Authors' Response to Comments from Referee #1

We would like to thank Referee #1 for his/her useful comments and suggestions that helped to improve the quality of this manuscript. Reponses to these comments are provided below.

Specific Comments:

P1L15: I found the abstract too long, written like a conclusion section. Please try to make it more concise and underline the key messages from your study.

Response: Thanks for the suggestions. The abstract has been condensed.

P1L27-28: I think you oversell the performance of the model in reproducing the vertical profile of BC. I don't think you can say that the simulations are "consistent" with the airborne measurements of BC. See for example Fig. 5 where the simulated profiles are flat, especially between 700 and 500 hPa, whereas the observations demonstrate a significant enhancement of BC concentrations. See more comments below (P15L14, P17L17 or P23L1).

Response: Thanks for pointing this out. We have clarified that our simulations underestimate BC concentrations in the middle troposphere in the abstract and the following paragraphs.

P6L4-7: The way it is phrased makes it difficult to understand what is new in this study. Do the authors develop the approach? Do they apply it to a different/longer period than that in Qi et al. (2017a)? Highlight the differences between the methodologies detailed in Kopacz et al. (2011) or Qi et al. (2017a).

Response: Qi et al. (2017a) has been revised to Qi et al. (2017b). We extended the time period from one month (April) in Qi et al. (2017b) to four months (each representing one season) and extended from the surface BC in Qi et al. (2017b) to the column BC that has implications for radiative forcing. We have highlighted these differences in the sentence as "We extend the application of this method to investigate the seasonal and annual responses of Arctic column BC to changes in regional emissions.". Kopacz et al. (2011) has been removed because this is a relatively old study and it not directly relevant to this study.

P6L8: Say that GEOS-Chem is a global CTM. It was only mentioned in the abstract.

Response: Done.

P7L5-6: Can you justify why no scattering corrections to the aethalometer measurements contrary to what is done for the PSAP data (P7L12)?

Response: Good question. The manufacturer's recommended MAC of 16.6 $m^2 g^{-1}$ is calibrated to account for multiple scattering (Sharma et al., 2017), thus no additional scattering corrections are necessary. We have revised the description to the following in the manuscript:
"This MAC value is recommended by the manufacturer for Model AE31 at 880 nm to account for absorption by BC and additional light scattering by both particles and filter fibers."

P10L11-16: How are fire emissions injected in the model? Are the emissions confined to the surface or does the model takes into account the variability in injection heights (function of burnt vegetation, intensity of fires, buoyancy, ...)?

Response: Fire emissions are injected into the boundary layer in the model. We have clarified this as "Biomass burning emissions are injected into the boundary layer in our simulations" in the manuscript.

P10L22: The description of the simulation is not given in this study. The authors give only a reference to an "old" paper (Park et al., 2003), which cannot be taken as a good reference to fully decribe what has been really done in this work. Please give additional details: period of the simulation, horizontal and vertical resolutions, . . . A lot of modifications have been included in the GEOS-Chem model from the study of Park et al. (2003). Some of them are likely to produce substantial changes in the BC distributions (e.g. wet deposition). Try to underline the main improvements.

Response: Thanks for the suggestion. We have included a detailed description of wet and dry deposition with major improvements since Park et al. (2003) in the model as the following in the manuscript:

"Dry deposition of BC aerosols adopts a standard resistance-in-series scheme as described in Zhang et al. (2001) with improvements on BC dry deposition velocity over snow and ice following Fisher et al. (2010) and Wang et al. (2011). Wet deposition of BC aerosols is initially described in Liu et al. (2001) and developed by Wang et al. (2011) to distinguish between liquid cloud (T > 268 K) in which 100 % hydrophilic BC is removed and ice cloud (T < 268 K) in which only hydrophobic BC is removed."

Additional details including the period of the simulation, horizontal and vertical resolutions are described on page 10 line 11-19.

P10L26: Does the model include wet deposition also in subgrid convective updrafts?

Response: Yes. This is described in Liu et al. (2001).

P13L8: The authors explain that EBC concentrations are biased high relative to rBC measurements because of absorbing components and errors in the coefficient used to derive BC concentrations from absorption coefficients. But the fact that EC concentrations are always larger than rBC is never explained. Is it similar to what was observed in other studies? Can it be caused by a difference in sampling the plumes?

Response: Thanks for point this out. Sharma et al. (2017) showed that some of the difference between EC and rBC could be explained by the presence of pyrolysis OC and carbonate carbon that might remain in aerosols after heating to 870 °C in the thermal method but were removed in aerosols at 3600°C in the refractory method. We have included this reason in the manuscript as "EC concentrations are lower than EBC concentrations from the Aethalometer, yet still high relative to rBC partly due to the presence of pyrolysis OC and carbonate carbon (Sharma et al., 2017)."

P13L14-16: May this discrepancy between aethalometer and PSAP measurements be also ascribed to differences in the calibration method: scattering corrections applied to PSAP but not

to aethalometer data? And/or spectral dependency of the absorption coefficient of other substances than BC?

Response: The questions on scattering corrections were explained in the response to P7L5-6.

To understand the effect of spectral dependency of the absorption coefficient of other substances (i.e. brown carbon) on our results, we examined the Aethalometer EBC measurements at 370 nm at Barrow to see if the summer peak still exists at 370 nm. The result showed a distinct peak in July and August with a concentration increased by 10 ng m^{-3} compared to June and September, indicating the existence biomass burning. Thus, the summer peak in Aethalometer measurements at Barrow was not affected by the wavelength of measurements, but was influenced by biomass burning that was missing in PSAP measurements. We have revised the manuscript to the following to clarify this point.

"The summer peak is also observed in Aethalometer EBC measurements at 370 nm that is sensitive to brown carbon, indicating the influence of biomass burning. Unintentional exclusion of biomass burning plumes in the local pollution data screening performed for PSAP measurements at Barrow could contribute to the bias between the PSAP and the Aethalometer there (Stohl et al., 2006)."

P14L11-14: The peak observed in model simulations at Barrow is explained by a wrong timing of biomass burning emissions. Shouldn't it be the case for all models compared in the Eckhardt et al. (2015) paper if they all use the same fire emissions (with the same timing)? What is the role of injection heights in summer? And what is the contribution of wet deposition processes in the different models?

Response: Eckhardt et al. (2015) used identical biomass burning emissions (GFED 3.1) for all the models in their study. They attributed the difference of simulated surface BC concentrations from different models not only in summer but also the whole year to the treatment of aerosol wet scavenging in the models.

It is not clear what the effect of injection heights is on Arctic surface BC concentrations because some models (i.e. FLEXPART and HadGEM3) do not show the summer peak with biomass burning emissions distributed evenly within the boundary layer, while some other models (i.e. ECHAM6-HAM2) do show the summer peak with the same biomass burning emissions algorithm.

We have revised the sentence from
"At Barrow all simulations show a distinct peak in July, which is due to the timing of biomass burning." to
"At Barrow all simulations show a distinct peak in July, which is partly due to the timing of biomass burning."

P15L14: I think this sentence oversells the model prediction. The main feature noticed in the observed BC profile is the significant enhancement between 700 and 500 hPa, which is not captured at all by the simulations. So the model does not "generally represent "the relative vertical distribution of BC. The next sentence in the manuscript is a much better way to say this.

Response: We have revised the sentence to "All simulations generally represent the near constant vertical distribution of BC measurements from the surface to 700 hPa, and the

decrease above 500 hPa, yet none represent the enhancement between 700-500 hPa."

P15L17-18: The way the sentence has been written gives the impression that most models also strongly underestimate BC in the mid or upper-troposphere. You cannot say this without also referring to more recent studies, highlighting either an overestimation aloft (e.g. Sharma et al. 2013 ; Wang et al., 2014), an improvement in the mid-troposphere (e.g. Breider al., 2014 ; Raut et al., 2017) or a very distinct behaviour between models (as in Eckhardt et al., 2015). In particular both Breider al. (2014) and Wang et al. (2014) used the GEOS-Chem model to represent BC in the Arctic and the results are not similar. This latter study suggested an incorrect production of convective precipitation in the summer in the Arctic. Can you give also some possible reasons to explain the underestimation of BC in your paper (e.g. emissions, plume injection, numerical diffusion, aerosol scavenging, ...) and some ways of improvements?

Response: Thanks for the suggestion. We have referred to Koch et al. (2009), Eckhardt et al. (2015), Breider et al. (2014) and Wang et al. (2011) in the discussion and have proposed some possible reasons for the underestimation as the following:

"The remaining underestimation of 14 ng m-3 RMSE in 500-700 hPa in the HTAP+flaring simulation is possibly due to insufficient magnitude or altitude comparisons of model with ARCTAS and ARCPAC measurements (Koch et al., 2009; Wang et al., 2011; Breider et al., 2014; Eckhardt et al., 2015) as proposed based on preferential sampling by the aircraft of plumes discussed further below."

P16L5-8: I cannot believe that missing plumes during one specific day (8 April 2015) could explain such a substantial discrepancy. Furthermore, writing "is perhaps due" is "perhaps" a bit too colloquial for a paper (but not a review!). I don't see what this sentence brings to the analysis.

Response: We have rephrased this sentence to indicate the potential sampling bias of plumes and have given another possible reasons as described in the response to the previous comment.

P16L24: What do you mean exactly by the "misrepresentation of these plumes"? Give rather the physical processes responsible for the strong BC underestimation: emissions? numerical diffusion?

Response: By misrepresentation, we meant low in magnitude. Emissions or numerical diffusion might be responsible for the underestimation. We have revised the sentence to "The underestimated magnitudes of these plume, likely related to emissions or numerical diffusion, may contribute to the underestimation of BC concentrations between 500-700 hPa in Fig. 5".

P17L12-13: It does not seem right in summer. Why are the BC columns so low in July near northern Russia? Are the flares off in summer? Is it realistic?

Response: We have included emissions for Jan, Apr and Jul at the bottom panel of Fig. 6. As shown by the figure, flaring emissions in July are very similar to those in the other months. The minor flaring contribution in summer is shown not only for the column but also at the surface where the flaring contribution is usually the largest, as shown in the red shadings of Fig. 3, which is consistent with Stohl et al. (2013). Thus, the flares are not off in summer but the effective wet scavenging in summer likely removes most of BC from anthropogenic sources including flaring. We have included this explanation in the

manuscript as the following:

"In July, the enhanced concentrations in western Siberia due to flaring are less obvious, due to more effective wet scavenging in summer."

P17L17: I would remove the word "vertical" and keep only the term "spatial" distributions.

Response: Done.

P18L2-4: This is not only caused by different transport patterns along which air masses reach the Arctic region, but also by different transport efficiencies due to scavenging.

Response: We have revised the sentence to "partly reflecting different transport pathways and scavenging efficiencies".

P19L8: I disagree with the use of the word "generally" here. A lot of studies have been focused on the same objective and they are not necessarily in agreement. How do the results of this study compare for example to Table A1 of Wang et al. (2014) who summarized twelve studies focusing on the source attribution of Arctic BC?

Response: Thanks for the suggestion. We have compared to Huang et al. (2010), Sharma et al. (2013), Shindell et al. (2008), Stohl (2006) and Ma et al. (2013) in Table A1 of Wang et al. (2014) and have revised the paragraph to the following

"The largest contribution from eastern and southern Asia to Arctic BC burden in this study is consistent with Ma et al. (2013) and Wang et al. (2014). However, some prior studies suggested that Europe had the largest contribution to Arctic BC burden (Stohl, 2006; Shindell et al., 2008; Huang et al., 2010a; Sharma et al., 2013) The difference likely arises from trends in anthropogenic emissions with reductions from Europe and increases in eastern and southern Asia as discussed further below."

P20L6-7: Another possible explanation could be that biomass burning plumes have been transported at a too low altitude. This would explain the strong overestimation at the surface and the marked underestimation aloft. Is it a possible scenario?

Response: In the response to P13L14-16, we have described that biomass burning plumes are likely missed by the PSAP measurements. Thus, it is not clear to us whether the discrepancy should be attributed to the simulation or the measurements.

P23L1: Here again, the simulated vertical BC profile is not consistent with that measured. The following sentence is a good way to say this.

Response: Revised.

P24L5-8: This is an odd conclusive remark for a study focused on model simulations. It may be better to insist on the model uncertainties in the result mentioned above mentioned and how they can be addressed in future work.

Response: Thanks for the suggestion. We have added "The considerable impact of emissions from China and Indo-Gangetic Plain on the Arctic deserves further investigation." to the conclusion.

Fig. 3: What is the uncertainty on simulated monthly BC concentrations? It could be estimated by the standard deviations of the model results used to derive the monthly averages. Please add them on Fig. 3 at least for the HTAP+flaring emission inventory.

