern Hemisphere, separately, obtained from Sand et al. (2016) and Shindell and Faluvegi (2009), to calculate the recent Arctic surface temperature change related to the variations in sulfate and BC radiative forcings over the different latitude bands during 1980–2018. This is different from the previous sections of source regions.

Line 330-333: again these are not relevant to this study. I think it is Ok if the authors want to compare the radiative forcing changes in Artic with other regions for the past 4 decades, but not necessary to distract the main point of the paper.

Response:

The aerosol-induced meridional gradient of temperature can also influence the Arctic climate by changing poleward heat transport. For example, BC at midlatitudes may increase the transport of heat into the Arctic by heating the atmosphere locally and increasing the meridional temperature gradient. This impact is directly related to changes in global aerosol emissions, so we believe it is an important factor to analyze and compare with changes within the Arctic.

Line 356-358: please explain why the BC changes over mid-latitude and tropics have positive climate effect and expand to Arctic?

Response:

The positive climate effect over mid-latitude and tropics is due to an increase of BC during 2014–2018 relative to 1980–1984. As we explained above, the remote impact from warming in mid-latitude and tropics on the Arctic is mainly through changes in the poleward heat transport. We didn't simulate such remote effects on Arctic temperature directly. Instead 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). The change in equilibrium temperature response is defined as $\Delta T = \sum_{j=LAT} \lambda_j * \Delta RF_j$. Δ represents the difference of the annual mean of a variable for a specific year compared to the average during 1980–1984 in this study. RF is radiative forcing due to aerosol-radiation or aerosol-cloud interactions associated with sulfate or black carbon. LAT represents latitudinal bands over the Arctic (60°N–90°N), Northern Hemisphere mid-latitudes (28°N–60°N), tropics (28°S–28°N) and Southern Hemisphere (90°S–28°S). Many studies used these climate sensitivity factors to estimate the Arctic temperature responses using RF calculated from different models (e.g., Sand et al., 2016). However, we note that, since the λ values were calculated with a different climate model (NASA-GISS), the estimated Arctic equilibrium temperature response based on these factors could be biased. We have now added this to the Methodology section.

Figures:

In Fig. 2 title, add the references that abbreviations for the regions could be found in Fig. 1

Response:

Done as suggested.

Fig. 3 I saw crosses, triangles, rectangles and dotted circles which are not explained in the legend. In the stacked contour plots, I think the authors refer light green for the Arctic? The Y axis for plots St. Nord Ny-Alesund and Kevo seems not right to me.

Response:

We have now revised the figure caption to clarify on these issues:

'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 colors represent modeled contributions from the Arctic (blue) and non-Arctic anthropogenic source region (green). The observations denoted by solid black circles are obtained from European Monitoring and Evaluation Programme and World Data Centre for Aerosols database (http://ebas.nilu.no) and Breider et al. (2017). Black triangles at Ny-Alesund for the period 1980–1981 show mean observations from Heintzenberg and Larssen (1983). Black diamond at Ny-Alesund in summer shows median non-sea-salt sulfate concentration from Maenhaut et al. (1989). Open circles in the spring for Ny-Ålesund are March–April mean values (Sirois and Barrie, 1999). Note that the vertical coordinates use logarithmic scales.'

In Fig 5 and figures below, the authors only show a list of the source regions, not all of them. I suspect that's because other regions' contribution to BC and sulfate in Arctic are very negligible? If so, how much is it? Is it magnitude level smaller than the CAS to sulfate, and EAS to BC? Also, how did the authors make the relative contribution equal to 100% if not all the regions included? I would also suggest the authors to reorganize the plot, so maybe the contour plots will be seen as smaller to largest, or vice-visa.

Response:

Figure 5 shows the time series (1980–2018) of absolute (left, μg $m^{-3}$) and relative (right, %) contributions of emissions from the major source regions to the simulated annual mean near-surface sulfate and BC concentrations averaged over the Arctic. The remaining source regions with annual contributions less than 3% are combined and shown as OTH (other regions, in figure S2). The total relative contribution considers all source regions, including the OTH, and equals to 100%. We have reorganized the contours to show an order of largest to smallest contributors except OTH.

Fig 9: this study's focus is on Arctic. This fig is not easy to distinguish the spatial patterns of temperature changes in Arctic.

Response:

As we explained above, RF changes in the lower latitudes are also important to affect the Arctic temperature. We agree that the spatial patterns of the small RF within the Arctic are not distinguishable, but the main purpose of this figure is not to focus on the Arctic RF variation.

References:

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.

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.

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Shindell, D., and Faluvegi, G.: Climate response to regional radiative forcing during the twentieth century, Nat. Geosci., 2, 294-300, https://doi.org/10.1038/ngeo473, 2009.

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**Manuscript # acp-2020-3**

**Responses to Referee #3**

Review of "Source attribution of Arctic aerosols and associated Arctic warming trend during 1980-2018" by Ren et al.

This paper presents a modelling study of the impacts of changing $SO_4$ and BC on the Arctic atmospheric composition, radiative forcing, and temperature. Modelled and measured $SO_4$ and BC are presented in the Arctic from 1980-2018 at a handful of surface measurement sites. A tagged version of CAM5 is used to quantify the source contributions from different continental geographic regions to the Arctic BC and $SO_4$ concentrations both at the surface and in the vertical column. The paper present interesting results that are important for understanding the rapidly warming Arctic. The authors conclude that about 20% of Arctic warming can be attributed to the combination of BC and $SO_4$.

We thank the reviewer for all the insightful comments. Below, please see our point-by-point response (in blue) to the specific comments and suggestions and the changes that have been made to the manuscript, in an effort to take into account all the comments raised here.

I suggest only the following minor revisions below before publishing:

lines 130-131: is there a primary reference for CAM5 and CESM that you can reference here?

Response:

Thanks for the suggestion. We have now added the primary reference for CESM as follows: "The global aerosol-climate model CAM5, which is the atmospheric component of the earth system model CESM (Community Earth System Model, Hurrell et al., 2013) 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)."

lines 143-144: what is the source for the specified sea surface temperatures, sea ice concentrations, etc?

Response:

Sea surface temperatures and sea ice concentrations are created from the merged Reynolds/HADISST products, as described in Hurrell et al. (2008). Solar radiation and GHGs follow the CMIP6 configuration for AMIP-type of simulations. We have now included these details in the manuscript.

lines 209-210: was the modelled precipitation compared to measured precipitation? Was wet deposition of model validated against measurements?

Response:

The performance of CAM5 in aerosol wet deposition and transport to the Arctic has been specifically evaluated and improved in previous studies (e.g., Liu et al., 2011; Wang et al., 2013; Qian et al., 2014; Yang et al., 2018a. To address this comment and follow a suggestion from one of the other reviewers, we have revised the sentence to "According to previous CAM5 studies on aerosol wet removal and long-range transport, the model underestimates aerosol concentrations in spring, likely due to biases in parameterizations of convective transport and wet scavenging of aerosols (Bond et al., 2013, Liu et al., 2011, Wang et al., 2013; Qian et al., 2014; Yang et al., 2018a)."

Fig 5/line 241: it needs to be clarified that Fig 5 is the model average in the Arctic (>66.5 °N).

Response:

Following the suggestion, we have now revised the sentence to "The absolute and relative source contributions of emissions from the major source regions to the simulated annual mean near-surface sulfate and BC concentrations averaged over the Arctic (66.5°N–90°N) are shown in Fig. 5."

line 252: was that rise in BC seen in the observations? e.g., consistent with BC seen at Alert?

Response:

Yes, we have now revised the sentence to "Simulated Arctic BC concentration also shows a considerable decline before 2000, but a slight rise after 2000, which is consistent with the BC observations at Alert."

line 263: "in the Arctic" ... and Russia?

Response:

Yes, we have now revised the sentence to "To further reduce present-day or future aerosols in the Arctic, efforts can be made to control local sources in the Arctic as well as emissions from Russia."

line 316: is the effect of BC deposition on snow/reduction of albedo included in this? I think not because that effect is discussed later, but could clarify here that this value is just for atmospheric BC effect.

Response:

No, the effect of BC deposition on snow/reduction of albedo is not included in it. This value is for atmospheric BC effect only. We have now revised the text to "The Arctic sulfate exerts a negative $RF_{ari}$ primarily by scattering incoming solar radiation back into the space, with the forcing in a range of -0.4~0 $Wm^{-2}$. The atmospheric 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."

Section 5/line 400: Can you add some discussion as to how the model bias affects your conclusions? E.g. would your estimates of $SO_4$ and BC temperature impacts be greater or lesser if the model were corrected to accurately reflect the measurements?

Response:

Thanks for the suggestion. We have now revised the sentence to "Considering that the model underestimates the magnitude of sulfate and BC concentrations, the estimated impact on Arctic temperature from sulfate and BC could be even larger if the model were able to accurately reproduce the measurements in the Arctic."

Data availability: please add where the Arctic BC & $SO_4$ measurements can be found in this section (e.g., EBAS database link).

Response:

Added.

Figs 1-2, and 5-7: please make sure the regional colours are consistent in all of these plots. e.g., colour X for RBU, colour Y for EUR, etc, in all 5 figures the same.

Response:

We have now made the regional colors consistent in all plots.

Fig 3 (4): Clarify in the caption that the black is from measurements, and the blue and green are modelled. E.g., "Measured seasonal means are denoted by...". "Stacked contours represent the modelled Arctic..."

Response:

Thanks for the suggestion. We have now revised the figure caption to:

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 colors represent modeled contributions from the Arctic (blue) and non-Arctic anthropogenic source region (green). The observations denoted by solid black circles are obtained from European Monitoring and Evaluation Programme and World Data Centre for Aerosols database (http://ebas.nilu.no) and Breider et al. (2017). Black triangles at Ny-Alesund for the period 1980–1981 show mean observations from Heintzenberg and Larssen (1983). Black diamond at Ny-Alesund in summer shows median non-sea-salt sulfate concentration from Maenhaut et al. (1989). Open circles in the spring for Ny-Ålesund are March–April mean values (Sirois and Barrie, 1999). Note that the vertical coordinates use logarithmic scales.

Fig 3: why is Barrow not shown? Fig 4: why is St Nord not shown? Fig 5: specify that this is the Arctic (>66.5 °N) average. As mentioned above, use the same regional colour scheme here as in Fig 1(a) & Fig 2. Fig 6 & 7: match the regional colours to Fig 5.

Response:

The data of Barrow and St Nord sites are relatively scarce. We only selected sites with more than 20 observation samples.

Following the suggestion, the caption Figure 5 has been revised to “Time series (1980–2018) of absolute (left, μg $m^{-3}$) and relative (right, %) contributions of emissions from the major source regions to the simulated annual mean near-surface sulfate and BC concentrations averaged over the Arctic (66.5°N–90°N).

Fig 2, Fig 5, Fig 6 and Fig 7 have now been revised to use the same regional color scheme.

Reference:

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.

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# Source attribution of Arctic black carbon and sulfate aerosols and associated Arctic surface warming ~~trend~~ during 1980–2018

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## Abstract

Observations show that the concentrations of Arctic sulfate and black carbon (BC) aerosols have declined since the early 1980s~~, which~~. Previous studies have reported that reducing sulfate aerosols 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 affecting changes in the Arctic surface temperature ~~change over the past four decades (~~from 1980~~–~~ to 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.~~All aerosol radiative impacts are considered including aerosol-radiation and aerosol-cloud interactions, as well as black carbon deposition on snow and ice-covered surfaces. Within the Arctic, sulfate reductions caused a top-of-the-atmosphere (TOA) warming

of 0.11 and 0.25 W m$^{-2}$, respectively, through aerosol-radiation and aerosol-cloud interactions. While the changes in Arctic atmospheric BC has little impact on local radiative forcing, the decrease of BC in snow/ice led to a net cooling of 0.05 W m$^{-2}$. By applying climate sensitivity factors for different latitudinal bands, global changes in sulfate and BC during 2014–2018 (with respect to 1980–1984) exerted a +0.088 K and 0.057 K Arctic surface warming, respectively, through aerosol-radiation interactions. Through aerosol-cloud interactions, the sulfate reduction gave an Arctic warming of +0.193 K between the ~~first~~two time periods. The weakened BC effect on snow/ice albedo led to an Arctic surface cooling of –0.041 K. The changes in atmospheric sulfate and ~~last five years of 1980–2018~~BC outside the Arctic totally produced an Arctic warming of +0.25 K, the majority of which is due to the mid-latitude ~~emission change~~changes in radiative forcing. 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 ~~associated with~~through enhanced poleward heat transport~~ from the aerosol-induced stronger meridional temperature gradient.~~. The combined ~~aerosol~~total effects of sulfate and BC ~~together produce~~produced an Arctic surface warming of +0.297 K~~ during 1980–2018~~, explaining approximately 20% of the observed Arctic warming ~~during~~since the ~~same time period~~early 1980s.

