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.

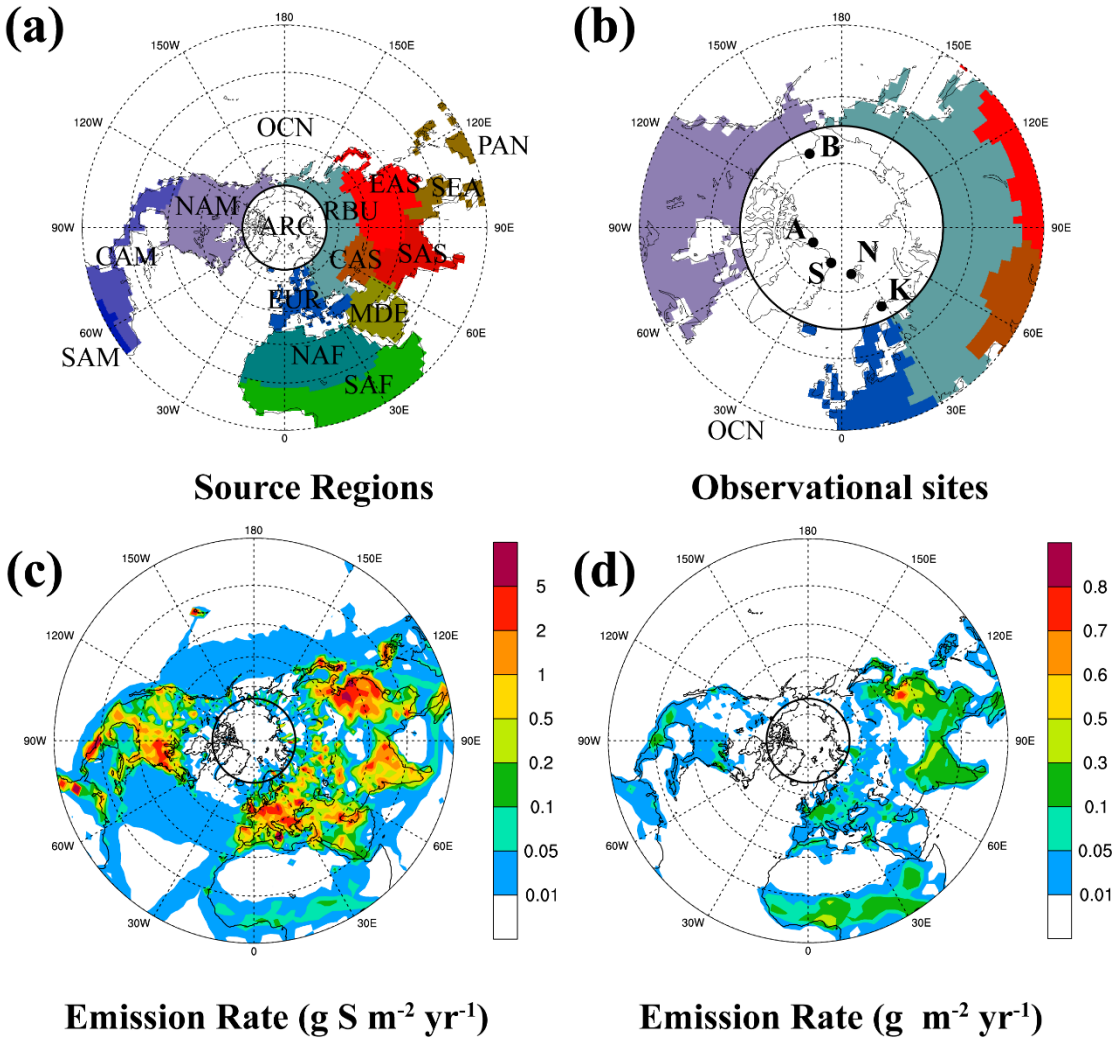

Figure 1. (a) Sixteen anthropogenic source regions (Europe (EUR), North America (NAM), Central America (CAM), South America (SAM), North Africa (NAF), South Africa (SAF), the Middle East (MDE), Southeast Asia (SEA), Central Asia (CAS), South Asia (SAS), East Asia (EAS), Russia-Belarus-Ukraine (RBU), 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:

Abbatt, J. P. D., Leaitch, W. R., Aliabadi, A. A., Bertram, A. K., Blanchet, J.-P., Boivin-Rioux, A., Bozem, H., Burkart, J., Chang, R. Y. W., Charette, J., Chaubey, J. P., Christensen, R. J., Cirisan, A., Collins, D. B., Croft, B., Dionne, J., Evans, G. J., Fletcher, C. G., Galí, M., Ghahremaninezhad, R., Girard, E., Gong, W., Gosselin, M., Gourdal, M., Hanna, S. J., Hayashida, H., Herber, A. B., Hesaraki, S., Hoor, P., Huang, L., Hussherr, R., Irish, V. E., Keita, S. A., Kodros, J. K., Köllner, F., Kolonjari, F., Kunkel, D., Ladino, L. A., Law, K., Levasseur, M., Libois, Q., Liggio, J., Lizotte, M., Macdonald, K. M., Mahmood, R., Martin, R. V., Mason, R. H., Miller, L. A., Moravek, A., Mortenson, E., Mungall, E. L., Murphy, J. G., Namazi, M., Norman, A.-L., O'Neill, N. T., Pierce, J. R., Russell, L. M., Schneider, J., Schulz, H., Sharma, S., Si, M., Staebler, R. M., Steiner, N. S., Thomas, J. L., von Salzen, K., Wentzell, J. J. B., Willis, M. D., Wentworth, G. R., Xu, J.-W., and Yakobi-Hancock, J. D.: Overview paper: New insights into aerosol and climate in the Arctic, Atmos. Chem. Phys., 19, 2527–2560, https://doi.org/10.5194/acp-19-2527-2019, 2019.