Response: Done.

Fig. 4: Could you add an errorbar on the ground mean concentrations?

Response: Done. But only one year (2009) of ground measurement is available for Ny Alesund, so no error bar presents in the figure.

Fig. 5: This caption is good, but some of this text should also be reported in the manuscript, in particular L6-7 to understand how the analysis is performed.

Response: Done.

Fig. 6: This is not any flaring plume in July. Is it normal?

Response: There is flaring plume in July but the magnitude is smaller than the other seasons because of more effective wet scavenging in summer. This has been described in more details in the response to P17L12-13.

Fig. 8: Why aren't there error bars at Barrow and Ny Alesund stations as at Alert?

Response: Thanks for pointing out this error. Corrected.

Fig. 9: Why don't you take into account 12 months to build the annual map? I think this is confusing. It is fine to present maps for particular months (Jan, Apr, Sept) but I don't undertand the purpose of computing an annual average based on only 3 months, when 12 have actually been simulated.

Response: We do not have a full year simulation with the adjoint model because it is time consuming. We assume the four months (Jan, Apr, Jul and Sept) are representative of four seasons. We have revised the caption to "The annual map is the average of contributions in January, April, July and September calculated with the adjoint model."

Table 1: Why don't you also include rRMSE in this stable? The discussion relative to this section would be clearer.

Response: We have included rRMSE in Table 2.

Technical comments:
In many places, add a space between value and unity, e.g. P2L6, P7L22, P8L4, P11L4, P15L8, P15L16, P15L17, P16L1, P16L18, P16L26, P18L9, P18L22, P23L3, P23L11, P23L15.

Response: Done.

P3L8: Remove the dot before the parenthesis.

Response: Done.

P7L21: Neodymium dopped YAG is generally abbreviated as Nd:YAG, not Ni-YAG.

Response: Done.

P9L25: ACP journal recommands to write Figure instead of Fig. at the beginning of a sentence.

Response: Done.

P10L6: Replace semi-colon by comma.

Response: Done.

P17: section should be abbreviated as Sect. in ACP.

Response: Done.

P13L10-15: There is a typo in aethalometer L10, L11 and L15.

Response: Done.

P22L4: "to a half of the impact": this is not very clear. Please reformulate.

Response: Done.

P27L10-14: Evangeliou et al. (2016): the paper has been published in ACP in June 2016. It is not a discussion paper any longer.

Response: Done.

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Authors' Response to Comments from Referee #2

We sincerely thank the Referee #2 for taking the time to review our paper and for providing constructive suggestions for improvement. Reponses to these comments are provided below.

In this paper, Xu and coauthors use the GEOS-Chem transport model to quantify the contributions from different regions to the Arctic black carbon burden during three years – 2009, 2011, and 2015. They first validate the model with surface-based monthly mean observations and with measurements from two springtime aircraft campaigns. They find relatively good agreement between the model and observed concentrations. For two Arctic sites (but not a third), this agreement improves when they include in their model an inventory of gas flaring emissions from western Siberia. Sensitivity studies with the forward model yield the contributions from different regions to Arctic BC, while simulations with the adjoint version of GEOS-Chem provide spatially-resolved information on these contributions.

The main findings of this paper are as follows: Anthropogenic BC from eastern and southern Asia dominate the Arctic BC burden in spring and contribute about one-third of the annual burden, with larger contributions aloft than near the surface. Anthropogenic BC from northern Asia are important BC in the lower troposphere, especially in spring. Biomass burning contributes 25% of Arctic BC annually. Results from the adjoint point to interesting influences on Arctic BC from regions as far south as the Indo-Gangetic Plain.

Response: Thank you.

Main criticisms.

1. This paper moves forward the research on the origins of Arctic haze, providing in particular an update on how recent increases in anthropogenic BC from Asia may affect the Arctic. However, the authors do not make clear how their work builds on four recent GEOS-Chem studies that focus wholly or in part on Arctic BC: Wang et al. (2011, 2014) and Breider et al. (2014, 2017). No doubt the authors were unaware of the 2017 paper, but the other three papers were published well before this one was submitted. Only Wang et al. (2011) is mentioned, and that only in passing. It is in particular concerning that the authors do not make clear whether they took advantage of the improvements in BC wet deposition of Wang et al. (2011, 2014). Did the authors include the snow scavenging scheme and the improvements to washout and rainout from

Wang et al. (2011)? What about improvements to the impaction scavenging (Wang et al., 2014)? As is, the text cites only the wet deposition scheme of Liu et al. (2001). If the authors chose not to implement the Wang et al. (2011, 2014) improvements to wet deposition, the reader will want to know the rationale and what difference it would make if these improvements had, in fact, been included.

Brieder et al. (2014) focused on Arctic haze in 2008, and Brieder et al. (2017) examined the evolution of Arctic haze from 1980 to 2010. The authors can easily make the case that by simulating Arctic haze in 2009, 2011, and 2015, their paper provides an update to the Breider research, especially in light of increasing Asian emissions. But first they need to compare their approach and results very carefully with those in the earlier work. For example, the Brieder papers make use of a different emission inventory than does the current paper, and the reader will want to know how these inventories differ. As another example, Brieder et al. (2014) appears to capture the mid-tropospheric peak in BC, while the current work does not. Again the reader will want to understand this discrepancy.

Responding to criticism #1 will require some effort. A close reading of the four relevant papers is necessary, and a detailed account of how the current paper moves beyond the previous papers is expected by the reader.

Response: Thanks for these suggestions. We use version 10 of GEOS-Chem, which was the latest version available at the start of this work. Thus the wet deposition of Wang et al. (2011) was implemented in our simulation. We have clarified this in text. The developments of Wang et al. (2014) were not implemented into GEOS-Chem until version 11, and thus were not included here. Furthermore, these developments have little effect in the simulations of Arctic BC as indicated by sensitivity simulations in the supporting information of Wang et al. (2014).

This manuscript is not intended to be a follow-up study of Breider et al. (2014) or Breider et al. (2017). Instead, this is an independent project (hence different emission inventories and model parameters) with different objectives. Breider et al. (2014) and Breider et al. (2017) studied major near-term climate forcers including BC in the Arctic with an emphasis on their roles in Arctic warming, whereas we aim to interpret recent measurements to investigate geographical sources and their contributions to Arctic BC. Thus providing updates to Breider et al.'s research is not our purpose. The different emission years of Breider et al. (2014) likely contribute to differences in the middle troposphere due to different biomass burning. However, we have included a table (Table 1) with detailed regional anthropogenic and biomass burning emissions, and have given possible reasons for the discrepancy in the middle troposphere as the following to help readers understand our simulations.

"The remaining underestimation of 14 ng m-3 RMSE in 500-700 hPa in the HTAP+flaring simulation is possibly due to insufficient magnitude or altitude comparisons of model with ARCTAS and ARCPAC measurements (Koch et al., 2009; Wang et al., 2011; Breider et al., 2014; Eckhardt et al., 2015) as proposed based on preferential sampling by the aircraft of plumes discussed further below."

2. The conclusion section lacks discussion. Why should readers care about these new results? For example, what are the implications for their findings for regional climate in the Arctic? The introduction mentions some of the probable effects of BC on regional climate, and how the meteorological impacts of atmospheric BC likely differ with altitude. What does this altitude

variation in forcing mean for Arctic haze of Asian origin? In addition, Brieder et al. (2017) suggests that the 1980-2010 trends in Arctic haze have contributed to regional warming. How do the new results build on Brieder et al. (2017)? How are emissions in Asia projected to change in the future, and what are the probable consequences for Arctic climate? Is gas flaring around the Arctic expected to ramp up in future decades?

Response: Thanks for the suggestion. We have included more discussions in the conclusion as the following:
"The increasing BC fraction from eastern and southern Asia at higher altitudes could have significant implications for the Arctic warming by extending the trend in increasing BC radiative forcing efficiency found by Breider et al. (2017) driven by strong increase with altitude of the direct radiative forcing of BC (Zarzycki and Bond, 2010; Samset and Myhre, 2015). Besides, anthropogenic emissions of BC in southern Asia are projected to increase under several IPCC scenarios (Streets et al., 2004; Bond et al., 2013). The climate implications of BC emissions within the Arctic are concerning given their disproportionate warming effects and the potential for increasing Arctic shipping activity as ice cover declines (Sand et al., 2013)."

3. The introduction lacks key information but is nonetheless too long. First, the authors should describe what is known about the seasonal variation of transport to the Arctic at the beginning of the paper. As is, this information appears scattered through the paper as a kind of recurring explanation for the modeled results. It would be easier for the reader to encounter this information in a succinct paragraph in the beginning, and then be reminded of how transport influences Arctic as the results emerge.

That said, the authors should condense much of the other background information in the introduction, beginning at line 14 on page 3 and continuing to the end of that section. For example, the reader doesn't need to know every published estimate of the influence of biomass burning on Arctic BC. Details of the Arctic aircraft campaigns can be saved for later in the paper.

Response: Thanks for the suggestion. We have included the description of transport to the Arctic in the introduction as the following and have condensed the other background information.

"Analysis of observations have revealed that Arctic BC is primarily transported from regions outside the Arctic (Klonecki et al., 2003; Stohl, 2006). In winter, northern Eurasia is the primary source where air masses are cold enough to penetrate the polar dome into the Arctic lower troposphere (Stohl, 2006). Air masses from the relatively warm mid-latitudes (i.e. North America and Asia) are forced to ascend above the polar dome to the Arctic middle and upper troposphere (Law and Stohl, 2007). In spring, the warming of the surface leads to higher potential temperature over the Arctic and the northward retreat of the polar dome, facilitating the transport of air masses from mid-latitude regions to the Arctic (Stohl, 2006). However, large uncertainties remain in sources and geographical contributions to Arctic BC that require additional interpretation of observations to address."

4. The authors make much of recent increases in Asian BC emissions, but use anthropogenic emissions only for 2010 and GFED emissions for 2009, 2011, and 2014. These emissions are applied to GEOS-Chem simulations driven by 2009, 2011, and 2015 meteorological fields. The reader is curious if there are implications in using constant anthropogenic emissions and GFED emissions from a mismatched year. Also of interest is whether the authors see much interannual

variation in transport over the three model years.

Response: The time frame for the "recent" increase in Asian BC emissions is from 2000s to 2010. We have clarified this as the following in the manuscript:
"The main difference is due to emission trends such that our anthropogenic BC emissions from eastern and southern Asia are generally 30 % higher than those in earlier studies (e.g. Shindell et al., 2008; Sharma et al., 2013) due to rapid development since 2000 and that our anthropogenic BC emissions in Europe are half those in prior studies due to European emission controls.".

We assume no significant change of Asian BC emissions from 2010 to 2015 because Asian BC emission growth plateaued after 2010 (Crippa et al., 2016). We also assume that using GFED 2014 emissions for 2015 simulation has little influence on our results because no abnormal forest fires have been reported for 2014 and 2015. These assumptions have been included in the manuscript on page 8 line 12-13 and page 9 line 22-23.

We do not see much inter-annual variation in transport over the three model years because the simulated vertical profiles of 2009 and 2011 campaign years are similar to each other and that the contribution from eastern and southern Asia pattern remain similar. Both 2009 and 2011 profiles show uniform (coefficient of variance of 0.08 for 2009 and 0.13 for 2011) distribution below 700 hPa and larger variation above 700 hPa. The 2015 profile exhibits a distinct enhancement in the middle troposphere that may be affected by plumes.

Minor criticisms.

Page 1, line 16. Run-on sentence.

Response: We have revised it to "Black carbon (BC) contributes to the Arctic warming, yet sources of Arctic BC and their geographic contributions remain uncertain".

Page 2, line 28. What is meant by "near-surface"?

Response: We have revised it to "Near-surface (< 1 km) BC particles".

Page 4, line 11. Reader is curious why published BC measurements may be biased. Section 2.1. Years of measurements should be stated.

Response: We have revised the sentence to "Furthermore, evidence is emerging that the BC observations to which many prior modeling studies compared may have been biased by 30 % (Sinha et al., accepted) or a factor of 2 (Sharma et al., 2017) due to other absorbing components in the atmospheric aerosol."

Years of measurements have been included in Sect. 2.1.

Page 9, line 3. The authors should consider a table providing BC emissions by region, as in Breider et al. (2014).

Response: Done.

Page 12, line 25. Reader is confused why the measurements at Ny Alesund are halved.

Response: The measurements at Ny-Ålesund were not halved by us. We have clarified this as
"Restricting measurements to common years changes monthly means by less than 13 %, except for a 40 % change at Ny-Ålesund in April that arises from limited data coverage in common years since PSAP measurements for April at Ny-Ålesund is not available in 2009."