## 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~~found that aerosols may have warmed the Arctic surface during 1976-2010 based on model sensitivity experiments. The aerosols that caused Arctic warming are not only from local emissions. ~~Previous studies~~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 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 understanding of their past and present impacts on the Arctic climate. In situ observations of sulfate and BC concentrations in the Arctic (e.g., at Alert, Barrow, Station Nord, and Zeppelin) have shown a declining trend since the 1980s (Gong et al., 2010; Heidam et al., 1999; Hirdman et al., 2010; Quinn et al., 2009; Sharma et al., 2004; Sharma et al., 2006; Sinha et al., 2017; Sirois and Barrie, 1999). Based on the chemical transport model (GEOS-Chem) simulations, Breider et al. (2017) found that annual sulfate and BC concentrations decreased by 2–3% per year over the Arctic. during 1980-2010. McConnell et al. (2007) presented a historical BC trend derived from ice-core records, showing that BC concentration had been declining steadily after the peak around 1910.

Source attribution analysis of atmospheric aerosols in the Arctic, which can help to understand aerosol trends, is extremely important for air pollution research. There is less local anthropogenic aerosol emission in the Arctic region than in polluted regions of the world. Pollutants in the Arctic are generally from mid-latitude areas by long-distance transport (Fisher et al., 2011; Wang et al., 2014). Recent studies have found that Arctic aerosols mainly originated from Eurasia, Southeast Asia, Siberia and North America (Fisher et al., 2011; Qi et al., 2017; Sharma et al., 2013; Stohl, 2006). The contribution of Eurasia to Arctic sulfate and BC aerosols concentration is dominant in the lower atmosphere, while South and Central Asia contributed the most at high altitudes (e.g., Wang et al., 2014). In general, Northern Europe and Russia, with large industrial emissions, are the main source region of Arctic BC aerosols in spring (Rahn et al., 1977; Rahn, 1981; Raatz and Shaw, 1984; Barrie, 1986; Koch and Hansen, 2005; Sharma et al., 2006; Stohl, 2006). Shindell et al. (2008) studied the sensitivity of simulated Arctic aerosol concentrations to emissions perturbations in 2001 and found that European emissions contributed to Arctic sulfate concentrations near the surface and at 500 hPa by 73% and 51%, respectively. East Asia has the largest contribution at 250 hPa, reaching 36%. Based on simulations of a chemical transport model, Fisher et al. (2011) concluded that West Asia emissions dominated wintertime Arctic sulfate concentration, with contributions between 30% and 45%. In the past few decades, anthropogenic emissions 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~~Sulfate and BC concentrations from the CAM5-EAST model and observations 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, Hurrell et al., 2013) 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 and number concentrations of sulfate~~, BC, primary organic aerosol (POA), second organic aerosol (SOA), mineral dust, and sea salt particles~~ are predicted ~~with a~~ for the three lognormal modes (i.e., Aitken, accumulation, and coarse modes) of the three-mode modal aerosol module~~. Aerosol number concentration is also predicted for each mode. Particles~~ (Liu et al., 2012) in ~~the different size modes~~ CAM5. Aerosols are ~~assumed to be externally mixed and~~ internally mixed in the same ~~mode~~aerosol mode and then externally mixed between modes. Within each mode, sulfate is internally mixed with primary/secondary organic matter, BC, mineral dust, and/or sea salt. BC is mixed with other aerosol species (e.g., sulfate, POA, SOA, sea salt, and dust) in the accumulation mode immediately after being emitted into the atmosphere without considering explicit aging processes. 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. Sea surface temperatures and sea ice concentrations are created from the merged Reynolds/HADISST products, as described in Hurrell et al. (2008). Solar radiation and GHGs follow the CMIP6 configuration for AMIP-type of simulations. 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.

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

We focus on the Arctic (66.5°N–90°N) as the receptor region in this study. According to source region definition of the Hemispheric Transport of Air Pollution model experiment phase 2 (HTAP2), sulfate and BC from 16 regions are tagged (Fig. 1): 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, hereafter Russia), Pacific-Australia-New Zealand (PAN), the Arctic (ARC), Antarctic (ANT), and Non-Arctic/Antarctic Ocean (OCN). Note that the OCN tag includes sources from oceans and volcanic eruptions.

## 2.3 Radiative Forcings and Temperature Response

Radiative forcing (RF) 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). Aerosols interact with stratiform clouds through two-moment microphysics, in which the nucleation of stratiform cloud droplets is based on the scheme of Abdul-Razzak and Ghan et al. (2000). Although aerosols have no microphysical impact on convective clouds, the ambient temperature and convection can be affected by BC-induced atmospheric heating. 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). ~~2.3~~The change in equilibrium temperature response is defined as $\Delta T = \sum_{j=LAT} \lambda_j * \Delta RF_j$. Δ represents the difference of the annual mean of a variable for a specific year compared to the average during 1980–1984 in this study. RF is radiative forcing due to aerosol-radiation or aerosol-cloud interactions associated with sulfate or black carbon. LAT represents latitudinal bands over the Arctic (60°N–90°N), Northern Hemisphere mid-latitudes (28°N–60°N), tropics (28°S–28°N) and Southern Hemisphere (90°S–28°S). Many studies used these climate sensitivity factors to estimate the Arctic temperature responses using RF calculated from different models (e.g., Sand et al., 2016). However, we note that, since the λ values were calculated with a different climate model (NASA-GISS), the estimated Arctic equilibrium temperature response based on these factors could be biased.

## 2.4 Aerosol and Precursor Emissions

In order to simulate the long-term temporal variations in aerosols, historical anthropogenic (Hoesly et al., 2018) and biomass combustion (van Marle et al., 2017) emissions of aerosols and precursor gases during 1980–2014 are used in the simulation following the CMIP6 (Coupled Model Intercomparison Project Phase 6) protocol. For the most recent years (2015-2018), yearly interpolated emissions from the ~~SSP2~~SSP (Shared Socioeconomic Pathways) 2-4.5 scenario are used, which is the modest scenario compared to other SSPs and is widely utilized in many MIPs (O'Neill et al., 2016~~) are used. Figures~~). Figure 1 and Figure 2 (Figure S1) 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~~values 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~~the emission 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~~,~~ and Russia ~~and the Arctic~~ decreased by 45.2~~%, 44.1~~% and ~~38.3~~44.1%, 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~~5 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 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~~According to previous CAM5 studies on aerosol wet removal and long-range transport, the model underestimates aerosol concentrations in spring, likely due to biases in ~~simulated precipitation and aerosol wet removal during the transport to high latitudes.~~parameterizations of convective transport and wet scavenging of aerosols (Bond et al., 2013; Liu et al., 2011; Wang et al., 2013; Qian et al., 2014; Yang et al., 2018a). 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~~,~~ (69°N, 27°E), 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 in spring and ~~BC in spring (~~summer~~)~~ decreased at a rate of -3.18% ~~(-1.92%)~~ and -~~2.89% (-~~1.~~74%)~~92% per year, respectively, which ~~is~~are similar to -4.37% ~~(-3.26%)~~ and -3.~~01% (-2.82%)~~26% 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,~~decreasing rates of BC in spring and summer were -2.89% and -1.74%, respectively, that are also consistent with the observed ~~decreasing trends~~values of -~~2.89% (-0.47%) per year.~~3.01% and -2.82%.

Observational data are very limited in the Arctic, especially the long-term 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 of emissions from the major source regions to the simulated annual mean near-surface sulfate and BC concentrations averaged over the Arctic (66.5°N–90°N) are shown in Fig. 5 and Fig. S2. 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 the average of 0.447 $\mu g/m^3$ during 1980–1984, the simulated annual sulfate concentration over the Arctic has a decrease of 42.8% (0.191 $\mu g/m^3$) in 2014–2018 (Table 1). Sulfate concentration shows a considerable decreasing trend from 1980 to 2000~~ and~~, which then slows 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% (0.083 $\mu g/m^3$) and -18.8% (0.084 $\mu g/m^3$) of the decline of the Arctic sulfate concentrations, respectively. The change in emissions from Central Asia and North America, respectively, explains -1.6% (0.007 $\mu g/m^3$) and -3.4% (0.015 $\mu g/m^3$) of the reduced concentration.

Simulated Arctic BC concentration also shows a considerable decline before 2000, but a slight rise after 2000~~.~~, which is consistent with the BC observations at Alert. Overall, the average concentration of BC in the Arctic had a decrease of 22.98% ~~in 2014–2018~~(3.7 ng/m$^3$ relative to the 1980–1984 average of 16.1 ng/m$^3$) in 2014–2018, mainly due to the reductions in emissions originating from the Arctic and Russia, which lead to 9.32% (1.5 ng/m$^3$) and 14.91% (2.4 ng/m$^3$) of the ~~concentration~~ decrease (Table 1). Sources in Europe, North America, and East Asia account for less than 4% of the changes in Arctic near-surface BC concentration~~ in range of ±1–3%.~~. The remaining source regions (Central America, South America, North Africa, South Africa, the Middle East, Southeast Asia, Central Asia, South Asia, Pacific-Australia-New Zealand, Antarctic, and Non-Arctic/Antarctic Ocean) have no substantial impact on the BC concentration in the Arctic (total contribution less than 2%) due to the 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~~local sources in the Arctic as well as emissions from Russia. The industry and energy sectors account for the majority of local sources in the Arctic (Fig. S4). Reducing the emissions of industry and energy sectors may be effective for the reduction of local sulfate and BC concentrations 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 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.~~Below 1 km, Arctic local emissions account for the majority of Arctic sulfate and BC concentrations. For BC and sulfate located between 1 km and 5 km, emissions from Russia are the major sources. Above 8 km, East Asia and South Asia are the major source regions of the Arctic aerosol concentrations, which is consistent with results using other models (e.g., Shindell et al., 2008). Arctic and Russia have their maximum absolute contributions at 0.2 km and 1.4 km, respectively. Europe and North America have their maximum absolute contributions around 2 km. The contribution of East Asia and South Asia increases with the increase of altitude, reaching their maximum contribution values at 8 km and 11 km, respectively. Previous studies also pointed out that, in April, BC showed a high concentration in the mid-troposphere of the Arctic, mainly due to the effect of Asian anthropogenic aerosols, that are transported to the Arctic through warm conveyor belt (Wang et al., 2011). Evidence from aircraft and ground-based measurements showed that eastern and southern Asia source regions contributed the most to the BC concentration in the Arctic mid-troposphere, while northern Asia dominated the contribution to the Arctic surface BC (Abbatt et al., 2019).

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~~Below 6 km, 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~~Between 10–15 km, contributions from South

Asia 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~~troposphere 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~~Similar to sulfate, BC concentrations contributed by East Asia and South Asia ~~show an increasing trend~~increased in the high altitudes~~,~~ (Breider et al. 2017, Fisher et al., 2011; Qi et al., 2017; Sharma et al., 2013; Stohl, 2006), mainly due to increased emissions in these two regions, offsetting the decrease in column burden owing to the reduced ~~concentration in the lower altitudes.~~loading in the lower atmosphere. Similar to our findings, Breider et al. (2017) found that the simulated decrease in aerosol optical depth in the Arctic from 1980 to 2010 was driven by a strong decrease in aerosol loading at lower altitudes due to the emission changes in West Eurasia, Russia and North America and an increase in aerosols at higher altitudes resulting from the changes in emissions in regions such as South Asia and East Asia.

~~Linear~~A linear regression approach is applied in order to analyze the 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~~South and ~~South~~East Asia, resulting in an insignificant change of total BC burden during 1980–2018. All trend values mentioned above are statistically significant at the 95% confidence level.