Abdul-Razzak, H. and Ghan, S. J.: A parameterization of aerosol ac- tivation: 2. Multiple aerosol types, J. Geophys. Res., 105, 6837–6844, https://doi.org/10.1029/1999JD901161, 2000.

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

AMAP, The Impact of Black Carbon on Arctic Climate (2011), By: P.K. Quinn, A. Stohl, A. Arneth,T. Berntsen, J. F. Burkhart, J. Christensen, M. Flanner, K. Kupiainen, H. Lihavainen, M. Shepherd, V. Shevchenko,H. Skov, and V. Vestreng, AMAP Tech. Rep., 4, 72 pp., Arctic Monitoring and Assessment Programme (AMAP), Oslo, 2011.

Bond, T. C., Zarzycki, C. M., Flanner, M. G., and Koch, D., Quantifying immediate radiative forcing by black carbon and organic matter with the Specific Forcing Pulse, Atmos. Chem. Phys., 11(4), 1505–1525, https://doi.org/10.5194/acp-11-1505-2011, 2011.

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.

Fisher, J. A., Jacob, D. J., Purdy, M. T., Kopacz, M., Le Sager, P., Carouge, C., Holmes, C. D., Yantosca, R. M., Batchelor, R. L., Strong, K., Diskin, G. S., Fuelberg, H. E., Holloway, J. S., Hyer, E. J., McMillan, W. W., Warner, J., Streets, D. G., Zhang, Q., Wang, Y., and Wu, S.: Source attribution and interannual variability of Arctic pollution in spring constrained by aircraft (ARCTAS, ARCPAC) and satellite (AIRS) observations of carbon monoxide, Atmos. Chem. Phys., 10, 977–996, https://doi.org/10.5194/acp-10-977-2010, 2010.

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

Flanner, M. G., Zender, C. S., Hess, P. G., Mahowald, N. M., Painter, T. H., Ramanathan, V., and Rasch, P. J.: Springtime warming and reduced snow cover from carbonaceous particles, Atmos. Chem. Phys., 9, 2481–2497, https://doi.org/10.5194/acp-9-2481-2009, 2009.

Liu, X., Easter, R. C., Ghan, S. J., Zaveri, R., Rasch, P., Shi, X., ... & Conley, A., Toward a minimal representation of aerosols in climate models: Description and evaluation in the Community Atmosphere Model CAM5, Geosci. Model Dev., 5(3), 709. https://doi.org/10.5194/gmd-5-709-2012, 2012.

Samset, B. H., Myhre, G., Herber, A., Kondo, Y., Li, S.-M., Moteki, N., Koike, M., Oshima, N., Schwarz, J. P., Balkanski, Y., Bauer, S. E., Bellouin, N., Berntsen, T. K., Bian, H., Chin, M., Diehl, T., Easter, R. C., Ghan, S. J., Iversen, T., Kirkevåg, A., Lamarque, J.- F., Lin, G., Liu, X., Penner, J. E., Schulz, M., Seland, Ø., Skeie, R. B., Stier, P., Takemura, T., Tsigaridis, K., and Zhang, K.: Modelled black carbon radiative forcing and atmospheric lifetime in AeroCom Phase II constrained by aircraft observations, Atmos. Chem. Phys., 14, 12465–12477, https://doi.org/10.5194/acp-14-12465-2014, 2014.

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

Shindell, D. T., Chin, M., Dentener, F., Doherty, R. M., Faluvegi, G., Fiore, A.

M., Hess, P., Koch, D. M., MacKenzie, I. A., Sanderson, M. G., Schultz, M. G., Schulz, M., Stevenson, D. S., Teich, H., Textor, C., Wild, O., Bergmann, D. J., Bey, I., Bian, H., Cuvelier, C., Duncan, B. N., Folberth, G., Horowitz, L.W., Jonson, J., Kaminski, J.W., Marmer, E., Park, R., Pringle, K. J., Schroeder, S., Szopa, S., Takemura, T., Zeng, G., Keating, T. J., and Zuber, A.: A multi-model assessment of pollution transport to the Arctic, Atmos. Chem. Phys., 8, 5353–5372, https://doi.org/10.5194/acp-8-5353-2008, 2008.

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.

Stohl, A.: Characteristics of atmospheric transport into the Arctic troposphere, J. Geophys. Res.-Atmos., 111, D11306, https://doi.org/10.1029/2005JD006888, 2006.

Wang, Q., Jacob, D. J., Fisher, J. A., Mao, J., Leibensperger, E. M., Carouge, C. C., Le Sager, P., Kondo, Y., Jimenez, J. L., Cubison, M. J., and Doherty, S. J.: Sources of carbonaceous aerosols and deposited black carbon in the Arctic in winter-spring: implications for radiative forcing, Atmos. Chem. Phys., 11, 12453–12473, https://doi.org/10.5194/acp-11-12453-2011, 2011.

Wang, H., Rasch, P. J., Easter, R. C., Singh, B., Zhang, R., Ma, P. L., Qian, Y., Ghan, S. J., and Beagley, N.: Using an explicit emission tagging method in global modeling of source‐receptor relationships for black carbon in the Arctic: Variations, sources, and transport pathways, J. Geophys. Res., 119, 12,888-12,909, https://doi.org/10.1002/2014JD022297, 2014.