Page 20, line 20. How "substantially" are shipping emissions expected to increase and over what time frame?

Response: We have clarified these in the manuscript as "This source is expected to increase by 16 % by 2050. (Winther et al., 2014)".

Page 20, line 25. The authors state: "The main difference is due to emission trends that our anthropogenic emissions from eastern and southern Asia are generally 30% higher than those in other studies." Are these increases due to increased development in Asia? Please remind the reader what time frame is being considered here.

Response: We have clarified these in the manuscript as "The main difference is due to emission trends that our anthropogenic emissions from eastern and southern Asia are generally 30 % higher than those in earlier studies (e.g. Shindell et al., 2008; Sharma et al., 2013) due to rapid development since 2000".

Page 21, lines 12-21. Using the adjoint, the authors find that emissions as far south as the Indo-Gangetic Plain influence Arctic BC. This is new information. How confident are the authors of the GEOS-Chem simulation in this region (and in China)?

Response: Emissions are a major source of uncertainty in the simulation of the contributions from the Indo-Gangetic Plain and China to the Arctic. The emissions in China and the Indo-Gangetic Plain in the HTAP v2 inventory originate from the MICS Asia inventory that represent the best estimate of emissions in Asia (Li et al., 2017). However, uncertainties still exist, so we suggested further investigations in the conclusion as the following:
"The considerable impact of emissions from China and Indo-Gangetic Plain on the Arctic deserves further investigation."

Figure 1. Are these total BC emissions or just anthropogenic?

Response: We have revised the caption to "The colormap indicates annual total BC emissions averaged over 2009, 2011 and 2015 as used in the GEOS-Chem simulation."

Figure 3. Error bars on most measurements look very small. Please check the magnitudes. What are the years of the measurements?

Response: Error bars of measurements at Alert were not included for the clarity of the figure, but error bars of the best estimate of BC measurements (mean EC and rBC measurements) at Alert were included. The error bar magnitudes have been corrected at Barrow and Ny Alesund. Measurement years are included in the legend.

Figure 4. Please put error bars on the ground-based measurements.

Response: Done. But only one year (2009) ground measurement is available for Ny Alesund,

so no error bar presents in the figure.

Figure 5. Please state in the caption the year and season of the measurements and model results.

Response: Done.

Figure 7. Consider making a 4-panel plot with two new panels showing the stacked percent contribution of each region to the BC at different altitudes. The two new panels would have altitude on the y-axis, and percent contribution from 0-100% along the x-axis. In any case, the two existing panels look strangely elongated.

Response: Thanks for the suggestion. We have changed the figure to the 4-panel plot as suggested.

Figure 8. Measurements should have error bars.

Response: Done.

Table 1. Table should include footnotes so that the reader does not have to scramble through the text to learn what the different scenarios mean. Also, it's not that clear that the vertical RMSE is meaningful since it varies so much with altitude.

Response: Thanks for the suggestion. We have included footnotes in Table 2. The vertical RMSE for simulations with different emissions shows the improvement with seasonal residential heating and flaring emissions in simulating vertical distributions of BC concentrations.

References:

Breider, T. J., Mickley, L. J., Jacob, D. J., Wang, Q., Fisher, J. A., Chang, R. Y.-W. and Alexander, B.: Annual distributions and sources of Arctic aerosol components, aerosol optical depth, and aerosol absorption, J. Geophys. Res. Atmos., 119(7), 4107–4124, doi:10.1002/2013JD020996, 2014.

Breider, T. J., Mickley, L. J., Jacob, D. J., Ge, C., Wang, J., Payer Sulprizio, 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(6), 3573–3594, doi:10.1002/2016JD025321, 2017.

Crippa, M., Janssens-Maenhout, G., Dentener, F., Guizzardi, D., Sindelarova, K., Muntean, M., Van Dingenen, R. and Granier, C.: Forty years of improvements in European air quality: regional policy-industry interactions with global impacts, Atmos. Chem. Phys., 16(6), 3825–3841, doi:10.5194/acp-16-3825-2016, 2016.

Li, M., Zhang, Q., Kurokawa, J., Woo, J.-H., He, K., Lu, Z., Ohara, T., Song, Y., Streets, D. G., Carmichael, G. R., Cheng, Y., Hong, C., Huo, H., Jiang, X., Kang, S., Liu, F., Su, H. and Zheng, B.: MIX: a mosaic Asian anthropogenic emission inventory under the international collaboration framework of the MICS-Asia and HTAP, Atmos. Chem. Phys., 17(2), 935–963, doi:10.5194/acp-17-935-2017, 2017.

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(23), 12453–12473, doi: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. Atmos., 119(22), 12,888-12,909, doi:10.1002/2014JD022297, 2014

Authors' Response to Comments from Referee #3

We thank anonymous referee #3 for the helpful suggestions and questions, which have led to valuable improvements in our manuscript. Reponses to these comments are provided below.

GENERAL COMMENTS

The present paper describes results from air campaigns and an effort to understand the transport and origin of BC from different regions and emission sectors through modelling. The paper does not really include any new story information about the transport, missing sources or origin of BC to the Arctic. This is very obvious, because the authors frequently justify most of their sentences with references
throughout the whole manuscript. So, what the authors claim in the present study has been already well described in previously published articles, although values are different. For instance, the contribution from Europe or Asia to BC in an Arctic station may differ in the present manuscript compared to other paper. However, this is still nothing new, as it may differ due to the use of different models or due to different lifetime of BC within each of the models or for any other reason that induces modelling
uncertainty. Nevertheless, measurements in the Arctic are very useful and generally lack and especially measurements from air campaigns. So, I would suggest that the authors should focus more on the measurements from the air campaigns and try to shorten the manuscript by removing the trivial statements about issues that have already been published elsewhere. Since the editor thinks that the present manuscript is novel enough to get out for a review, then I think that it deserves publication. It is very well written and it was a pleasure to reading it, although improvements can be applied, in order to be clearer and more concise. I could not find any weak point except those that I already pointed out. Everything flows well in it. Therefore, I only have some minor comments.

Response: Thanks for these. Novelties of this paper include 1) interpretation of new airborne measurements at Alert in the Arctic, 2) the first comparison with a chemical transport model of rBC measrements at Alert, 3) more accurate surface measurements used for model evaluation and source attribution, 4) improved understanding of how differet emission inventories affect comparison with observations, 5) source attribution using the adjoint of the GEOS-Chem model to understand the importance of specific sources, and 6) identification of the Tarim oilfield and Indo-Gangetic plain as important sources. We have revised the manuscript to highlight these novelties and have condensed less novel material.

SPECIFIC COMMENTS

Please shorten the Abstract. E.g., Page 2 – Line 9-10 is a trivial statement and can be removed from the abstract. Please follow this pattern and mention the most important points of your paper only and not all the conclusions!

Response: Done.

P 5 – L 16: Should it be "state-of-the-art" instead of "state-of-the-science"?

Response: Revised.

P 5 – L 15 until the end of paragraph: You are describing methodology in the Introduction. Please remove all these details from this chapter!

Response: Done.

P 5 – L 21 until the end of paragraph: Again you describe methodological issues that do not belong there, but rather in the next section of your paper.

Response: We have condensed this paragraph but we think it deserves a brief description of the motivation to use this method in the introduction because the adjoint of the GEOS-Chem simulation results are a highlight of this manuscript.

P 6 – L 26: EMEP and WDCA are mentioned for the first time in the manuscript and need explanation. Please do the same elsewhere (e.g., SP2).

Response: We have written out EMEP, WDCA and SP2 where they appear for the first time in the manuscript

P16 – comments on Fig. 6: I had really hard times to follow this part and I think it is due to the poor labeling on the Figure. Therefore, I would suggest to put 6 small letters on each of the figures and point them in the text, so the reader knows to which of the figures you refer in the text.

Response: Done.

P16 – L19-20 and L21: You are talking about the origin of the plume that arrives at the hotspot areas, but evidence is lacking. You have to point to respective figures somewhere or then remove these lines, because they cannot stand alone without any justification.

Response: Origin removed.

P17 – L5-19: In my opinion column concentrations at the bottom panels of Fig.6 there do not say much. I think it is necessary to show the same maps with emissions. Preferably, add another panel (bottom) and show emissions in the same periods as with the column concentrations.

Response: Thanks for the suggestion. We have included emissions at the bottom panel of Fig. 6.

Source attribution of Arctic black carbon constrained by aircraft and surface measurements

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[2]Harvard-Smithsonian Center for Astrophysics, Cambridge, MA, USA

[3]Atmospheric Science and Technology Directorate/Science and Technology Branch, Environment and Climate Change Canada, Toronto, Ontario, Canada

[4]Department of Chemistry, University of Toronto, Toronto, Canada

[5]Alfred Wegener Institute, Helmholtz Centre for Polar and Marine Research, Bremerhaven, Germany

[6]Department of Mechanical Engineering, University of Colorado, Boulder, CO, USA

Abstract

Black carbon (BC) contributes to Arctic warming, yet sources of Arctic BC and their geographic contributions remain uncertain. We interpret a series of recent airborne (NETCARE 2015, PAMARCMiP 2009 and 2011 campaigns) and ground-based measurements (at Alert, Barrow and Ny-Ålesund) from multiple methods (thermal, laser incandescence and light absorption) with the GEOS-Chem global chemical transport model and its adjoint to attribute the sources of Arctic BC. This is the first comparison with a chemical transport model of refractory BC (rBC) measurements at Alert. The springtime airborne measurements performed by the NETCARE campaign in 2015 and the PAMARCMiP campaigns in 2009 and 2011 offer BC vertical profiles extending to above 6 km across the Arctic and include profiles above Arctic ground monitoring stations. Our simulations with the addition of seasonally varying domestic heating and of gas flaring emissions are consistent with ground-based measurements of BC concentrations at Alert and Barrow in winter and spring (rRMSE < 13 %), and with

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airborne measurements of the BC vertical profile across the Arctic (rRMSE=17 %) except for an underestimation in the middle troposphere (500-700 hPa).

Sensitivity simulations suggest that anthropogenic emissions in eastern and southern Asia have the largest effect on the Arctic BC column burden both in spring (56 %) and annually (37 %), with the largest contribution in the middle troposphere (400-700 hPa). Anthropogenic emissions from northern Asia contribute considerable BC (27 % in spring and 43 % annually) to the lower troposphere (below 900 hPa). Biomass burning contributes 20 % to the Arctic BC column annually.

At the Arctic surface, anthropogenic emissions from northern Asia (40 % - 45 %) and eastern and southern Asia (20 % - 40 %) are the largest BC contributors in winter and spring, followed by Europe (16 % - 36 %). Biomass burning from North America is the most important contributor to all stations in summer, especially at Barrow.

Our adjoint simulations indicate pronounced spatial heterogeneity in the contribution of emissions to the Arctic BC column concentrations, with noteworthy contributions from emissions in eastern China (15 %) and western Siberia (6.5 %). Although uncertain, gas flaring emissions from oilfields in western Siberia could have a striking impact (13 %) on Arctic BC loadings in January, comparable to the total influence of continental Europe and North America (6.5 % each in January). Emissions from as far as the Indo-Gangetic Plain could have a substantial influence (6.3 % annually) on Arctic BC as well.

1. Introduction

The Arctic has warmed rapidly over the last few decades at a rate about twice the global mean (AMAP, 2011; AMAP, 2015). By directly absorbing solar radiation, black carbon (BC) contributes substantially to the warming, impacting the Arctic in multiple ways (Flanner et al., 2007; Ramanathan and Carmichael, 2008; Shindell and Faluvegi, 2009; Bond et al., 2013; Sand et al., 2016). Near-surface (< 1 km) BC particles over a highly reflective surface (i.e. snow and ice in the Arctic) warm the atmosphere, and

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subsequently the surface (Shaw and Stamnes, 1980; Quinn et al., 2008). BC particles well above the surface warm the layer in which they reside and increase the stability of the Arctic atmosphere (e.g. Brock et al., 2011). Deposition of BC onto snow and ice can reduce surface albedo and enhance light absorption by snow and ice (Wiscombe and Warren, 1980; Chýlek et al., 1983), and trigger chain reactions involving the acceleration of snow aging (Clarke and Noone, 1985; Hansen and Nazarenko, 2004), leading to accelerated melting (Quinn et al., 2008; Namazi et al., 2015). The modified local radiative balance exerted by deposited BC has the potential to further affect climate at a larger scale (Flanner et al., 2007; Doherty et al., 2010).