## 4. Aerosol ~~radiative~~ Radiative ~~forcing~~ Forcing and ~~associated~~ Associated Arctic ~~warming~~Warming

Both sulfate and BC influence the Arctic climate through perturbing atmospheric and 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~~averaged over 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~~. The atmospheric 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~~.~~, which is similar to the values of 0.1~0.6 $Wm^{-2}$ estimated in previous studies (Koch and Hansen, 2005; Flanner et al., 2009; AMAP, 2011; Bond et al., 2011; Samset et al., 2014; Wang et al., 2014). 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~~regional aerosol trends in affecting the Arctic warming, we ~~look~~looked into the temporal variation of annual mean ~~$RF_{ari}$~~ radiative forcing of sulfate and BC in different ~~latitude~~latitudinal bands during 1980–2018 (Fig. 10). Within the Arctic (~~66.5~~60°N–90°N), the ~~negative $RF_{ari}$~~magnitude of sulfate $RF_{ari}$ 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~~60°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

$Wm^{-2}$ to 0.74 $Wm^{-2}$ in the mid-latitudes and from 0.51 $Wm^{-2}$ to 0.76 $Wm^{-2}$ in the tropics, while the BC $RF_{ari}$ over the Arctic has no obvious change during this time period.

Systematic assessment of the impact of aerosols on Arctic warming since 1980s requires quantifying the Arctic temperature responses to changes in radiative forcing of different aerosol species over different regions. Here, we apply Arctic climate sensitivity factors, defined as the Arctic temperature response per unit ~~radiation~~radiative forcing, for each short-lived climate forcers over the Arctic, mid-latitudes of the Northern Hemisphere, tropics and Southern Hemisphere from Sand et al. (2016) and Shindell and Faluvegi (2009) to calculate the recent Arctic surface temperature change related to the variations in sulfate and BC radiative forcings over the different ~~latitude~~latitudinal bands during 1980–2018 (Fig. 11 and Table 3). This method has been widely adopted to examine the Arctic temperature response to aerosol forcings (e.g., Breider et al., 2017; Flanner, 2013; Sand et al., 2016; Shindell and Faluvegi, 2009; Yang et al., 2018c).

It is estimated that, between 1980–1984 and 2014–2018, changes in total $RF_{ari}$ of sulfate and BC produce a surface warming of +0.145 K over the Arctic, with +0.088 K (61%) contributed by the sulfate forcing change and the remaining explained by the BC forcing change. The sulfate-related Arctic warming is mainly due to the decrease in sulfate in mid-latitudes that enhances the temperature gradient between the mid-latitudes and Arctic, resulting in a strengthened meridional heat transport and, therefore, the Arctic warming of +0.059 K. The change in Arctic local $RF_{ari}$ of sulfate provides +0.035 K of the surface warming, while the forcing change in the tropics has a negligible influence on the Arctic temperature change. The Arctic temperature responses to increases in BC $RF_{ari}$ over the mid-latitudes and tropics are +0.029 K and +0.031 K, respectively, related to the enhanced poleward heat transport from the warming radiative impact in the mid-latitudes, while changes in the Arctic BC $RF_{ari}$ only exert a weak cooling of -0.005 K. Overall, the $RF_{ari}$ change over the mid-latitudes provides the strongest warming effect (+0.088K) to the Arctic compared to other latitude bands, owing to the aerosol-induced increase in the poleward heat transport.

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

Based on the Arctic climate sensitivities, impacts of changes in radiative forcing due to aerosol-cloud interactions ~~($RF_{aci}$)~~ of sulfate are also estimated. The sulfate $RF_{aci}$ provides an Arctic warming of +0.193 K between 1980–1984 and 2014–2018, with

+0.165 K contributed by the $RF_{aci}$ change over the mid-latitudes and +0.078 K resulting from the 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~~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)~~Different from the emission perturbation method that was often used in previous studies, in this study, the EAST was implemented in CAM5 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~~. Considering that the model underestimates the magnitude of sulfate and BC ~~to some extent~~concentrations, the estimated impact on Arctic temperature from sulfate and BC could be even larger if the model were able to accurately reproduce the measurements in the Arctic.

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 the annual mean concentrations during 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 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 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.

Many studies have examined possible mechanisms that can explain the recent Arctic warming, but the quantitative importance of these mechanisms is still on debate (e.g., Breider et al., 2017; Navarro et al. 2016). Among these mechanisms, some are related to roles of aerosols in changing the Arctic temperature. Shindell and Faluvegi (2009) found that aerosols may have warmed the Arctic surface due to emission reductions during 1976-2010. Breider et al. (2017) estimated that emission reductions in anthropogenic aerosols during 1980–2010 had contributed to a net warming at the Arctic surface by +0.27 ± 0.04 K using the GEOS-Chem model, which is consistent with our results. However, they did not take into consideration of the radiative forcing from aerosol-cloud interactions and deposition of BC to snow and ice surfaces. Navarro et al. (2016) presented simulations with an Earth system model and showed that the reduction in European $SO_2$ emission over 1980–2005 has caused an Arctic warming by

0.5 K on annual average as a result of the enhanced poleward heat transport, which is larger than our estimates likely due to different emissions and models used here and in Navarro et al. (2016).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. Previous studies have reported large discrepancies of aerosol and precursors emissions in China between MEIC (Multi-resolution Emission Inventory for China) and CMIP6 emission inventories (e.g., Paulot et al., 2018). The CMIP6 emissions dataset shows similar decreasing trends in anthropogenic $SO_2$ and BC emissions over China since 2011 as in the MEIC inventory (Fig. S3). However, the decrease of CMIP6 anthropogenic $SO_2$ and BC emissions by 39% and 0.5%, respectively, in 2017 compared to 2010 is less than the corresponding magnitude of 62% and 27% in MEIC (Zheng et al., 2018). It indicates that the increase in aerosol contribution from East Asia during the recent decade and its impact on Arctic surface temperature could be overestimated in this study. 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 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).

***Data availability.***

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

***Competing interests.***

The authors declare that they have no conflict of interest.

***Author contribution*.**

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

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

| | **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% |

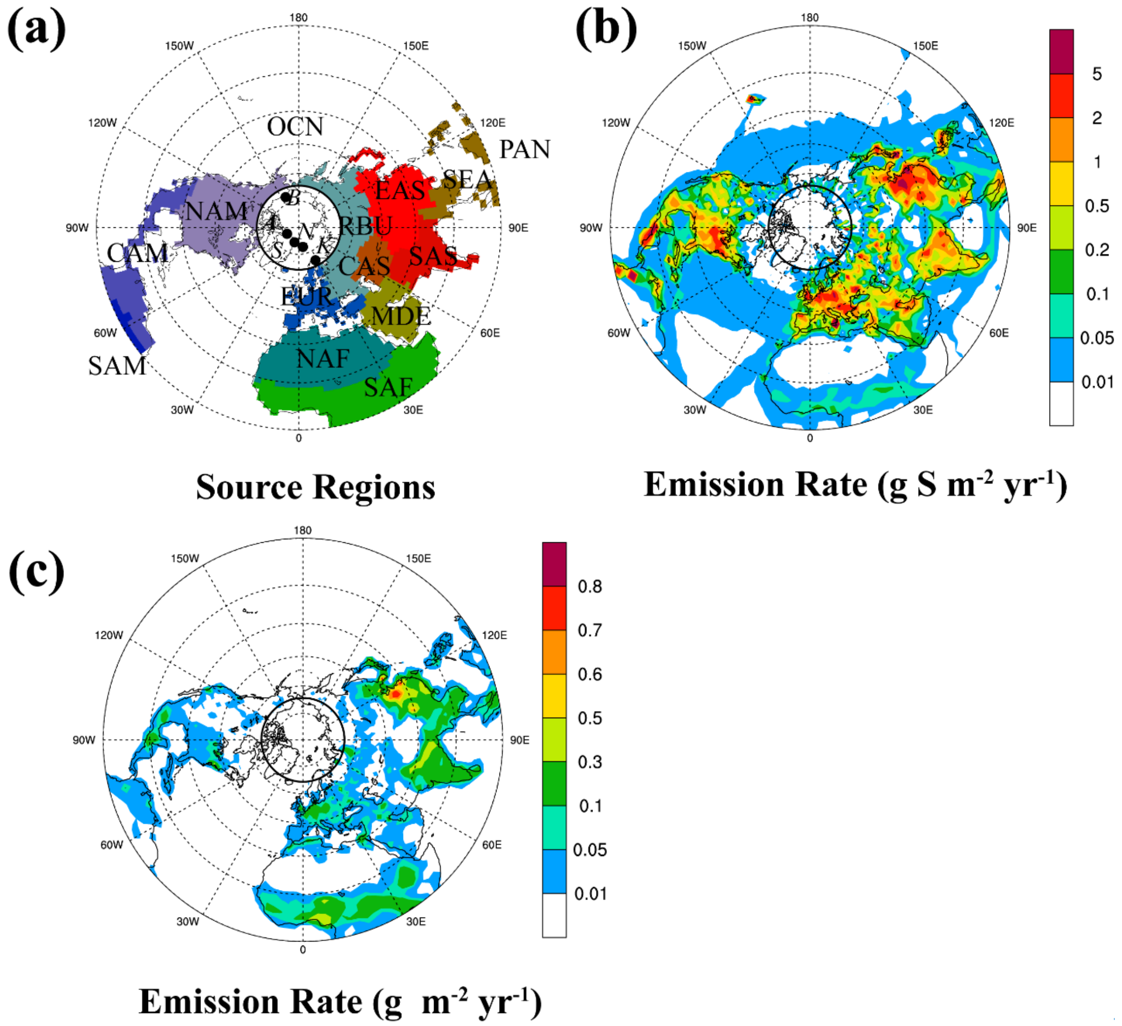
**Table 2.** Trends in annual mean sulfate and BC concentrations (% per decade) in surface air and in the column contributed by 16 anthropogenic source regions during 1980–2018 relative to the 39-year averages of total concentrations. The boldface values are statistically significant at the 95% confidence level based on F-test.

| 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%** |

**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 ($\lambda$, 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 $\lambda$ 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).


(a)
OCN
PAN
SEA
EAS
NAM
RBU
CAM
SAS
CAS
EUR
MDE
NAF
SAF
SAM
Source Regions
(b)
5
2
1
0.5
0.2
0.1
0.05
0.01
Emission Rate (g S m-2 yr-1)
(c)
0.8
0.7
0.6
0.5
0.3
0.1
0.05
0.01
Emission Rate (g m-2 yr-1)

