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.”

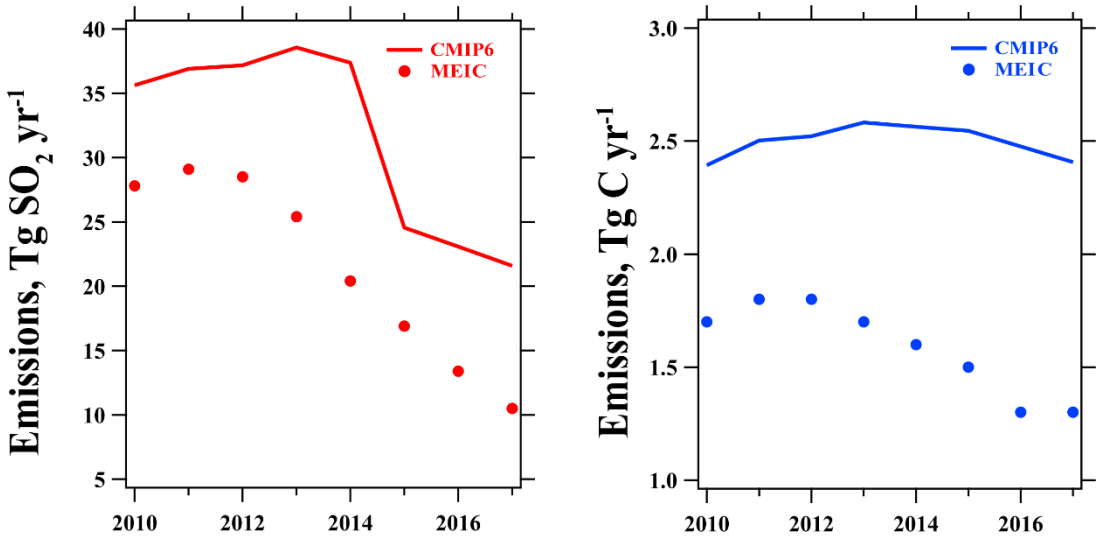

Figure S3. Annual anthropogenic emissions of SO_2 and BC in China from CMIP6 (solid lines) and MEIC (dotted lines).

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.