Analysis of observations have revealed that Arctic BC is primarily transported from regions outside the Arctic (Klonecki et al., 2003; Stohl, 2006). In winter, northern Eurasia is the primary source where air masses are cold enough to penetrate the polar dome into the Arctic lower troposphere (Stohl, 2006). Air masses from the relatively warm mid-latitudes (i.e. North America and Asia) are forced to ascend above the polar dome to the Arctic middle and upper troposphere (Law and Stohl, 2007). In spring, the warming of the surface leads to higher potential temperature over the Arctic and the northward retreat of the polar dome, facilitating the transport of air masses from mid-latitude regions to the Arctic (Stohl, 2006). However, large uncertainties remain in sources and geographical contributions to Arctic BC that require additional interpretation of observations to address.

Elevated BC concentrations in the Arctic especially in winter and spring have been observed over the past few decades (Delene & Ogren, 2002; Sharma, et al., 2006; Eleftheriadis et al., 2009; Yttri et al., 2014). Some studies attributed the surface BC primarily to emissions in high-latitude regions including Europe and northern Eurasia (e.g. Stohl, 2006; Shindel et al., 2008; Hirdman et al., 2010; Wang et al., 2014) while others found eastern and southern Asia had the largest contribution (Koch and Hansen, 2005; Ikeda et al., 2017). Some studies suggested that Europe was the dominant source of BC aloft (Stohl, 2006; Huang et al, 2010b) while others found eastern and southern Asia was the most important source (Sharma et al., 2013; Wang et al., 2014; Ikeda et al., 2017)

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in the middle troposphere. Recent work by Stohl et al. (2013) and Sand et al. (2016) raised questions about prior studies by identifying the importance of seasonally varying residential heating and by suggesting a significant overlooked source from gas flaring in high-latitude regions. In addition to anthropogenic emissions, biomass burning is another important source of Arctic BC (Stohl et al., 2007; Warneke et al., 2009; Yttri et al., 2014; Evangeliou et al., 2016), yet its contribution remains uncertain. Furthermore, evidence is emerging that the BC observations to which many prior modeling studies compared may have been biased by 30 % (Sinha et al., accepted) or a factor of 2 (Sharma et al., 2017) due to other absorbing components in the atmospheric aerosol. Additional attention is needed to these issues.

BC emissions in mid- and low-latitude regions increase the Arctic climate forcing efficiency by altering the BC vertical distribution (Breider et al., 2017). Thus it is also crucial to quantify the source contributions to the vertical distribution of Arctic BC. However, vertical profiles in the Arctic have been scarce (Jacob et al., 2010; Brock et al., 2011) and anomalously influenced by biomass burning (Warneke et al., 2009). The NETCARE (Network on Climate and Aerosols: Addressing Key Uncertainties in Remote Canadian Environments, http://www.netcare-project.ca) aircraft campaign in 2015 and the PAMARCMiP (Polar Airborne Measurements and Arctic Regional Climate Model Simulation Project) aircraft campaigns in 2009 and 2011 offer a new dataset of BC measurements across the Arctic.

Source attributions of pollution in the Arctic are commonly estimated by back-trajectory analysis (Huang et al., 2010a; Harrigan et al., 2011; Barrett et al., 2015; Liu et al., 2015), and by sensitivity simulations using chemical transport models (Fisher et al., 2010; Sharma et al., 2013; Mungall et al., 2015; Evangeliou et al., 2016). These traditional approaches have been insightful, but suffer from coarse regional estimates of the source location. The adjoint of a global chemical transport model (Henze et al., 2007) efficiently determines the spatially resolved source contribution to receptor locations by calculating the gradient of a cost function (e.g. Arctic column BC concentrations) with respect to the perturbations of the initial conditions (e.g. emissions). This approach has been

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successfully applied to quantify source contributions to Arctic surface BC in April 2008 (Qi et al., 2017b). We extend the application of this method to investigate the seasonal and annual responses of Arctic column BC to changes in regional emissions.

In this study, we first evaluate the BC concentrations simulated with the GEOS-Chem global chemical transport model with surface and aircraft measurements in the Arctic to assess the quality of different emission representations. Then sensitivity simulations are conducted to assess the regional contributions to the observed BC in the Arctic. We subsequently use the adjoint of the GEOS-Chem model to investigate the spatially resolved sensitivity of Arctic BC column concentrations to global emissions.

2. Method

2.1 Surface measurements of BC in the Arctic

Surface BC mass concentrations are measured at three Arctic stations: Alert (Nunavut, Canada; 62.3° W, 82.5° N), Barrow (Alaska, USA; 156.6° W, 71.3° N) and Ny-Ålesund (Svalbard, Norway; 11.9° E, 78.9° N). Station locations are shown in Fig. 1. Following the recommendations of Petzold et al. (2013), measurements of BC based on light absorption are here referred to as equivalent BC (EBC); measurements based on a laser induced incandescence technique (e.g. single particle soot photometer; SP2) are referred to as refractory BC (rBC); and measurements based on a thermal volatilization in an oxygen-enriched environment are referred to as elemental carbon (EC).

EBC mass concentrations derived from an AE-31 Aethalometer (Magee Scientific Inc.) at Alert for 2011-2013 are obtained from Environment and Climate Change Canada and those at Barrow for 2010-2014 and Ny-Ålesund for 2009-2010 are obtained from the EMEP (European Monitoring and Evaluation Programme) and WDCA (World Data Centre for Aerosols) database (http://ebas.nilu.no/). The Aethalometer measures the absorption of light at 880 nm transmitted through particles that accumulate on a quartz fiber filter and relates the change of light absorption to light absorption coefficients (σ_{ap})

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using Beer's Law. EBC mass concentrations are derived from σ_{ap} by adopting a mass absorption cross-section (MAC) of 16.6 m^2 g^{-1} at all stations. This MAC value is recommended by the manufacturer for Model AE31 at 880 nm to account for absorption by BC and additional light scattering by both particles and filter fibers.

EBC mass concentrations are also derived from a particle soot absorption photometer (PSAP, Radiance Inc.) that operates on a similar principle to the Aethalometer at the three stations. PSAP measures the absorption of light at 530 nm. σ_{ap} data at Alert for 2011-2013 are obtained from Environment and Climate Change Canada, and σ_{ap} data at Barrow for 2009-2015 and Ny-Ålesund for 2009-2014 are obtained from the EMEP and WDCA database (http://ebas.nilu.no/). σ_{ap} has been corrected for scattering following Bond et al. (1999) and is further reduced by 30 % at all stations following Sinha et al. (accepted). σ_{ap} values less than the detection limit (0.2 Mm^{-1}) are excluded. Recent evidence is emerging that the MAC is lower than the traditional value of 10 m^2 g^{-1}, with recent effective MAC values ranging from 8 m^2 g^{-1} (Sharma et al., 2017) to 8.7 m^2 g^{-1} (Sinha et al., accepted). We adopt the average of these two values (8.4 m^2 g^{-1}) for application to PSAP measurements at all three sites.

Two additional measurements of BC mass concentrations are available at Alert for 2011-2013: rBC and EC. rBC is measured via laser induced incandescence by an SP2 instrument (Droplet Measurement Technologies Inc., Boulder, CO). The SP2 uses a high intensity laser (Ni:YAG) operating at 1064 nm wavelength to selectively heat individual particles up to 4000K. At such high temperature, the non-refractory components evaporates and rBC mass is proportional to the intensity of the emitted incandescent light. The incandescence signal is calibrated using Aquadag particles of known size selected with a differential mobility analyzer (Sharma et al., 2017). The detection range of the SP2 at Alert spans approximately between 75 nm and 530 nm volume-equivalent diameter (Sharma et al., 2017), assuming an rBC density of 1.8 g cm^{-3} (Bond and Bergstrom, 2006). A lognormal function fit over the range of 80-225 nm is applied to calculate rBC concentrations over the 40-1000 nm size range that increases the rBC concentrations by about 50 % (Sharma et al., 2017).

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EC measurements at Alert are inferred from weekly-integrated samples of particles collected on quartz filters with a 1µm upper size cut and analyzed using an in-house thermal technique referred to as EnCan-total-900 (Huang et al., 2006). The EnCan-total-900 method has three temperature steps with different redox conditions: 550°C and 870°C under pure helium and 900°C under helium + 10 % oxygen. The retention times are 600 seconds at 550°C for organic carbon (OC), 600 seconds at 870°C for pyrolysis of OC and carbonate carbon, and 420 seconds at 900°C for EC. The 870°C pure helium step releases pyrolysis OC and carbonate carbon to minimize the effect of OC charring on EC.

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2.2 Aircraft measurements of BC in the Arctic

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Aircraft measurements are obtained from a series of recent campaigns that offer new measurements in the lower troposphere across the Arctic. The PAMARCMiP campaigns conducted springtime surveys of sea ice thickness, aerosol and meteorological parameters along the coast of the western Arctic onboard the Alfred Wegener Institute (AWI) Polar 5 aircraft. Data from two campaigns in April 2009 (Stone et al., 2010) and March 25th – May 6th 2011 (Herber et al., 2012) are used here. The NETCARE campaign in April 2015 continued and extended the PAMARCMiP campaigns observations using the Polar 6 aircraft. Flight tracks of each campaign are shown in Fig. 1. All three campaigns traveled along similar routes across the western Arctic and near long-term ground monitoring stations in the Arctic (Alert, Barrow and Ny-Ålesund). Measurements of rBC mass concentrations during all three campaigns were performed with the state-of-the-art SP2 (Droplet Measurement Technologies Inc., Boulder, CO) instrument. The SP2 used during the PAMARCMiP campaigns was previously described in Stone et al. (2010). The NETCARE 2015 campaign used the AWI's 8-channel SP2 with a detection range of 75 – 700 nm of volume-equivalent diameter (assuming a particle density of 1.8 g cm^{-3}) without corrections for particles outside the size range. The incandescence signal was calibrated with particles of Fullerene soot size selected with a differential mobility analyzer. The spatial and multi-year coverage of airborne measurements during these campaigns offer comprehensive representation of Arctic BC.

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2.3 Simulations of Arctic BC

We use the GEOS-Chem global chemical transport model (version 10-01; http://geos-chem.org/) and its adjoint (version 35) to simulate Arctic BC concentrations and their sensitivities to local emissions.

Figure 1 shows the annual mean BC emissions in our GEOS-Chem simulation averaged over 2009, 2011 and 2015. We develop the simulation here to use global anthropogenic emissions of BC from version 2 of the HTAP (Hemispheric Transport of Air Pollution; http://www.htap.org/) emission inventory for 2010 (Gilardoni et al., 2011; Janssens-Maenhout et al., 2015) with regional overwrites over the United States (NEI 2011) for the most recent year (2011). Global and regional BC emissions remain largely constant after 2010 (Crippa et al., 2016). The HTAP inventory is a compilation of different official emission inventories from MICS-Asia, EPA-US/Canada and TNO-Europe data, gap-filled with global emission data of EDGARv4.1. The HTAP contains BC emissions from all major sectors, including energy and industrial production, transport and residential combustion.

Table 1 contains the annual regional BC emissions used in the simulation. Total BC emissions from eastern and southern Asia exceed by more than a factor of 4 the BC emissions from either North America or Europe.

Figure 2 shows annual HTAP BC emissions and its seasonal variation over the Arctic and the Northern Hemisphere. The Bond et al. (2007) emission inventory for 2000 is included for comparison, since it has been widely used in modeling studies of Arctic BC (Shindell et al., 2008; Koch et al., 2009; Liu et al., 2011; Wang et al., 2011; Breider et al., 2014; Breider et al., 2017; Qi et al., 2017a; Qi et al., 2017b). The Bond et al. (2007) inventory is based on energy consumption in 1996 and contains similar emission sectors as in the HTAP. The HTAP annual emissions over the Northern Hemisphere exceed those in Bond et al. (2007) by 30 %, with a substantial difference in China and India where HTAP

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emissions are double those of Bond et al. (2007). A considerable increase of global energy consumption since 2001 especially in China and India contributes to the difference (Zhang et al., 2009; Li et al., 2017). Both inventories have low BC emissions within the Arctic. Figure 2 also shows the seasonal variation of HTAP emissions that are high in winter and spring and low in summer over the Northern Hemisphere, owing to the seasonal variation of emissions from residential heating in the HTAP. Bond et al. (2007) emissions are non-seasonal.

We also include additional BC emissions from gas flaring in the oil and gas industry taken from version 5 of the ECLIPSE (Evaluating the climate and Air Quality Impacts of short-Lived Pollutants) emission inventory (Klimont et al., 2016; http://eclipse.nilu.no). Gas flaring emissions of BC are calculated based on gas flaring volumes developed within the Global Gas Flaring Reduction initiative (Elvidge et al., 2007, 2011) with emission factors derived on the basis of particulate matter and soot estimates from CAPP (2007), Johnson et al. (2011) and US EPA (1995). Despite the small percentage (~5 %) of flaring in total anthropogenic BC emissions over the Northern Hemisphere, flaring from Russia alone accounts for 93 % of total anthropogenic BC emissions within the Arctic in the ECLIPSE inventory.