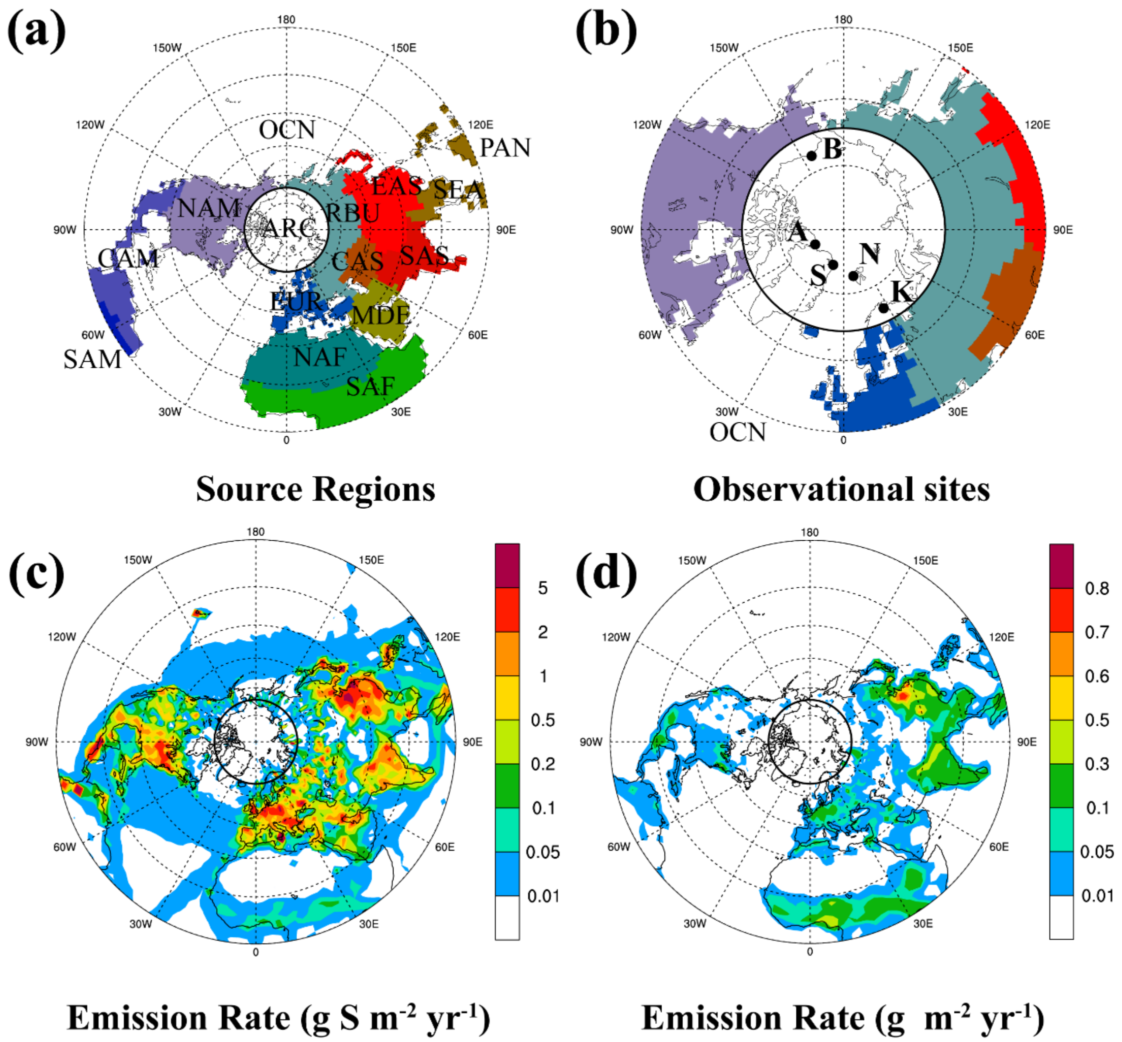
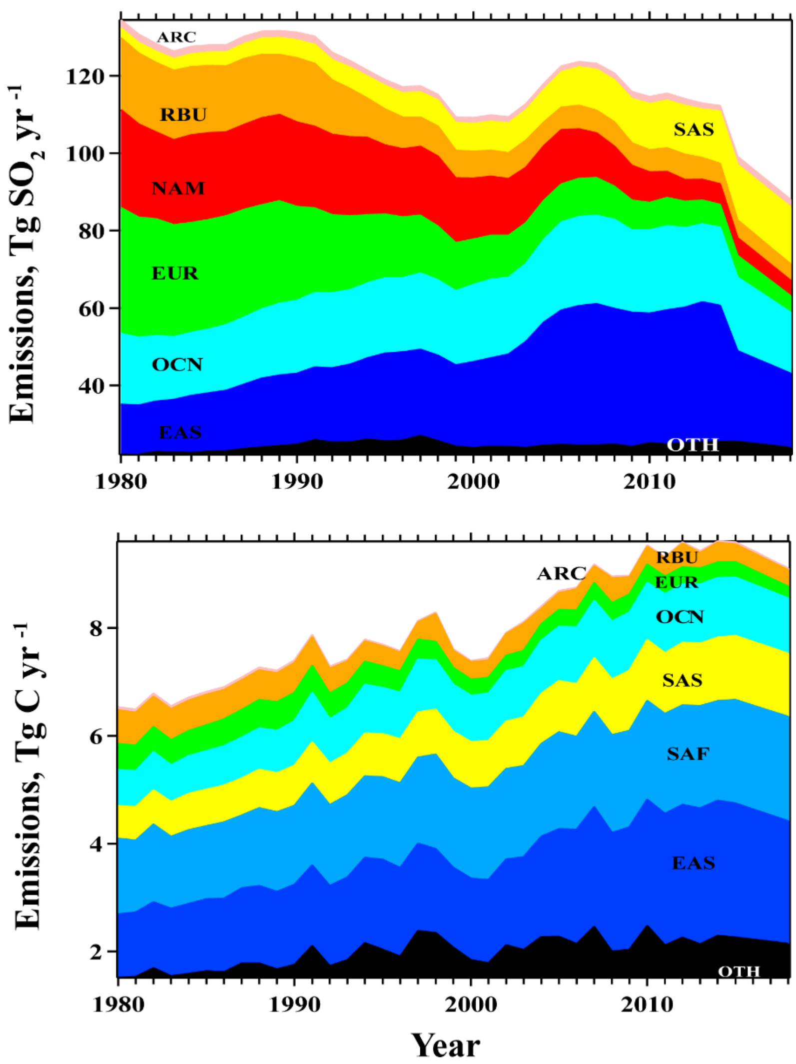



**Figure 1.** (a) Sixteen anthropogenic source regions (Europe (EUR), North America (NAM), Central America (CAM), South America (SAM), North Africa (NAF), South Africa (SAF), the Middle East (MDE), Southeast Asia (SEA), Central Asia (CAS), South Asia (SAS), East Asia (EAS), Russia-Belarus-Ukraine (RBU), 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~~Dots in (b) mark 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). Spatial distribution of annual mean (c) $SO_2$ (g S m$^{-2}$ yr$^{-1}$) and (d) BC (g C m$^{-2}$ yr$^{-1}$) emissions averaged over 1980-2018. The thick black circles mark the Arctic



Emissions, Tg $SO_2$ yr $^{-1}$
ARC
RBU
NAM
EUR
OCN
EAS
SAS
OTH
120
100
80
60
40
1980
1990
2000
2010
Emissions, Tg C yr $^{-1}$
ARC
RBU
EUR
OCN
SAS
SAF
EAS
OTH
8
6
4
2
1980
1990
2000
2010
Year


(66.5°N–90°N).

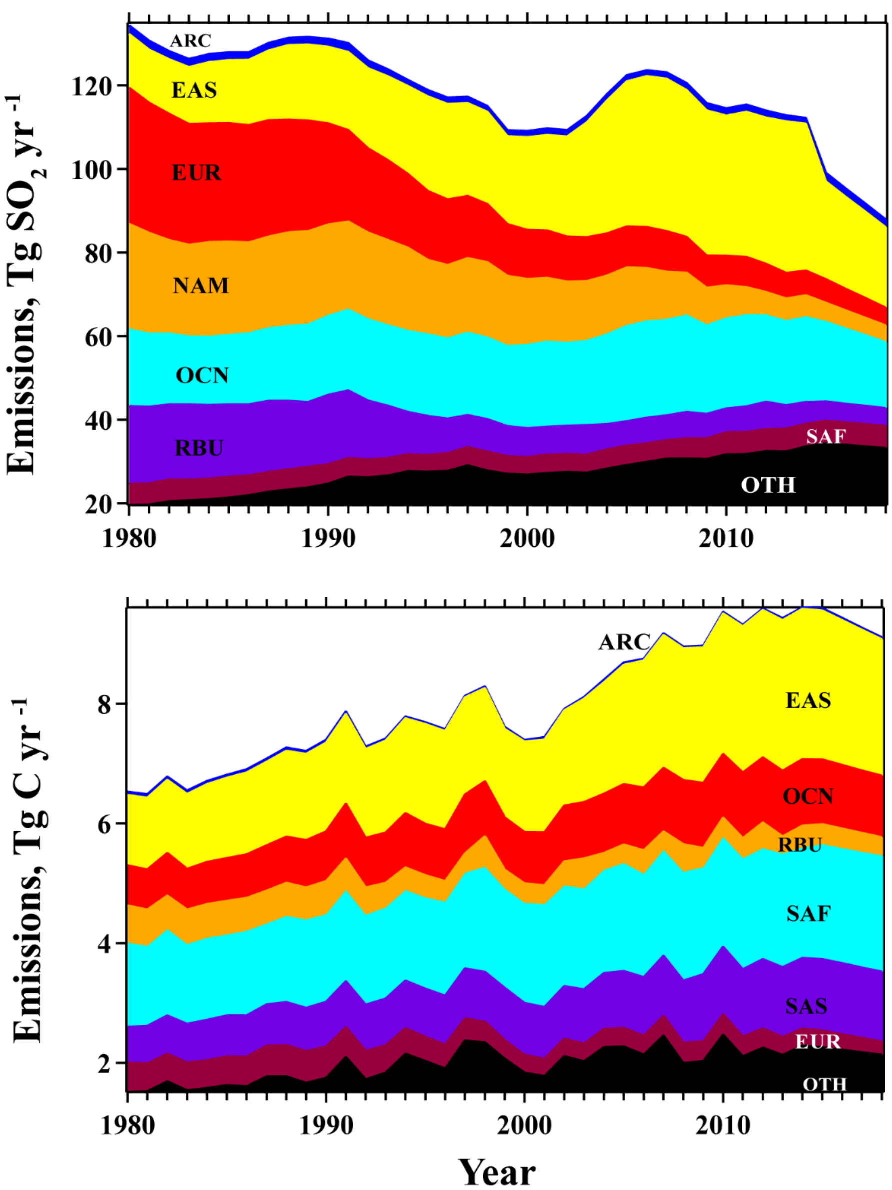



**Figure 2.** Time series of global total anthropogenic emissions of (top) $SO_2$ (Tg $SO_2$ $yr^{-1}$) and (bottom) BC (Tg C $yr^{-1}$), classified by key anthropogenic source regions. Emissions from other regions (OTH) include those of ANT, CAM, CAS, MDE, NAF, PAN, SAM, SEA, and SAF/NAM can be found in figure S1. Abbreviations for the regions can be found in Fig. 1.

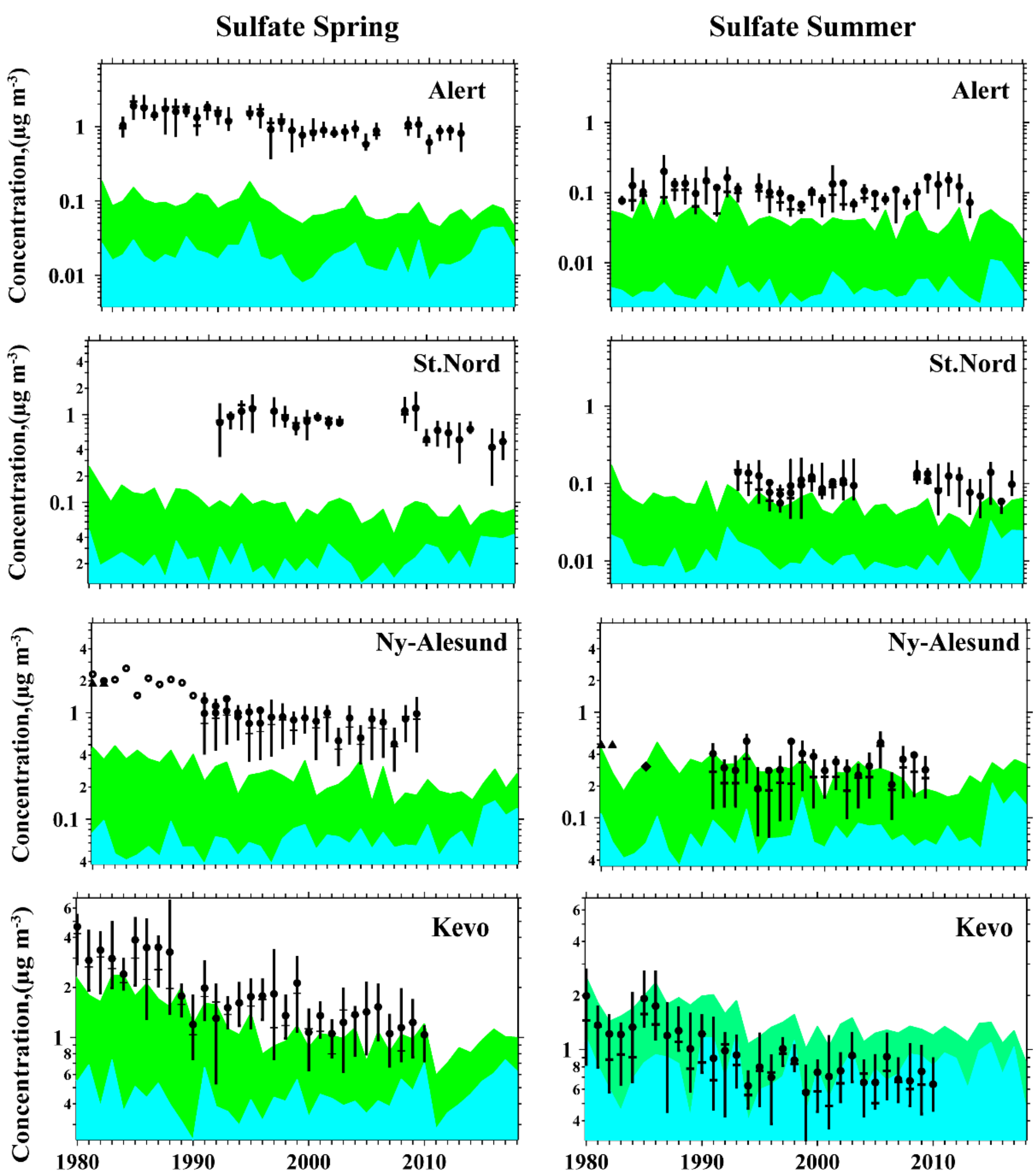



**Figure 3.** Surface concentrations of sulfate aerosols (μg $m^{-3}$) in spring (March–May) and summer (June–August) at four locations (Alert, Station Nord, Ny-Alesund, Kevo) in the Arctic during 1980–2018. Seasonal means are denoted by solid black circles, medians as short horizontal bars, and the 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.~~ Stacked colors represent modeled contributions from the Arctic (blue) and non-Arctic anthropogenic source region (green). The observations denoted by solid black circles are obtained from European Monitoring and Evaluation Programme and World Data Centre for Aerosols database (http://ebas.nilu.no) and Breider et al. (2017). Black triangles at Ny-Alesund for the period 1980–1981 show mean observations from Heintzenberg and Larssen (1983). Black diamond at Ny-Alesund in summer shows median non-sea-salt sulfate concentration from Maenhaut et al. (1989). Open circles in the spring for Ny-Ålesund are March–April mean values (Sirois and Barrie, 1999). Note that the vertical coordinates use logarithmic scales.