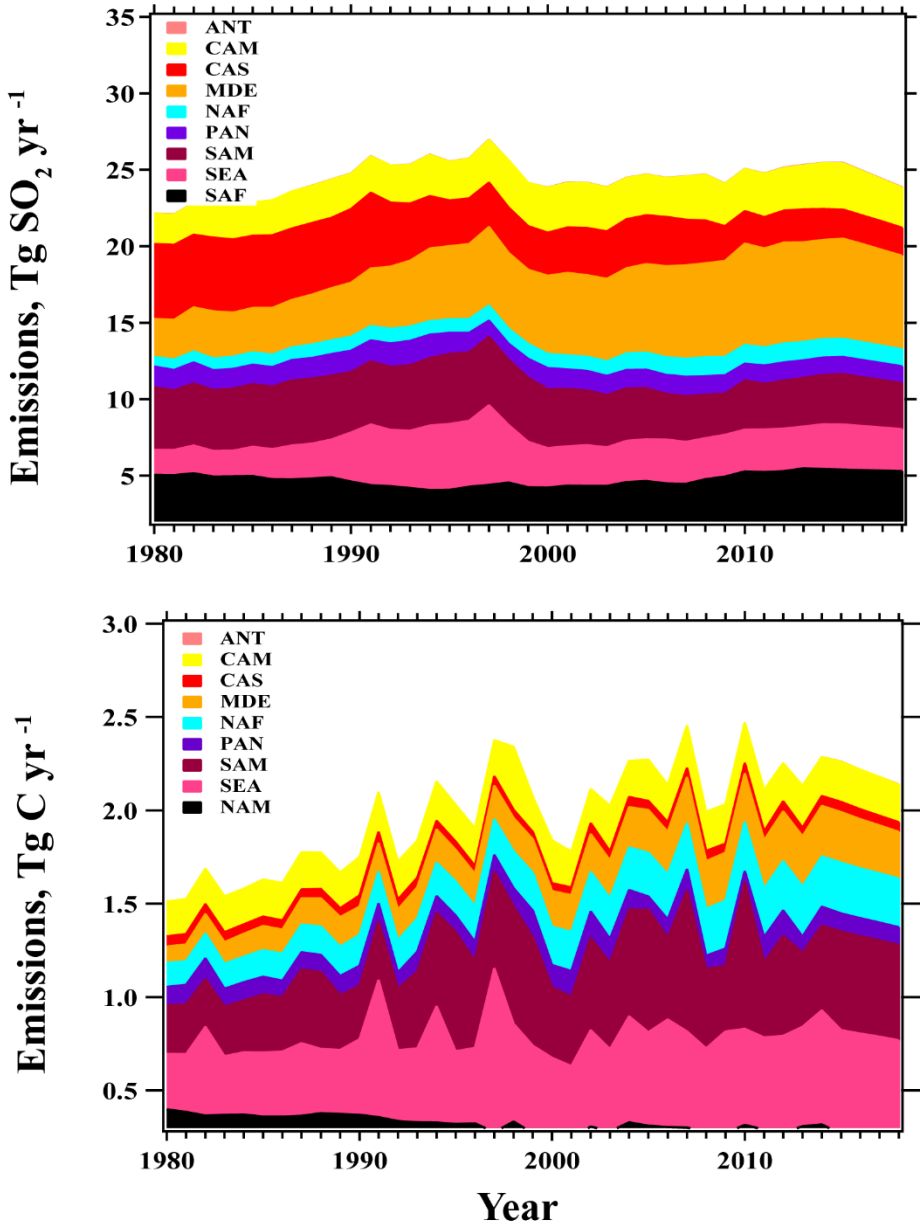

Figure S1. Time series of annual anthropogenic (top) SO_2 (Tg SO_2 yr^{-1}) and (bottom) BC (Tg C yr^{-1}) emissions from other regions of Fig. 2 individually.

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.

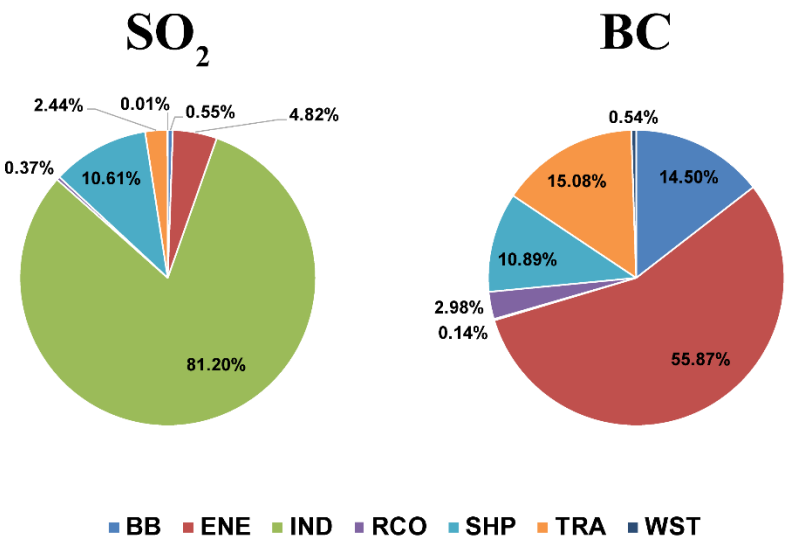

Figure S4. Annual mean of SO_2 and BC emissions from individual sectors in the Arctic during 2010-2014. BB: biomass burning, ENE: energy, IND: industry, RCO: residential, SHP: international shipping, TRA: transportation, WST: waste treatment.

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:

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