Emissions from biomass burning are calculated from the GFED4 (Global Fire emissions Database version 4) inventory (Giglio et al., 2013). The GFED4 combines satellite information on fire activity and vegetation productivity to estimate globally gridded monthly burned area (including small fires) and fire emissions. We use emissions for 2009, 2011 and 2014 (the most recent year available) for the simulations of 2009, 2011 and 2015. The mismatch of emission year is unlikely to strongly influence the simulation as no abnormal fire activities were reported for 2014 and 2015. Biomass burning emissions are injected into the boundary layer in our simulations.

As discussed in Sect. 2.1, measurements of BC depend on the analysis method. However, it is ambiguous what analysis method is used to derive BC emission factors or BC speciation factors in particulate matter in various emission inventories (Bond et al.,

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2013). Therefore, we directly compare simulated BC concentrations with the best estimate of measured atmospheric BC.

The simulation of BC in GEOS-Chem is initially described in Park et al. (2003). BC emitted from all primary sources is in hydrophobic and hydrophilic states with a constant conversion time of one day. Dry deposition of BC aerosols adopts a standard resistance-in-series scheme as described in Zhang et al. (2001) with improvements on BC dry deposition velocity over snow and ice following Fisher et al. (2010) and Wang et al. (2011). Wet deposition of BC aerosols is initially described in Liu et al. (2001) and developed by Wang et al. (2011) to distinguish between liquid cloud (T > 268 K) in which 100 % hydrophilic BC is removed and ice cloud (T < 268 K) in which only hydrophobic BC is removed.

Our GEOS-Chem simulations are driven by Modern-Era Retrospective Analysis for Research and Applications (MERRA) meteorological fields from the NASA Global Modeling and Assimilation Office (GMAO) at 2° × 2.5° spatial resolution with 47 vertical levels from the surface to 0.01 hPa. We conduct simulations for 2009, 2011 and 2015 with a 10-minute operator duration for transport and a 20-minute operator duration for chemistry as recommended by Philip et al. (2016). We initialize the model with a 6-month spin-up before each simulation to remove the effects of initial conditions on aerosol simulations.

We conduct sensitivity simulations using the GEOS-Chem model to quantify the contributions of regional emissions to Arctic (hereafter refer to the region north of 66.5° N) BC concentrations by excluding the regional anthropogenic source. Regions are North America (180° W-50° W, 0° N – 80° N), Europe (50° W- 50° E, 30° N – 80° N), eastern and southern Asia (50° E – 150° E, 0° N – 50° N) and northern Asia (50° E – 180° E, 50° N – 80° N), as outlined in Fig.1. We also conduct sensitivity simulations to quantify the contribution of biomass burning from North America and from the rest of the world to Arctic BC concentrations. These simulations are initialized with a 6-month spin-up as well.

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We also apply the GEOS-Chem adjoint model to quantify the spatially resolved sensitivity of Arctic BC column concentrations to local emissions. A detailed description of the adjoint model is given in Henze et al. (2007). Here we briefly describe the concept in the context of our study. The adjoint model offers a computationally efficient approach to calculate the sensitivity of a model output scalar, the cost function, to a set of model input parameters such as emissions. In this study, we define the cost function as the column concentrations of BC north of 66.5° N. The adjoint model calculates the partial derivatives of this cost function with respect to the modeled atmospheric state in each model grid box at each time step. This calculation is performed iteratively backward in time through transport toward emissions to yield the sensitivity of the cost function with respect to emissions.

Our adjoint simulation is driven by GEOS-5 meteorology at 2° × 2.5° spatial resolution with 47 vertical levels from the surface to 0.01 hPa for 2011. Differences between MERRA meteorological fields that are used in the forward model and GEOS-5 meteorological fields that are used in the adjoint are negligible (r^2 = 0.99 for Arctic column BC concentrations for 2011) in the simulation of BC. Although the adjoint simulation is based on an earlier version (v8) of the GEOS-Chem model than the forward model version (v10-01) used in this study, the differences in BC concentrations at Arctic stations that are simulated with the adjoint and with the forward model are within 15 % (Qi et al., 2017b).

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2.4 Statistics

To assist with the evaluation of simulations, we define root mean square error (RMSE) and relative root mean square error (rRMSE) as

$$\mathrm{RMSE}=\sqrt{\frac{1}{N}\sum_{i=1}^{N}(C_m(i)-C_o(i))^2} \quad (1)$$

$$\mathrm{rRMSE}=100\,\%\times\frac{\mathrm{RMSE}}{\frac{1}{N}\sum_{i=1}^{N}C_m(i)} \quad (2)$$

where $C_m(i)$ is the model simulated concentration and $C_o(i)$ is the measurement concentration. N is the number of measurements.

3. Results

3.1 Evaluation of GEOS-Chem simulated BC concentrations in the Arctic

Figure 3 shows the seasonal variation of BC concentrations from measurements and simulations at the Alert, Barrow and Ny-Ålesund stations. Different black line types indicate different instruments. Slight differences exist in sampling periods from different instruments. Restricting measurements to common years changes monthly means by less than 13 %, except for a 40 % change at Ny-Ålesund in April that arises from limited data coverage in common years since PSAP measurements for April are not available at Ny-Ålesund in 2009. At Alert, a diversity of instruments offers valuable insight into the suite of BC measurements throughout the Arctic, and perspective on previous model comparison with only one instrument type. EBC concentrations measured by the Aethalometer are biased high by a factor of 2 relative to rBC measurements, due to the presence of absorbing substances other than BC (e.g. brown carbon and mineral dust), extinction issues associated with the filter matrix and uncertainties in MAC values (Sharma et al., 2017). EC concentrations are lower than EBC concentrations from the Aethalometer, yet still high relative to rBC partly due to the presence of pyrolysis OC and carbonate carbon (Sharma et al., 2017). PSAP EBC concentrations are close to the average of EC and rBC concentrations throughout the year. At Barrow, EBC concentrations from the Aethalometer are higher than those from the PSAP, especially in summer when the Aethalometer shows a pronounced increase in concentrations to around 55 ng m^{-3}, whereas PSAP measurements reach a minimum for the year of 10 ng m^{-3}. The summer peak is also observed in Aethalometer EBC measurements at 370 nm that is sensitive to brown carbon, indicating the influence of biomass burning. Unintentional exclusion of biomass burning plumes in the local pollution data screening performed for PSAP measurements at Barrow could contribute to the bias between the PSAP and the Aethalometer there (Stohl et al., 2006).

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Following Sharma et al. (2017), we treat the best estimate of measured BC surface concentrations at Alert as the average of rBC and EC measurements, as shown by the thick black line with squares in Fig. 3. Since the PSAP EBC concentrations are close to the average of rBC and EC measurements throughout the year at Alert, we adopt the PSAP EBC measurements as the best estimate of surface BC at Barrow and Ny-Ålesund. The seasonal variations of surface BC at the three sites show similar features, characterized by higher concentrations in winter and early spring than in summer. At Ny-Ålesund, peak months are March and April, slightly later than at the other sites (January and February). BC concentrations at Ny-Ålesund are generally lower than those at the other sites.

The surface BC concentrations from measurements are used to constrain emissions in the simulations. Table 2 summarizes the RMSE and rRMSE between measurements and different simulations. The green line in Fig. 3 shows simulated surface BC concentrations using anthropogenic emissions of BC from the Bond et al. (2007) non-seasonal emission inventory. Stohl et al. (2013) found that accounting for BC emissions from gas flaring and from seasonal variation of residential heating improved their simulation with a particle dispersion model (FLEXPART) during winter and early spring. Our simulation at Alert and Barrow in winter and spring is also improved by using the HTAP emissions that include seasonal variation of residential heating and by adding flaring emissions to the HTAP inventory, decreasing the bias by about a factor of 2 and reducing the rRMSE to 5.6 % at Alert and 13 % at Barrow. At Barrow all simulations show a distinct peak in July, which is partly due to the timing of biomass burning. Eckhardt et al. (2015) similarly observed enhanced concentrations in July at Barrow in three models (DEHM, CESM1-CAM5 and ECHAM6-HAM2) driven with the GFED3 inventory for biomass burning emissions. At Ny-Ålesund, all simulations overestimate measured concentrations for most of the year, potentially indicating insufficient wet deposition from riming in mixed phase clouds that occurs more frequently at this site (Qi et al., 2017a).

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Figure 4 shows vertical profiles of BC concentrations at Alert and Ny-Ålesund averaged from the NETCARE 2015, the PAMARCMiP 2009 and the PAMARCMiP 2011 campaigns, along with the best estimate of ground-based measurements of April BC concentrations averaged over 2009 and 2011. Barrow is not included here due to limited number of airborne measurements (a total of 12 measurements at all pressures). The measured profile at Alert exhibits layered structure with enhanced concentrations in the middle troposphere that are attributable to a plume on April 8th 2015 around 660-760 hPa with a peak concentration of 128 ng m^{-3}. The mean ground-based measurements of BC concentrations at Alert are higher than airborne measurements at the same pressure by ~10 ng m^{-3}. Including only rBC measurements in ground-based mean concentrations reduces the difference with airborne rBC measurements to less than 5 ng m^{-3}. At Ny-Ålesund, the measured vertical profile exhibits a zigzag shape that arises from averaging multiple years each with individual features. The mean April ground-based concentration (20 ng m^{-3}) is about half that of the airborne measurements (37 ng m^{-3}) at the same pressure.

Figure 5 shows spring vertical distributions of BC averaged over all points along the flight tracks of the three campaigns in Fig. 1 for measurements and simulations. Simulated vertical profiles of BC are coincidently sampled with airborne measurements for spring 2009, 2011 and 2015, and are averaged to the GEOS-Chem vertical resolution. The measured rBC concentrations remain roughly constant (~38 ng m^{-3}) from the surface to 700 hPa, followed by an enhancement to around 50 ng m^{-3} between 700 hPa – 500 hPa, and then a rapid decrease with altitude. This vertical distribution is similar to the measurements of the ARCTAS aircraft campaign in the Arctic in spring 2008 (Wang et al., 2011), though the magnitude of concentrations in this work is lower by a factor of about 2, likely because the Arctic was substantially influenced by strong biomass burning in northern Eurasia during the ARCTAS in spring 2008 (Warneke et al., 2009). All simulations generally represent the near constant vertical distribution of BC measurements from the surface to 700 hPa, and the decrease above 500 hPa, yet none represent the enhancement between 700-500 hPa. Despite the comparable distributions,

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the magnitudes of concentrations simulated with different emissions vary substantially. Their consistency with airborne measurements is summarized in Table 2.

Figure 5 shows that the apparent bias of 40 % rRMSE (17 ng m^{-3} RMSE) in simulated concentrations with the Bond et al. (2007) non-seasonal inventory is reduced to 27 % rRMSE (11 ng m^{-3} RMSE) by the HTAP inventory with non-seasonal residential heating. The improvement is larger aloft than near-surface, indicating that the increased BC emissions in Asia in the HTAP inventory (discussed in Sect. 2) substantially contributes to the improvement. The bias versus measurements is further reduced to 23 % rRMSE (9.4 ng m^{-3} RMSE) by the HTAP emissions with seasonal residential heating, with larger improvement below 600 hPa. Adding flaring emissions further improves the consistency (17 % rRMSE; 7.2 ng m^{-3} RMSE) with measurements at all levels with larger effects in the lower troposphere, especially near the surface where the RMSE is only 3.2 ng m^{-3}. The substantial portion (93 %) of flaring in BC emissions within the Arctic (Fig. 2) explains the larger effect near the ground. The remaining underestimation of 14 ng m^{-3} RMSE in 500-700 hPa in the HTAP+flaring simulation is possibly due to insufficient magnitude or altitude comparisons of model with ARCTAS and ARCPAC measurements (Koch et al., 2009; Wang et al., 2011; Breider et al., 2014; Eckhardt et al., 2015) as proposed based on preferential sampling by the aircraft of plumes discussed further below.