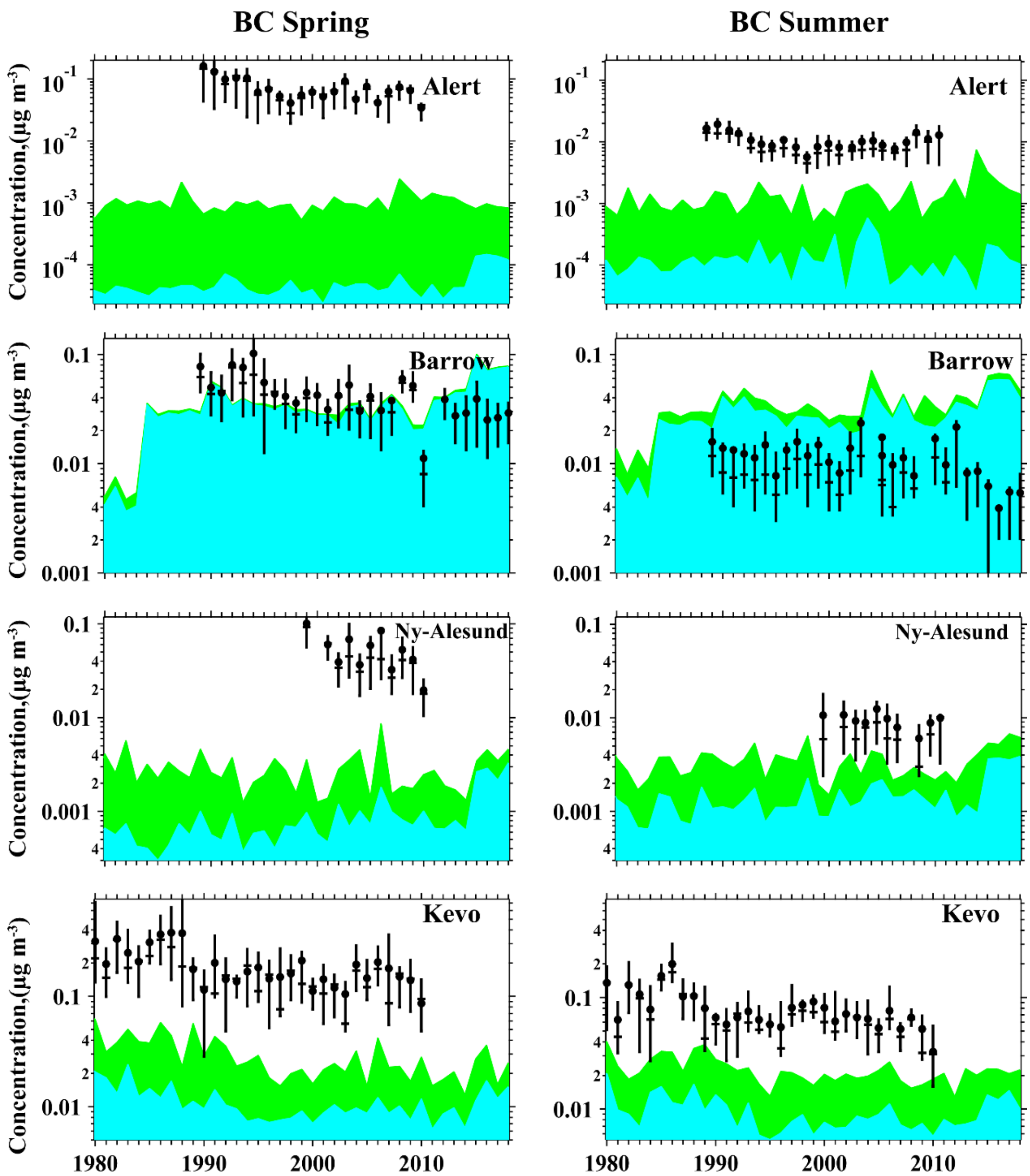
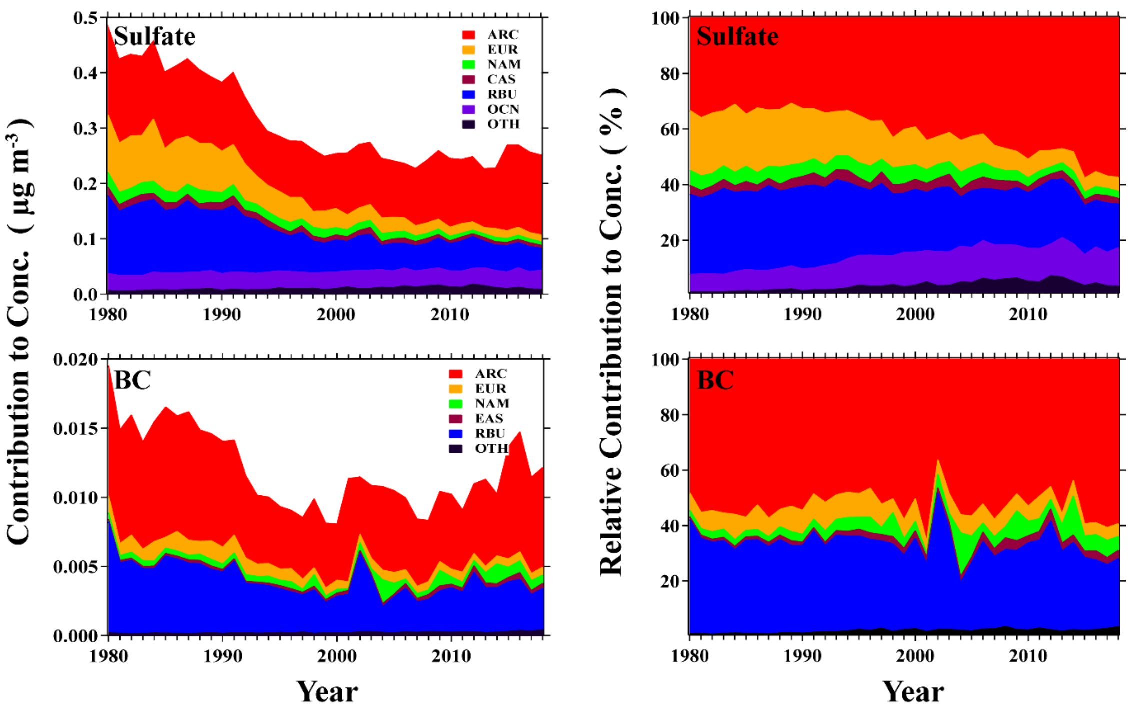



**Figure 4.** Same as Figure 3, but for surface BC (μg $m^{-3}$) at four (Alert, Barrow, Ny-Alesund, Kevo) Arctic sites.



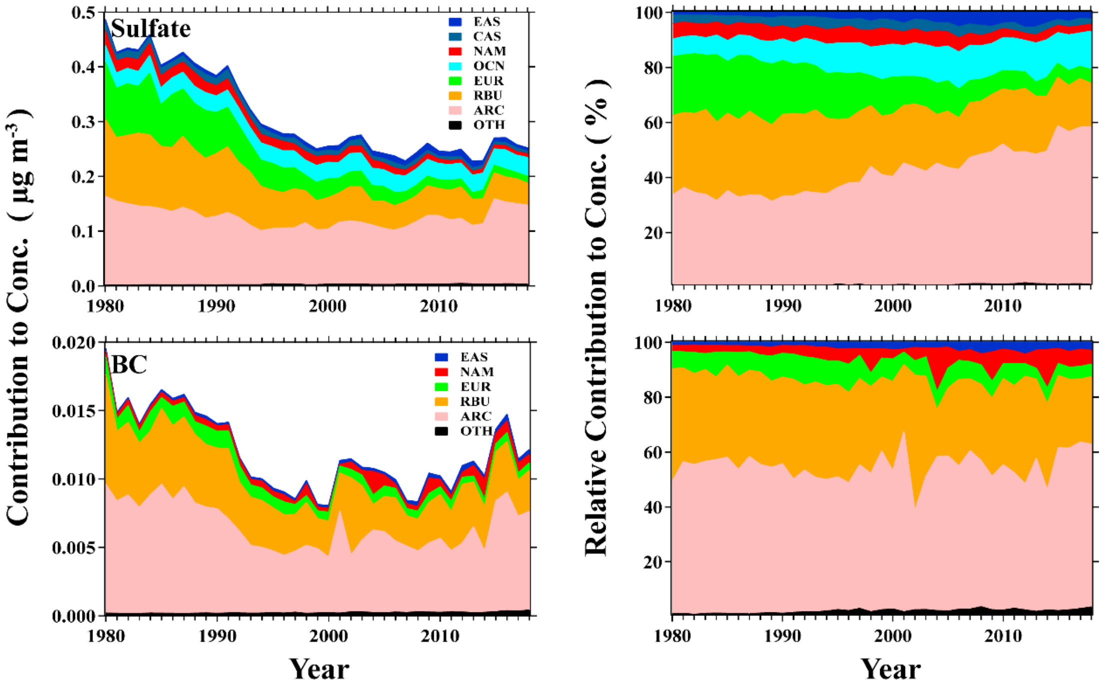
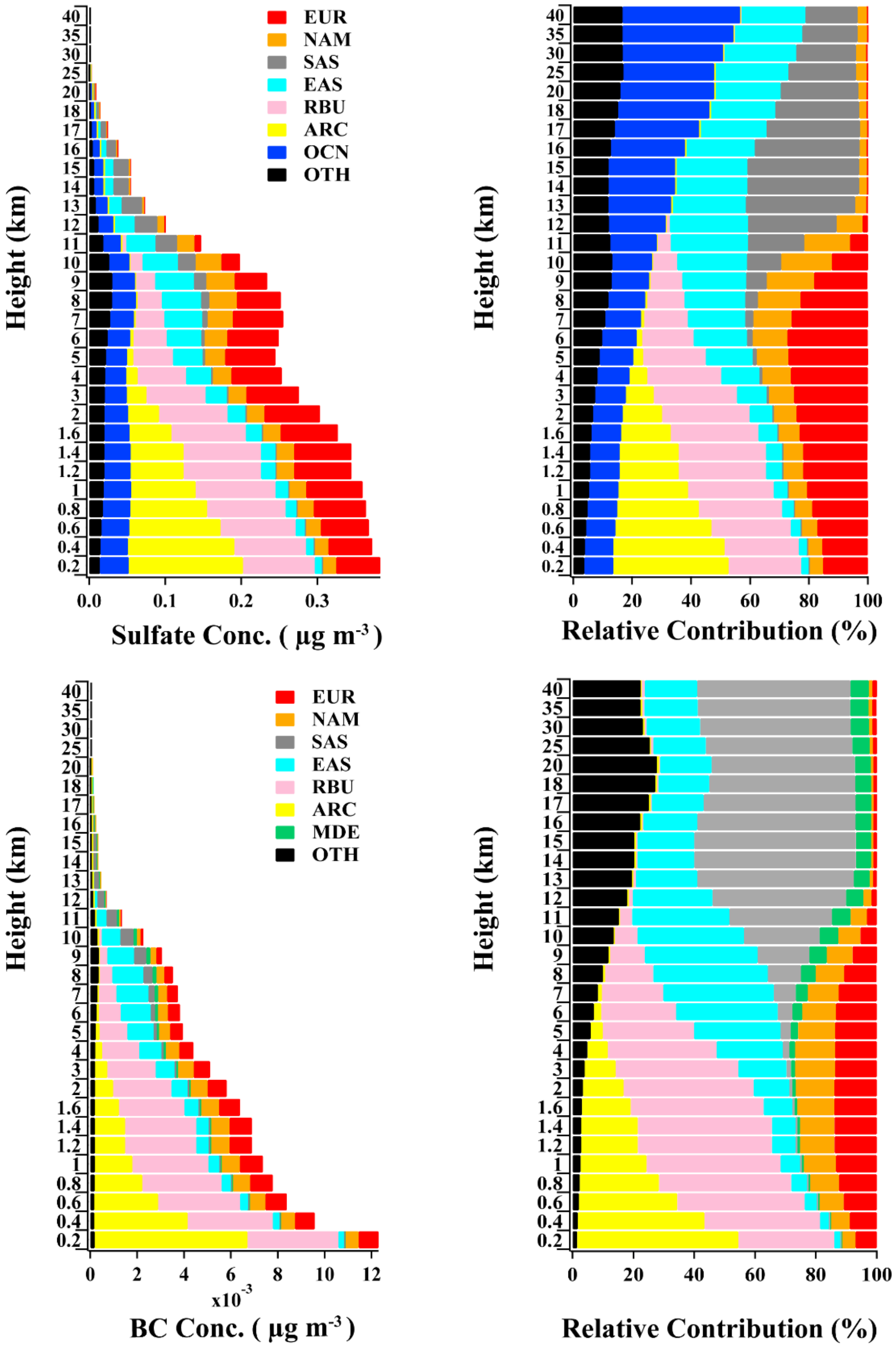




**Figure 5.** Time series (1980–2018) of absolute (left, μg m$^{-3}$) and relative (right, %) contributions of emissions from the major source regions to the simulated annual mean near-surface sulfate and BC concentrations averaged over the Arctic. (66.5°N–90°N). The remaining source regions with annual contributions less than 3% are combined and shown as OTH (other regions in figure S2).


EUR
NAM
SAS
EAS
RBU
ARC
OCN
OTH
Height (km)
Sulfate Conc. ( μg m$^{-3}$ )
Relative Contribution (%)
EUR
NAM
SAS
EAS
RBU
ARC
MDE
OTH
Height (km)
BC Conc. ( μg m$^{-3}$ )
x10$^{-3}$
Relative Contribution (%)

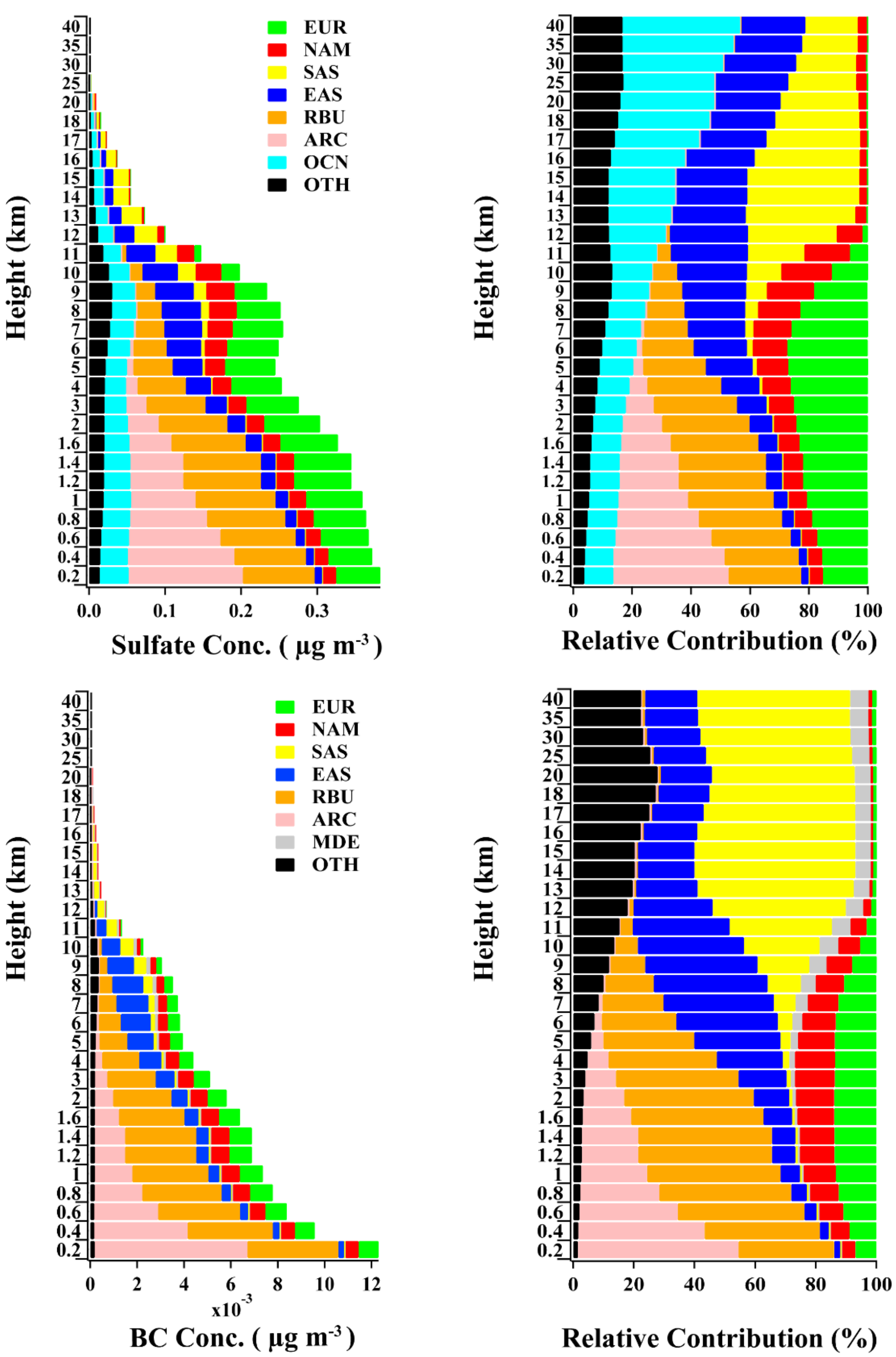



**Figure 6.** Annual mean vertical profile of sulfate (top) and BC (bottom) concentrations ($\mu g\ m^{-3}$) over the Arctic contributed by the tagged source regions (left) and their relative contributions (right, %) during 1980–2018. Sources with annual burden contributions less than 5% are combined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

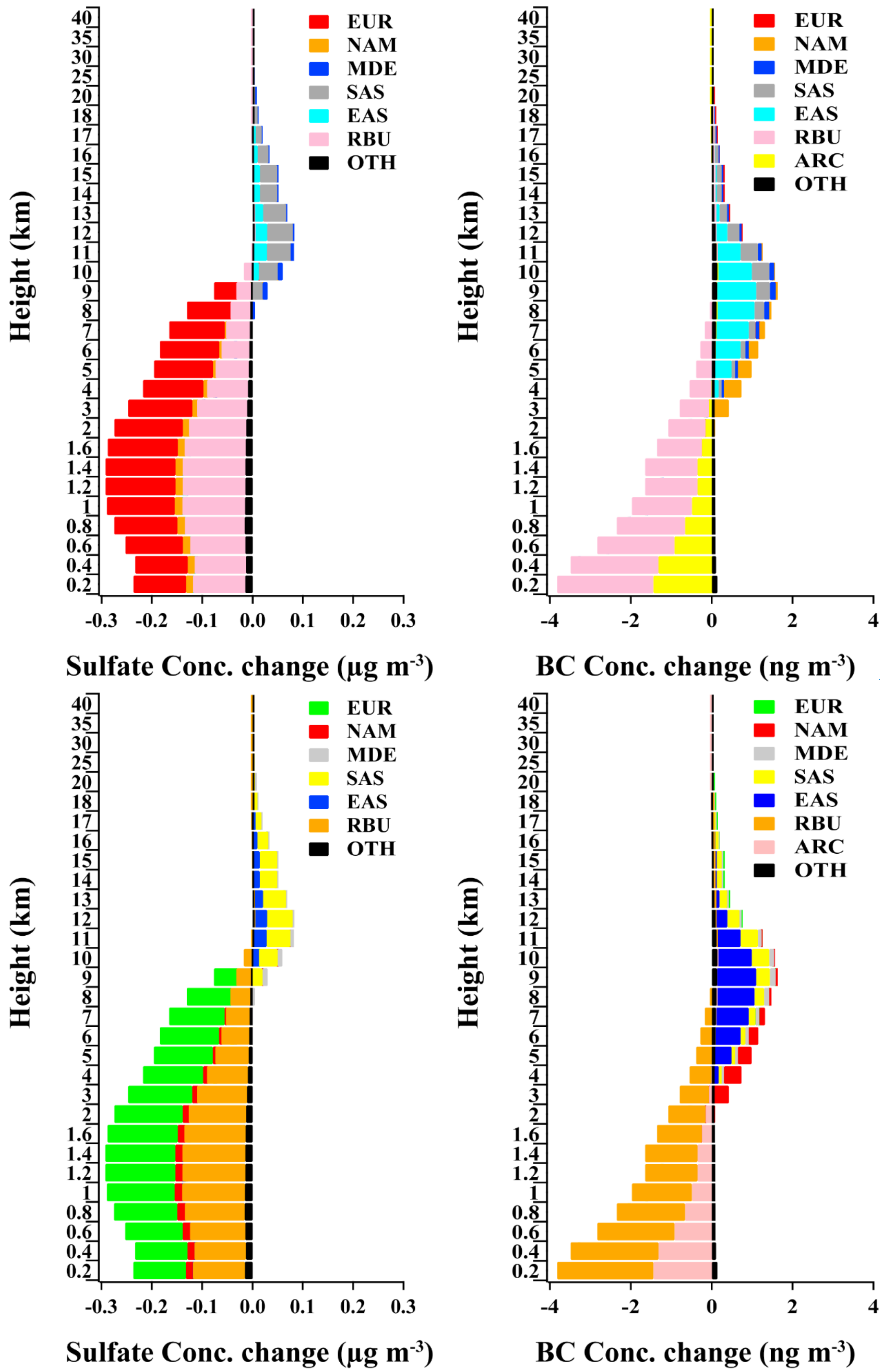
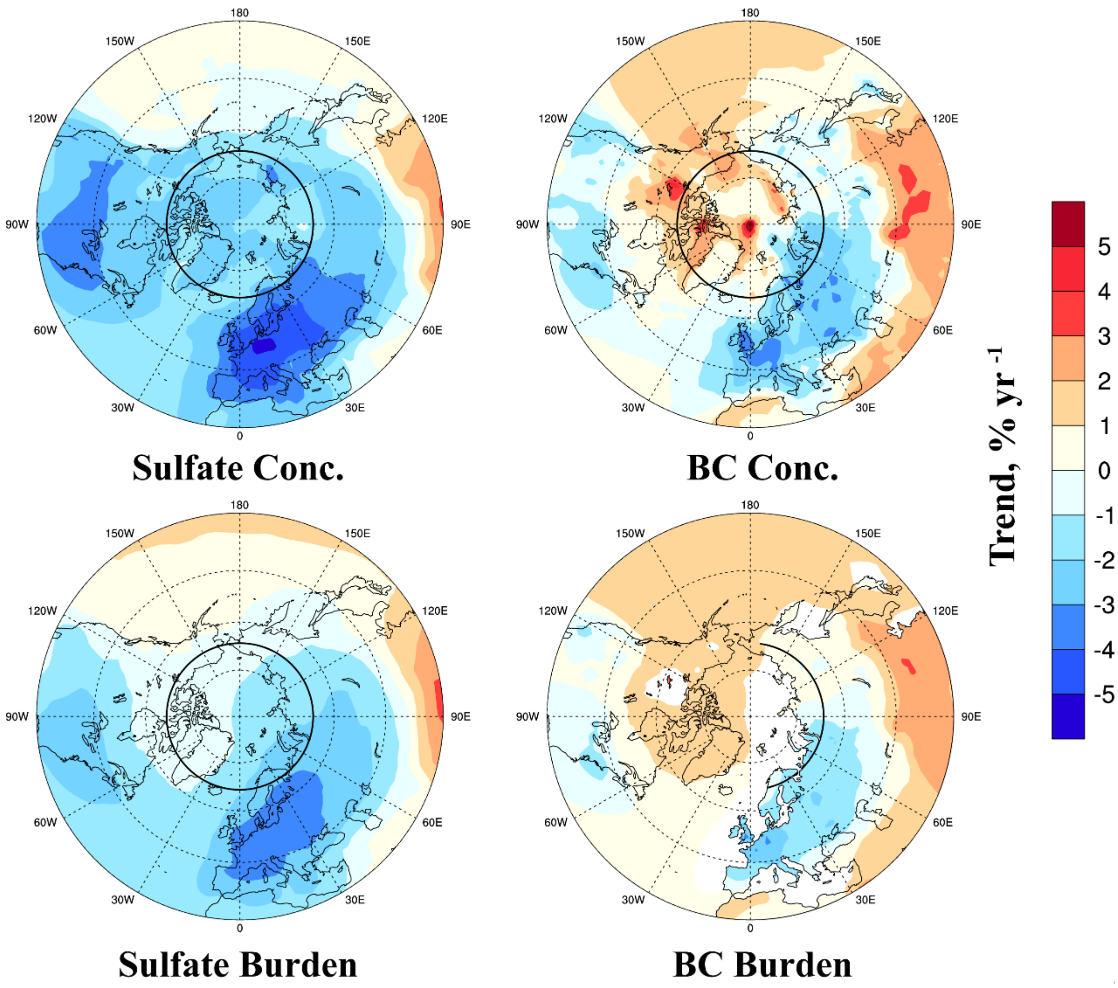