Figure 6 (A1 and A2) shows the spatial distribution of BC concentrations from aircraft measurements gridded onto the GEOS-Chem grid along with that from the HTAP+flaring simulation. The simulation represents well the spatial distribution of BC measurements, with concentrations of 30-70 ng m^{-3} near Barrow and Ny-Ålesund and lower concentrations of 20-40 ng m^{-3} near Alert, yet the simulation underestimates concentrations at three hotspots (labeled as a, b, c). Hotspot a is near Barrow along the coast of the Beaufort Sea that is affected by a plume around 800 hPa on April 6th 2011 and a plume around 500 hPa on April 20th 2015. Hotspot b is west of the Baffin Bay in Nunavut that is affected by a plume near 800 hPa on April 10th 2011. Hotspot c is near Ny-Ålesund that is caused by a plume at around 700 hPa on May 5th 2011. The

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underestimated magnitudes of these plumes, likely related to emissions or numerical diffusion, may contribute to the underestimation of BC concentrations between 500-700 hPa in Fig. 5. Fig. 6 (A3) shows mean simulated BC concentrations between 500-700 hPa in April. Concentrations are highest (~70 ng m^{-3}) in northeastern Russia and near Barrow, with a gradual decrease eastward to around 50 ng m^{-3} near Alert to reach the lowest concentrations of below 40 ng m^{-3} in the southern Arctic near Ny-Ålesund. This gradient illustrates the overall sources and transport pathways affecting BC in the Arctic middle troposphere in springtime. The next section will investigate the enhanced concentrations in northeastern Russia and their relation to sources in eastern and southern Asia.

Figure 6 (B1-B3) shows pan-Arctic spatial distributions of BC column (1000 hPa – 300 hPa) concentrations from the HTAP+flaring simulation for January, April and July. Strong spatial and seasonal variation is observed in BC columns with the highest overall concentrations in April and in the eastern Arctic. Emissions remain similar for the three months as shown in Fig. 6 C1-C3, indicating that the main reason for the seasonal variation of Arctic BC column is transport efficiency. In July, the enhanced concentrations in western Siberia due to flaring are less obvious, due to more effective wet scavenging in summer. North America exhibits remarkably high BC column in July (Fig. 6 B3) from biomass burning as will be discussed further in Sect. 3.2.

Since BC concentrations simulated with HTAP+flaring exhibit overall consistency with the measured seasonal variation, and the measured spatial distributions, we use this inventory in the following simulations for source attributions.

3.2 Source attribution of BC in the Arctic

Figure 7 (top left) shows the contribution of anthropogenic emissions from regions defined in Fig. 1, and of biomass burning from North America and the rest of the world, to springtime airborne BC along the flight tracks of the three aircraft campaigns in Fig. 1. Contributions are quantified by excluding regional emissions. At all levels, anthropogenic emissions explain more than 90 % of BC concentrations, of which 56 % is contributed by

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eastern and southern Asia, followed by Europe with a contribution of 19 %. Biomass burning is minor (~8 %) compared to anthropogenic emissions in the contribution to springtime Arctic BC loadings, and the biomass burning impact on the springtime Arctic almost exclusively originates from regions other than North America. The relative contribution of anthropogenic emissions from each source region varies with altitude, partly reflecting different transport pathways and scavenging efficiencies. The influence of eastern and southern Asia increases considerably with altitude, with a contribution of 66 % between 400 - 700 hPa and 46 % between 900 - 1000 hPa, because transport from mid-latitudes follows isentropic surfaces that slope upward toward the middle or upper troposphere in the Arctic (Klonecki et al., 2003). In contrast, the influence of northern Asia decreases rapidly with altitude by a factor of 10 from the surface to 400 -700 hPa, reflecting transport from sufficiently cold regions along the low-level isentropic surfaces into the Arctic and direct transport within the polar dome (Klonecki et al., 2003; Stohl, 2006). The impact of Europe is roughly uniform throughout the troposphere, suggesting both of the above pathways are possible.

The gas flaring contribution to the springtime vertical BC concentration is shown as the red shading in Fig. 5. The contribution decreases with altitude from ~20 % near the surface to < 10 % above 800 hPa because flaring occurs almost exclusively below 2 km a.s.l (Stohl et al., 2013) and because the high-latitude sources of flaring limit isentropic lifting in the polar dome (Stohl, 2006).

Figure 7 (top right) shows the annual mean vertical contribution of anthropogenic emissions from each source region and of biomass burning to Arctic BC. Anthropogenic emissions from eastern and southern Asia (37 %) and biomass burning emissions (25 %) are major sources of Arctic tropospheric BC, along with a substantial contribution (43 %) from anthropogenic emissions in northern Asia near the surface (900 -1000 hPa). Unlike in spring, roughly half of biomass burning BC originates from North America in the annual attribution. Compared to springtime, the annual anthropogenic contribution from eastern and southern Asia is smaller and that from northern Asia is substantially larger in the lower troposphere. This reflects that long-range transport from eastern and southern

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Asia is more favorable in spring due to warm conveyor belts (Liu et al., 2015), and that proximal transport from northern Asia is more efficient in winter owing to the extended Arctic front to the south of northern Asian sources (Stohl, 2006).

The dominant role of eastern and southern Asia in the middle troposphere is consistent with Ikeda et al. (2017) who studied the source attribution of Arctic BC using a tagged tracer method in GEOS-Chem with the HTAP v2.2 emission inventory. The largest contribution from eastern and southern Asia to Arctic BC burden in this study is also consistent with Ma et al. (2013) and Wang et al. (2014). However, some prior studies suggested that Europe had the largest contribution to the Arctic BC burden (Stohl, 2006; Shindell et al., 2008; Huang et al., 2010b; Sharma et al., 2013). The difference likely arises from trends in anthropogenic emissions with reductions from Europe and increases in eastern and southern Asia as discussed further below.

Figure 8 shows the simulated source attribution of surface BC at Alert, Barrow and Ny-Ålesund. For all stations, anthropogenic emissions from northern Asia, eastern and southern Asia, and Europe are major contributors to high concentrations of BC in winter and early spring. In summer, anthropogenic contributions decline rapidly while biomass burning predominantly from North America becomes the primary source. At Alert and Barrow, the largest contributions are anthropogenic emissions from northern Asia in winter (~50 %), and from eastern and sourthern Asia in spring (~40 %). Barrow shows a pronounced peak in summer, more than 90 % of which is explained by biomass burning from North America. At Ny-Ålesund, anthropogenic emissions in Europe and northern Asia are significant sources of BC in winter and early spring with a contribution of ~30 % from each source.

The contributions from gas flaring to surface BC concentrations are shown as the red shadings in Fig. 3. Flaring accounts for ~25 % of concentrations in winter and spring and less than 5 % in summer at all stations except Ny-Ålesund where flaring contributes 14 % of BC in summer. This result is consistent with Stohl et al. (2013) who studied the flaring contribution to surface BC concentrations at Arctic stations using the FLEXPART model.

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We also investigate the influence of international shipping from the HTAP v2 inventory for 2010 on Arctic surface BC concentrations, and found the contribution is less than 1 % at all stations owing to the small magnitude of emissions (< 1 % of total anthropogenic BC emissions globally and within the Arctic). This source is expected to increase by 16 % by 2050 (Winther et al., 2014).

Our source attribution of Arctic surface BC has consistencies with that of Koch and Hansen (2005) who investigated the origins of Arctic BC using a general circulation model and found that Russia, Europe and south Asia each accounted for 20 % - 30 % of springtime surface BC. However, some studies (e.g. Stohl, 2006; Shindell et al., 2008; Gong et al., 2010; Sharma et al., 2013) suggested lower contributions (< 10 %) from eastern and southern Asia and higher contributions (> 30 %) from Europe than our results. The main difference is due to emission trends such that our anthropogenic BC emissions from eastern and southern Asia are generally 30 % higher than those in earlier studies (e.g. Shindell et al., 2008; Sharma et al., 2013) due to rapid development since 2000 and that our anthropogenic BC emissions in Europe are half those in prior studies due to European emission controls.

Figure 9 shows the contributions to Arctic BC column concentrations from changes in local emissions in 2011 as calculated with the GEOS-Chem adjoint. Pronounced seasonal variation and spatial heterogeneity are found. Sources in January are strongly influenced by specific Asian regions including western Siberia, eastern China and the Indo-Gangetic Plain, whereas sources in other seasons are more widespread across Europe and North America. Several hotspots are found in each season. In January, oilfields in western Siberia have a total impact of 13 % on Arctic BC loadings, of which 4.4 % is from the Timan-Pechora basin oilfield and 6.4 % from the West Siberia oilfields, suggesting that the influence of western Siberia is comparable to the total influence of continental Europe and North America (~ 6.5 % each in January). Considerable flaring emissions (67 % of total flaring emissions north of 60° N in January) and close proximity to the Arctic contribute to the substantial influence of these oilfields in western Siberia. The Indo-Gangetic Plain also exhibits considerable impact (7.2 %) to the Arctic in January,

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reflecting the substantial emissions there as shown in Fig. 1. In April, the influence of western Siberia decreases to 4.4 % with the northward retreat of the Arctic front. In contrast, contributions from emissions in eastern China (25 %) and North America (8.2 %) are enhanced owing to the facilitated transport of air masses from warm regions (e.g. the US and Asia) in spring (Klonecki et al., 2003). Emission contributions to Arctic BC loadings are generally weak in July, but the Tarim oilfield in western China stands out as the second most influential (3.2 %) grid cell to the Arctic, which is comparable to the influence of half of continental Europe (6 %). The Tarim oilfield is located in a high altitude (~1000 m) arid region (Taklamakan Desert). Considerable flaring emissions, less efficient wet scavenging and elevation all facilitate its large contribution to the Arctic. The contribution from North America is the largest (13 %) in July, consistent with the remarkably high BC loadings over high-latitude North America as shown in Fig. 6 (B3). Annually, eastern China (15 %), western Siberia (6.5 %) and the Indo-Gangetic Plain (6.3 %) have the largest impact on Arctic BC loadings, along with a noteworthy contribution from the Tarim oilfield (2.6 %). At continental scales, eastern and southern Asia contributes 40 % to the Arctic BC loadings. Northern Asia, North America and Europe each make a contribution of ~10 %, consistent with the vertical source attribution from sensitivity simulations in Fig. 7 (right). BC emissions within the Arctic generally contribute less than 3 % of Arctic BC loadings in all seasons except for January (5 %).

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4. Conclusions

Airborne measurements of BC concentrations taken across the Arctic during the NETCARE 2015, the PAMARCMiP 2009 and the PAMARCMiP 2011 campaigns, along with long-term ground-based measurements of BC concentrations from three Arctic stations (Alert, Barrow and Ny-Ålesund) were interpreted with the GEOS-Chem chemical transport model and its adjoint to quantify the sources of Arctic BC. Measurements from multiple BC instruments (rBC, EC, EBC) were examined to quantify Arctic BC concentrations. We relied on rBC and EC measurements, and on EBC inferred from PSAP absorption measurements with a MAC calibrated to rBC and EC measurements. The new rBC measurements at Alert differed by up to a factor of 2 from

commonly used measurements as discussed by Sharma et al. (2017) and played a major role in our ability to simulate observations at Alert. Our simulations with the addition of seasonally varying domestic heating and of gas flaring emissions were consistent with ground-based measurements of the BC concentrations at Alert and Barrow in winter and spring (rRMSE < 13 %), and represented airborne measurements of BC vertical profile across the Arctic (rRMSE = 17 %), yet underestimated an enhancement of BC concentrations between 500-700 hPa that was affected by several plumes near Alert, Barrow and Ny-Ålesund. The weaker biomass burning influences on the airborne measurements used here than in prior ARCTAS and ARCPAC campaigns facilitated our interpretation for anthropogenic source attribution.

Sensitivity simulations with the GEOS-Chem model were conducted to assess the contribution of geographic sources to Arctic BC. The Arctic tropospheric BC burden was predominantly affected by anthropogenic emissions from eastern and southern Asia (56 % in spring and 37 % annually from 1000 hPa to 400 hPa) with larger contributions aloft (66 % in spring and 57 % annually between 400-700 hPa) than near the surface (46% in spring and 20 % annually below 900 hPa), reflecting long-range transport in the middle troposphere. Anthropogenic emissions from northern Asia had considerable contributions in the lower troposphere (27 % in spring and 43 % annually below 900 hPa) due to low-level proximal transport. Biomass burning contributed 25 % to the annual BC burden.

Surface BC was largely influenced by anthropogenic emissions from northern Asia (> 50 %) in winter and eastern and southern Asia in spring (~ 40 %) at both Alert and Barrow, and from Europe (~ 30 %) and northern Asia (~ 30 %) at Ny-Ålesund in winter and early spring. Biomass burning, primarily from North America, was the most important contributor to surface BC at all stations in summer, especially at Barrow.

Our adjoint simulations indicated pronounced spatial and seasonal heterogeneity in the contribution of emissions to Arctic BC column concentrations. Eastern China (15 %) and western Siberia (6.5 %) had a noteworthy influence on Arctic BC loadings on an annual

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average. Emissions from as far south as the Indo-Gangetic Plain also had a considerable influence (6.3 %) on the Arctic annually. The Tarim oilfield stood out as the second most influential grid cell with an annual contribution of 2.6 %. Gas flaring emissions from oilfields in western Siberia had a striking impact (13 %) on the Arctic BC burden in January, which was comparable to the total impact of continental Europe and North America (6.5 % each in January).

The increasing BC fraction from eastern and southern Asia at higher altitudes could have significant implications for Arctic warming by extending the trend in increasing BC radiative forcing efficiency found by Breider et al. (2017) driven by strong increase with altitude of the direct radiative forcing of BC (Zarzycki and Bond, 2010; Samset and Myhre, 2015). Furthermore, anthropogenic emissions of BC in southern Asia are projected to increase under several IPCC scenarios (Streets et al., 2004; Bond et al., 2013). The climate implications of BC emissions within the Arctic are concerning given their disproportionate warming effects and the potential for increasing Arctic shipping activity as ice cover declines (Sand et al., 2013). The considerable impact of emissions from China and Indo-Gangetic Plain on the Arctic deserves further investigation. Additional work to reconcile the different BC mass concentrations measured by different instruments would be valuable to reduce uncertainties in BC studies not only in the Arctic but also globally.

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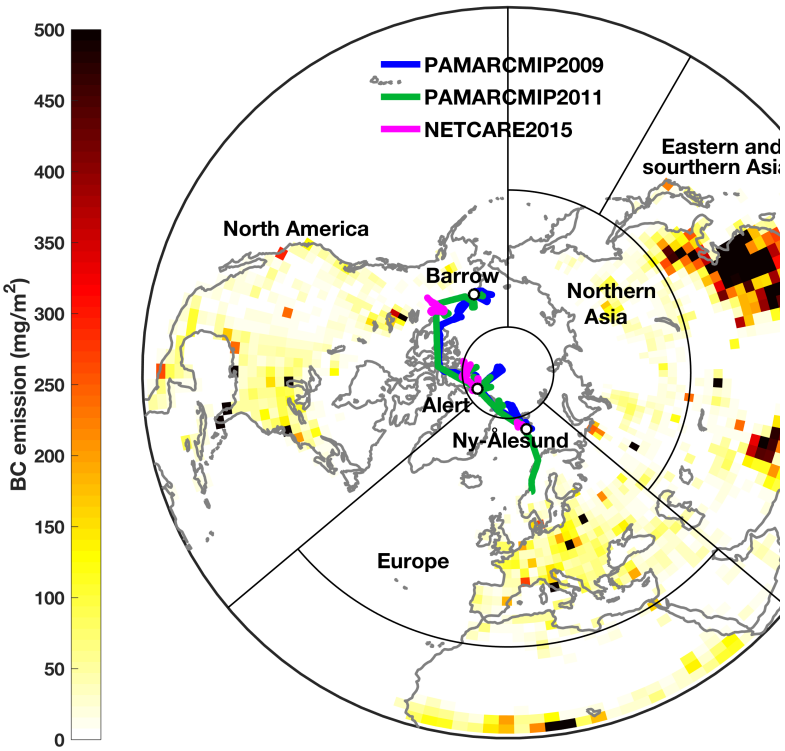

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Figure 1. The colormap indicates annual total BC emissions averaged over 2009, 2011 and 2015 as used in the GEOS-Chem simulation. Black open circles indicate the locations of ground monitoring stations (Alert, Barrow and Ny-Ålesund). Colored lines

indicate the flight tracks of the NETCARE 2015 (April 5th-21st), the PAMARCMiP 2009 (April 1st -25th) and the PAMARCMiP 2011 (Mar 30th – May 5th) campaigns. Black lines outline the source regions used in this study. Regional BC emissions are in Table 1.

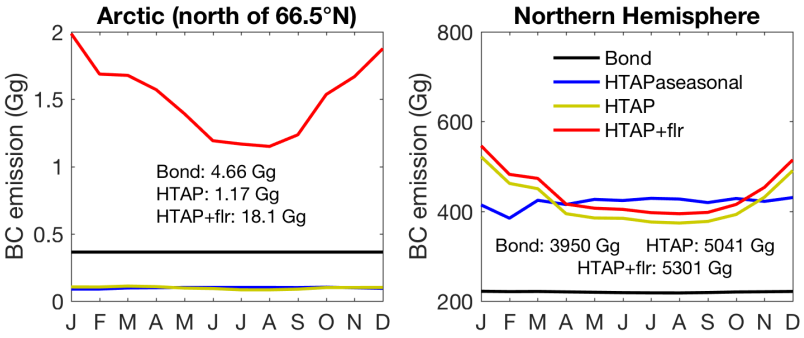

Figure 2. Anthropogenic BC emissions. Lines indicate monthly anthropogenic BC emissions from the Bond et al. (2007) non-seasonal inventory for 2000, the HTAP inventory for 2010, the HTAP inventory with non-seasonal emissions from residential heating, and the HTAP with additional flaring emissions for 2010. Annual values are given in the text.

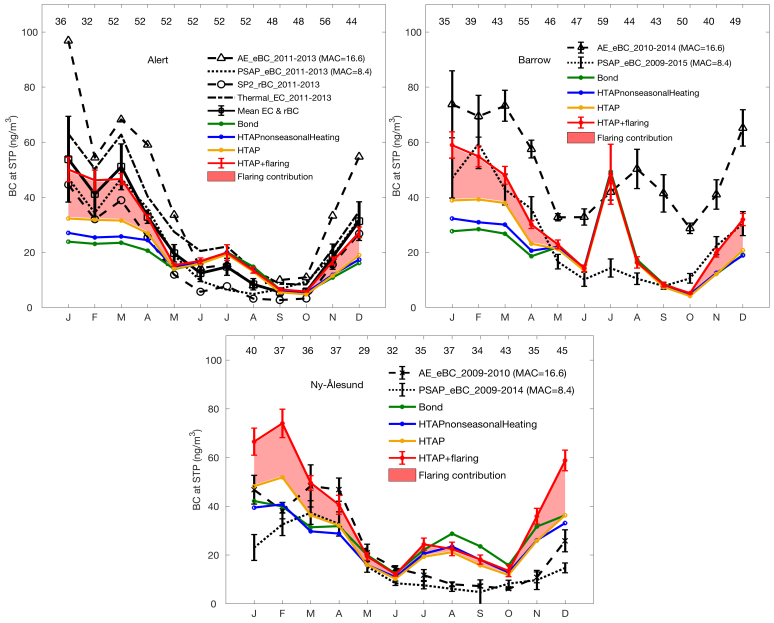

Figure 3. Seasonal variation of surface BC concentrations from measurements and simulations at selected Arctic stations. Black lines represent measurements from different instruments according to the legend. Error bars represent standard errors. The thick black line with squares at Alert is the average of rBC and EC concentrations. Error bars on the thick black line denote standard errors of monthly mean BC concentrations across instruments that are included in the calculation. Red shadings are the contributions from flaring to BC concentrations. Numbers below the top x-axis denote the total number of weekly observations from all available instruments in each month. Simulated monthly BC concentrations are the monthly averages of simulated concentrations for 2009, 2011 and 2015. Simulations use different emission inventories that are represented in color according to the legend. Error bars on the simulation represent standard errors. Concentrations from measurements and simulations are all calculated at standard temperature and pressure (STP).

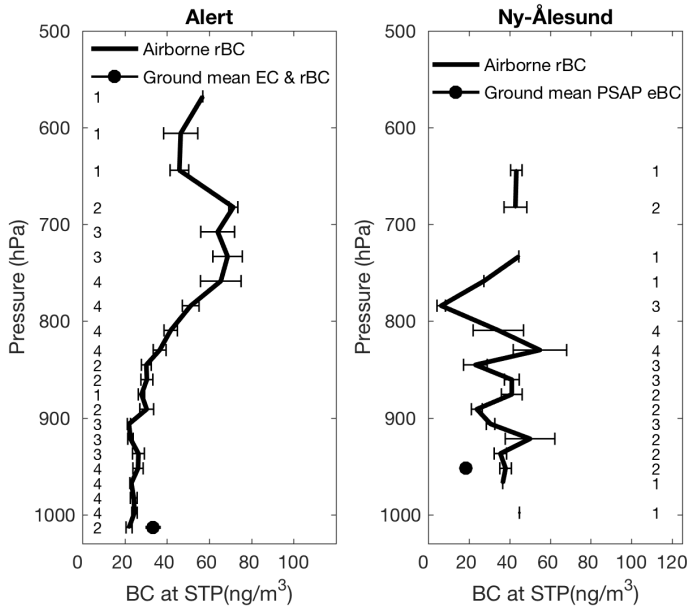

Figure 4. Vertical profile of BC concentrations averaged from all points along the flight tracks of the three aircraft campaigns (NETCARE 2015, the PAMARCMiP 2009 and the PAMARCMiP 2011) in Alert and Ny-Ålesund areas, along with the best estimate of April BC concentrations from ground-based measurements averaged for 2009 and 2011. The Alert area is defined as 59°W-65°W, 81.3°N-83.4°N and the Ny-Ålesund area is within 12°E-18°E, 77.8°N-79.1°N. Numbers along the y-axis are the number of airborne measurements in each pressure bin. All concentrations are presented at STP. Error bars on ground measurements are standard errors.

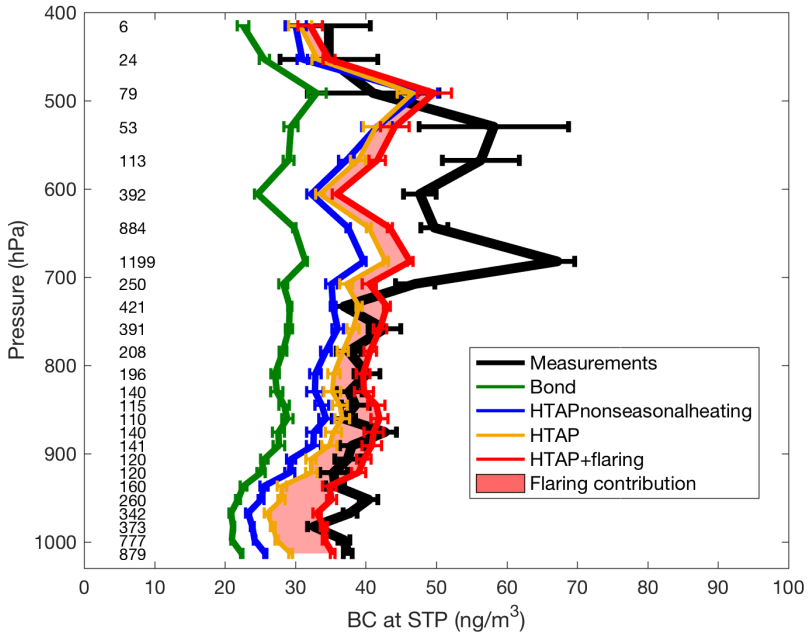

Figure 5. Mean spring vertical profiles of BC concentrations from measurements and simulations averaged over 50 hPa pressure bins from all points along the flight tracks of the NETCARE 2015, the PAMARCMiP 2009 and the PAMARCMiP 2011 campaigns. The red shading denotes the contribution of flaring to BC concentrations. Simulated vertical profiles of BC are coincidently sampled with airborne measurements for spring 2009, 2011 and 2015, and are averaged to the GEOS-Chem vertical resolution. Simulations include different emission inventories that are represented in different lines according to the legend. Error bars are standard errors. Numbers along the y-axis represent the number of measurements in each pressure bin. All concentrations are presented at STP.

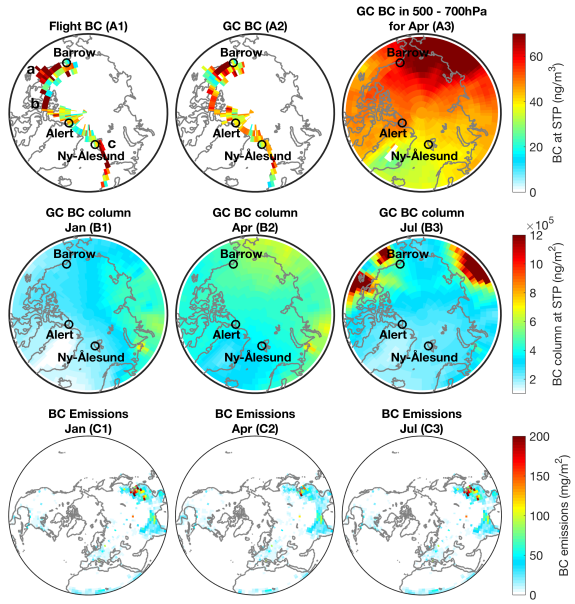

Figure 6. Top left: BC concentrations from the NETCARE 2015, PAMARCMiP 2009 and 2011 aircraft campaigns averaged on the GEOS-Chem grid, along with three hotspots labeled as a, b, c. Top middle: BC concentrations from GEOS-Chem simulations coincidently sampled with flight measurements. Top right: BC concentrations between 500-700 hPa simulated with the HTAP+flaring emissions in April averaged over 2009, 2011 and 2015. Circles are ground monitoring stations. Middle: pan-Arctic BC column concentrations simulated with the HTAP+flaring emissions for January (left), April (middle) and July (right) averaged over 2009, 2011 and 2015. All concentrations are at STP. Bottom: total BC emissions for January (left), April (middle) and July (right) averaged over 2009, 2011 and 2015. Letters and numbers in brackets refer to figure numbers.