**Figure 7.** Changes in annual mean vertical profile of sulfate (μg $m^{-3}$, left) and BC (ng $m^{-3}$, right) concentrations over the Arctic contributed by the tagged source regions

between 1980–1984 and 2014–2018.



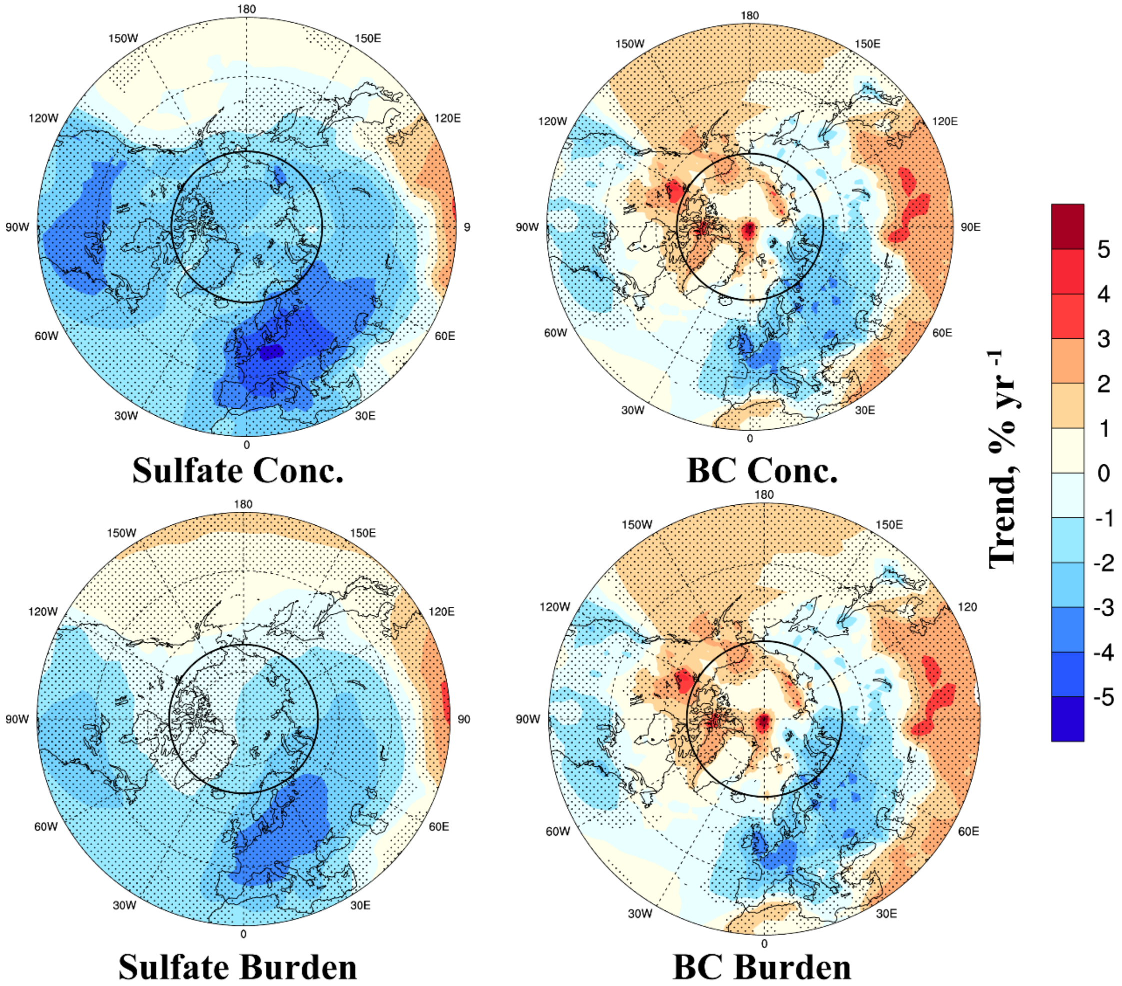



**Figure 8.** Spatial distribution of linear trends in annual mean sulfate (left) and BC (right) concentrations (% $yr^{-1}$) near the surface (top) and column burden (bottom) relative to the 39-year averages. The dotted areas indicate statistical significance with 95% confidence based on F-test.

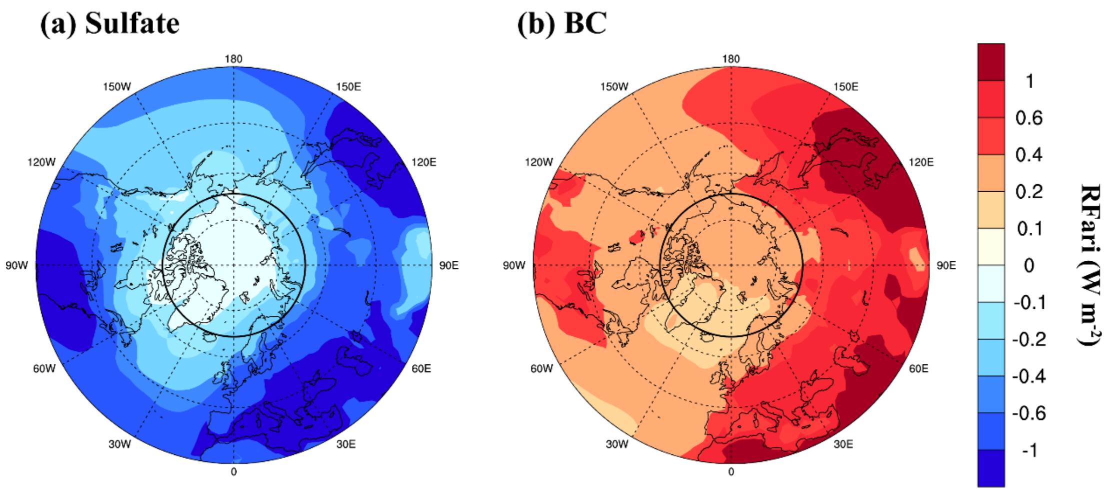
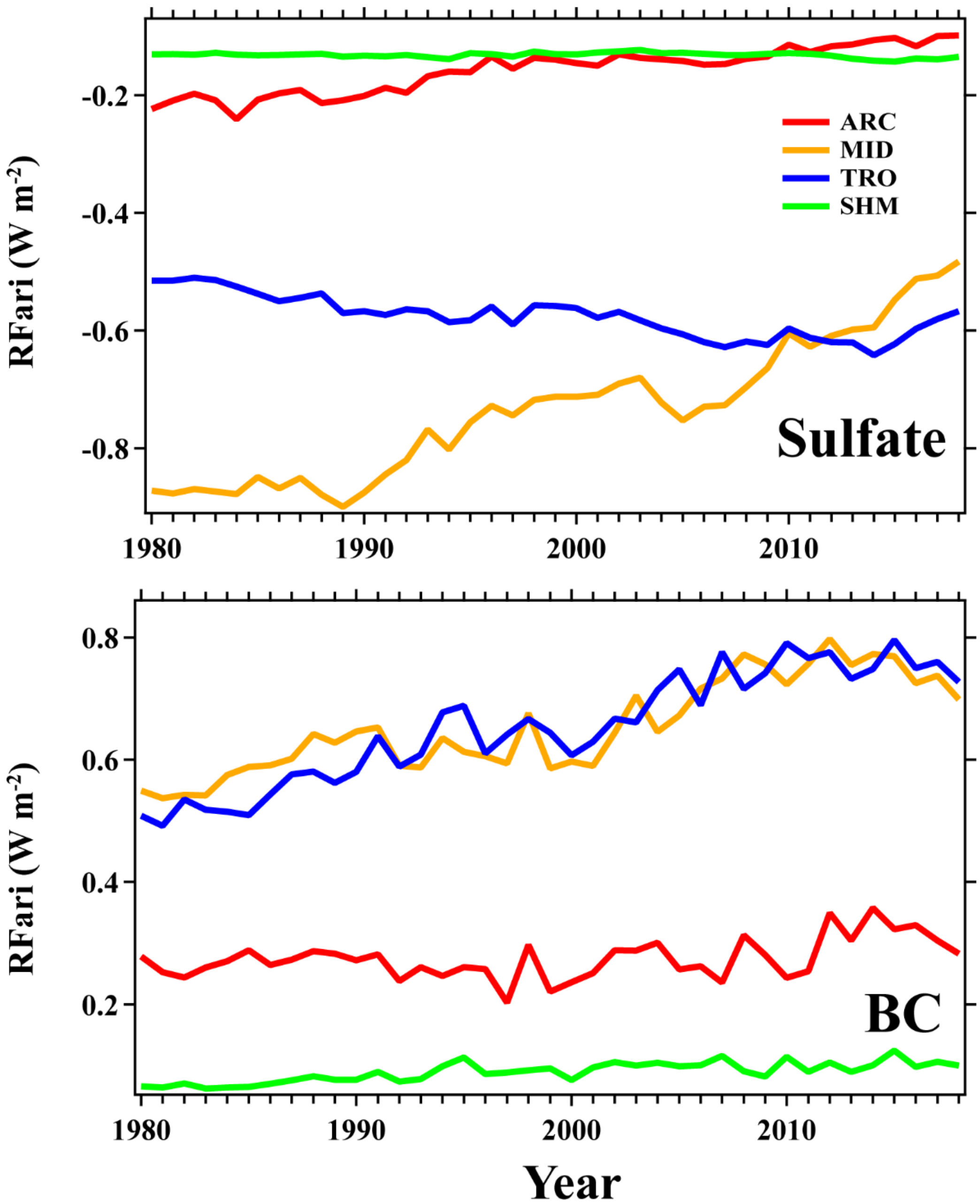



**Figure 9.** Spatial distribution of annual mean radiative forcing due to aerosol-radiation interactions ($RF_{ari}$) of (a) sulfate and (b) BC (W $m^{-2}$) at the TOA averaged over 1980–2018.


RFari (W m-2)
-0.2
-0.4
-0.6
-0.8
ARC
MID
TRO
SHM
Sulfate
1980
1990
2000
2010
RFari (W m-2)
0.8
0.6
0.4
0.2
BC
1980
1990
2000
2010
Year

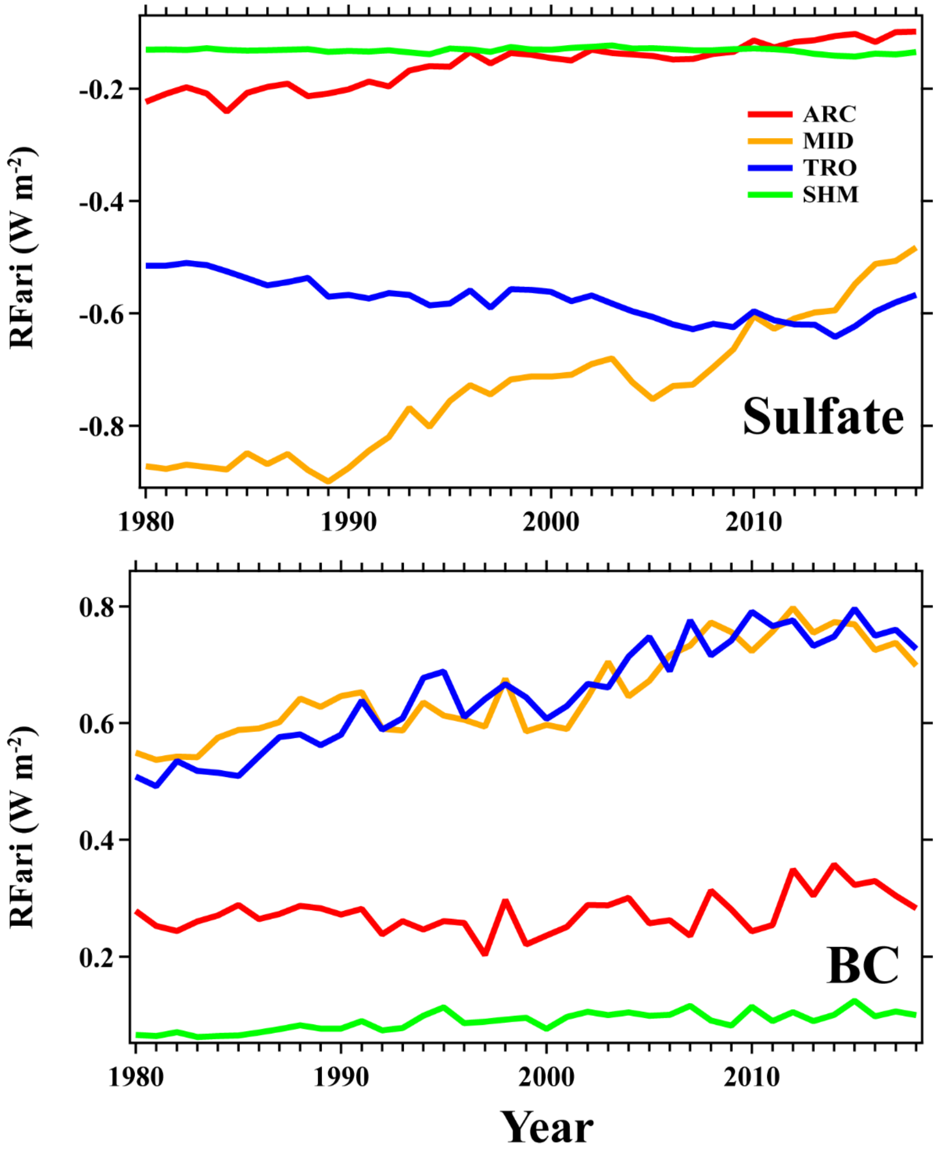
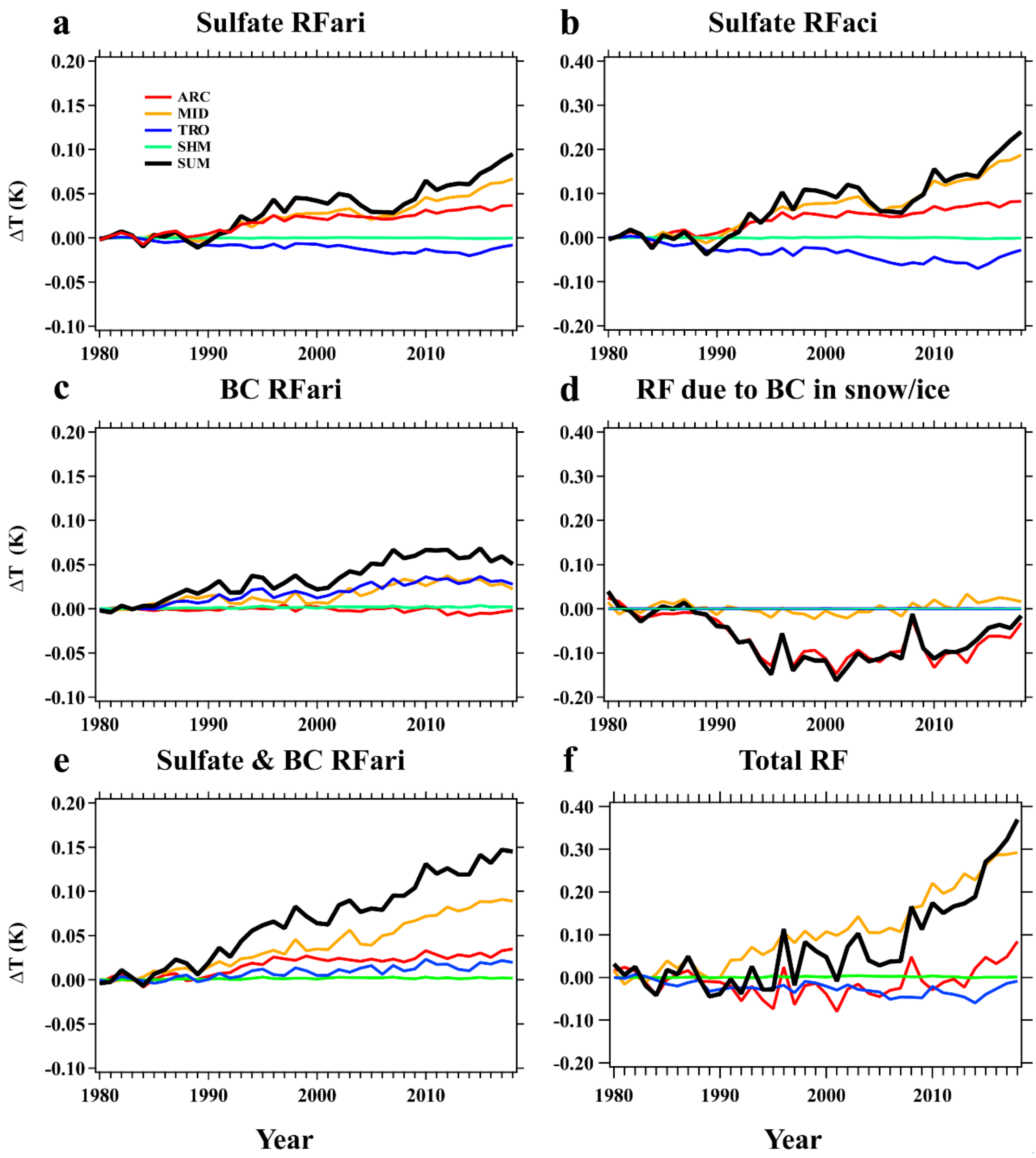



**Figure 10.** Time series (1980–2018) of annual radiative forcing due to aerosol-radiation interactions ($RF_{ari}$, W $m^{-2}$) of sulfate and BC over the Arctic (ARC, ~~66.5~~60°N–90°N), Northern Hemisphere mid-latitudes (MID, 28°N–~~66.5~~60°N), tropics (TRO, 28°S–28°N) and Southern Hemisphere (SHM, 90°S–28°S).


a
Sulfate RFari
b
Sulfate RFaci
c
BC RFari
d
RF due to BC in snow/ice
e
Sulfate & BC RFari
f
Total RF
ARC
MID
TRO
SHM
SUM
ΔT (K)
Year

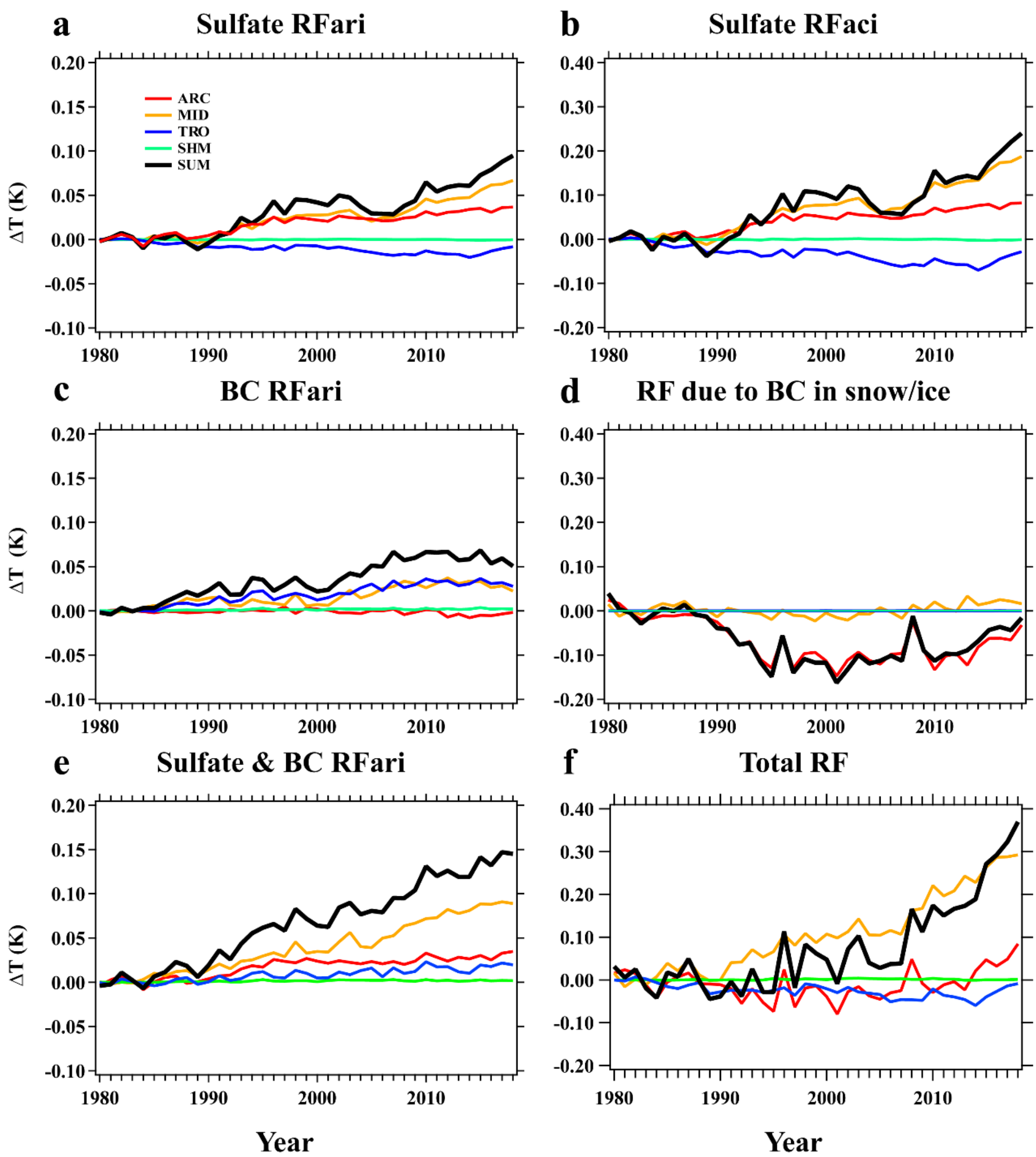



**Figure 11.** Time series (1980–2018) of the estimated response in surface temperatures (K) to the change in radiative forcing due to the aerosol-radiation interactions ($RF_{ari}$) of (a) sulfate, (c) BC, and (e) sum of sulfate and BC $RF_{ari}$, (b) radiative forcing due to aerosol-cloud interactions ($RF_{aci}$) 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).