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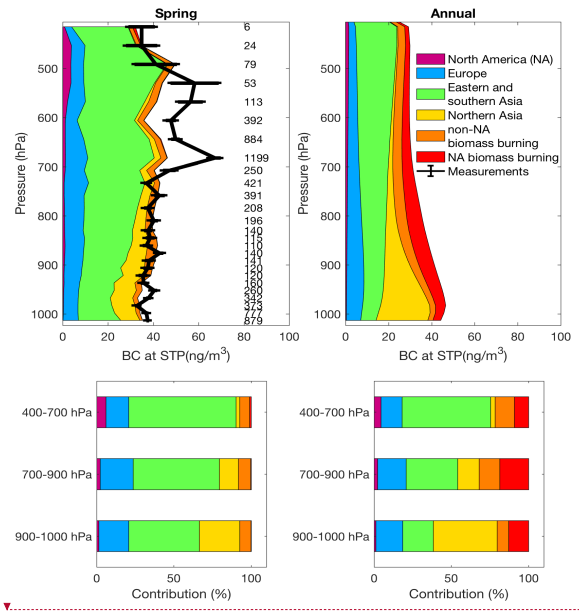

Figure 7. Top left: mean spring BC vertical profiles from flight measurements and simulations that are color-coded to anthropogenic sources from regions defined in Fig. 1. and biomass burning sources from North America and the rest of the world. Flight measurements and error bars are the same as in Fig. 5. Simulated vertical profiles of BC are taken coincidently with flight measurements. Numbers along the y-axis represent the number of measurements in each pressure bin. Top right: annual mean vertical profile of BC for the entire Arctic from simulations that are color-coded to source regions. Concentrations are all presented at STP. Bottom: regional contributions binned by pressure.

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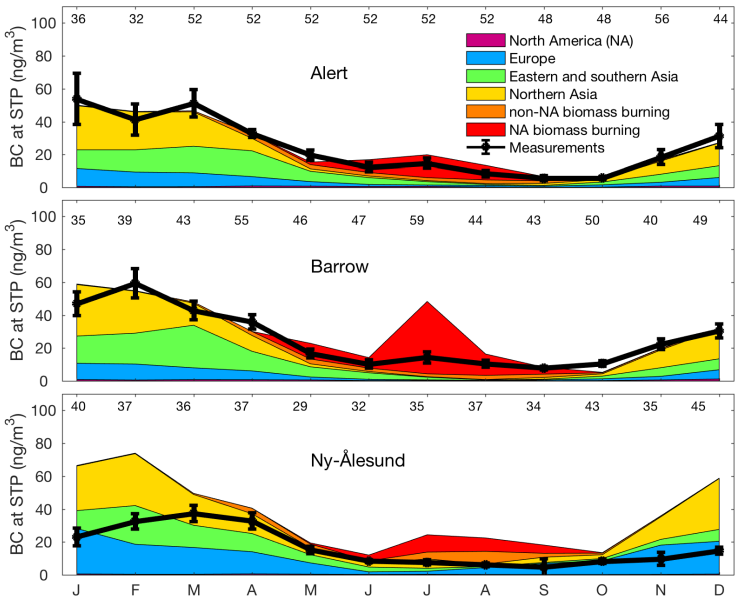

Figure 8. Monthly variation of BC surface concentrations at selected Arctic stations from measurements and simulations that are color-coded to anthropogenic sources from regions defined in Fig 1. and biomass burning sources from North America and the rest of the world. The measured monthly mean concentrations of BC and error bars are the same as the best estimate of surface BC concentrations in Fig. 3. Simulated monthly concentrations are monthly averages of 2009, 2011 and 2015. Numbers below the top x-axis denote the total number of weekly observations from all available instruments in each month. Concentrations are all presented at STP.

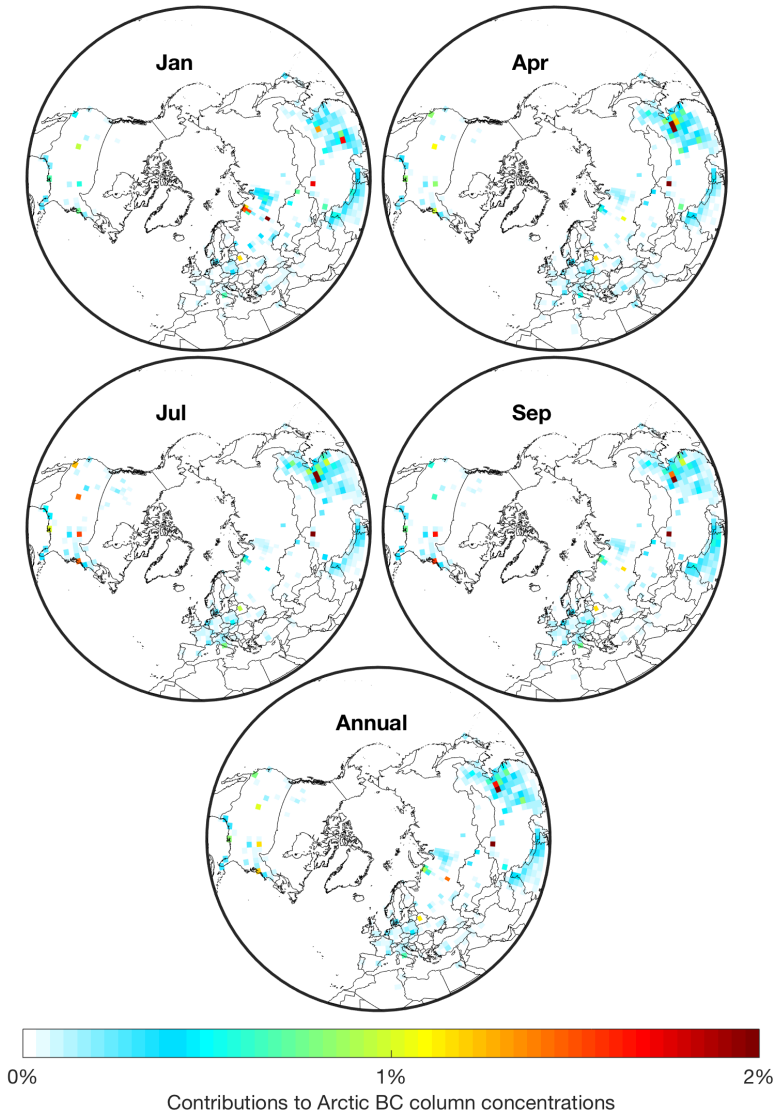

Figure 9. Contributions to Arctic BC column concentrations from changes in local emissions (as percent change in Arctic BC column concentration per fractional change in emissions) in 2011. Local emissions include anthropogenic and biomass burning

emissions. The annual map is the average of contributions in January, April, July and September calculated with the adjoint model.

Table 1. Regional annual BC emissions averaged over 2009, 2011 and 2015 as used in the GEOS-Chem simulations[a].

Emission Source ($Tg\ C\ yr^{-1}$)	North America	Europe	Northern Asia	Eastern and southern Asia
Anthropogenic[b]	0.62	0.48	0.11	3.36
Biomass burning	0.17	0.02	0.13	0.20

[a] Regions are outlined in Fig. 1.

[b] Including gas flaring, fossil fuel combustion and biofuel combustion.

Table 2. Summary of root mean square error (RMSE) and relative root mean square error (rRMSE) between simulations with different emissions and measurements for BC surface concentrations at Arctic stations (in reference to Fig. 3) and for vertical concentrations from airborne measurements (in reference to Fig. 5).

RMSE ($ng\ m^{-3}$; rRMSE)	Alert	Barrow	Ny-Ålesund	Vertical
Bond[a]	13 (55%)	17 (66%)	15 (88%)	17 (40%)
HTAPnonseasonalheating[b]	11 (48%)	16 (61%)	12 (71%)	11 (27%)
HTAPheating[c]	8.7 (37%)	13 (52%)	14 (82%)	9.4 (23%)
HTAPheatingflaring[d]	3.7 (16%)	11 (44%)	25 (150%)	7.2 (17%)

[a] Bond et al. (2007) emission inventory for 2000.

[b] HTAP v2 inventory for 2010 with non-seasonal residential heating

[c] HTAP v2 inventory for 2010 with seasonal residential heating

[d] HTAP v2 inventory for 2010 with seasonal residential heating and the addition of flaring emissions from the ECLIPSE v5 inventory

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. Early observations have identified anthropogenic emissions in northern Eurasia as the primary source of BC in the Arctic by analyzing the characteristics of chemical tracers (Lowenthal, Borys, & Mosher, 1997; Lowenthal & Kenneth, 1985). East and South Asia were excluded from these early studies because they were assumed to be unlikely sources due to the long distance (Cheng et al., 1993; Rahn, 1981). However, Koch and Hansen (2005) suggested that East and South Asia were comparable to Russia and Europe as sources to the Arctic surface BC, and were dominant sources of BC in the upper troposphere. Subsequent studies supported the importance of East and South Asia to the Arctic upper troposphere (Sharma et al., 2006; D. T. Shindell et al., 2008; Wang et al., 2011). Shindell et al. (2008) studied the sensitivity of Arctic BC concentrations to perturbations in emissions in each region using results from a coordinated model intercomparsion, and found that East and South Asia were indeed dominant sources in the Arctic upper troposphere, but at the surface, Europe remained the predominant contributor. Sharma et al. (2013) also found that East Asia had little influence at the surface but contributed substantially to atmospheric Arctic BC burden in winter.

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is a significant overlooked source of Arctic surface BC that remains missing from most inventories (e.g. Bond et al., 2007 inventory). Sand et al. (2016) found that Russian flaring emissions make the second largest contribution to the warming of Arctic surface temperature following Asian domestic emissions.

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In addition to anthropogenic emissions, another important periodic source of BC is biomass burning (Lavoué et al., 2000). For example, several simulations suggest that a severe air pollution episode in the European Arctic in 2006 spring (Stohl et al., 2007), and a strong increase in BC concentrations at four Arctic monitoring stations in summer 2004 are all attributable to intense biomass burning events in northern Eurasia and North America (Stohl et al., 2006). Subsequent studies support the large contributions of biomass burning to Arctic BC

concentrations (Evangeliou et al., 2016; Warneke et al., 2009; Yttri et al., 2014), yet disagree quantitatively. Warneke et al. (2009) suggested that biomass burning contributed at least 80% to the Arctic atmospheric BC burden in April 2008, whereas Wang et al. (2011) indicated that biomass burning contributed 50% of total BC in the Arctic tropospheric column during the same period. Evangeliou et al. (2016) found the contribution of biomass burning to Arctic surface BC as site-dependent, annually contributing 71% to surface BC at Alert, compared to 47% at Barrow. Additional interpretation of observations is needed to constrain this uncertain source.

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(Koch and Hansen, 2005; Sharma et al., 2013; Shindell et al., 2008; Wang et al., 2011).

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Since the latter has important implications for radiative forcing (Koch et al., 2009; B. H. Samset et al., 2014; Bjørn H. Samset & Myhre, 2011)

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All three campaigns were performed in spring when BC is most abundant, and traveled along similar routes across the entire western Arctic and near long-term ground monitoring stations in the Arctic (Alert, Barrow and Ny-Ålesund). Airborne measurements during all three campaigns were performed with the state-of-the-science single particle soot photometer (DMT-SP2; Stephens et al., 2003; Stone et al., 2010) for the measurement of refractory BC (rBC) mass concentrations. The spatial and multi-year coverage of airborne measurements during these campaigns offer comprehensive insight into BC distributions and high representativeness of characteristics of Arctic BC.

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that identifies possible source regions by tracking air mass flow

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that a perturbation is applied to emission sources and then compares to an unperturbed run to infer the influence of emissions on the simulation

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rBC concentrations were all measured with SP2 instruments (Droplet Measurement Technologies Inc., Boulder, CO) during the three campaigns.

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RMSE(ng m^{-3})	Alert	Barrow	Ny-Ålesund	Vertical
Bond	13	17	15	17
HTAPaseasonalheating	11	16	12	11
HTAPheating	8.7	13	14	9.4
HTAPheatingflaring	3.7	11	25	7.2