Response to comments of reviewer #1

This study examined the source contributions to Arctic black carbon (BC) in 2010 using the Flexpart Lagrangian transport model version 10.1 equipped with a new aerosol wet removal scheme. They found that Arctic BC at the surface and high altitudes is sensitive to emissions in high latitude and mid-latitude regions, respectively. About half of Arctic surface BC comes from anthropogenic emissions in Russia and 40% of BC at high altitudes comes from anthropogenic emissions in East Asia. The results can contribute to the Arctic aerosol community and the method with the new wet deposition scheme can improve high latitude aerosol simulation with lagrangian models.

Response:
We appreciate the reviewer for the positive evaluations. The comments are truly helpful in improving the work. Each of the comments is responded as follows.

Major comment:
It is good to add more literatures using models in the revised manuscript. However, I still feel the description lacks of details. The authors listed many literatures in lines 79-92, but they did not give their result quantitatively. They stated that 'Although numerous studies have been performed, results regarding regional contributions of BC sources in the Arctic are still inconclusive.' I was expecting to see the differences of BC source contributions between this study and other studies and the possible causes of the differences, but they only showed the similarity between the Flexpart Lagrangian transport model and GEOS-Chem in the last part of the paper. I agree that using the new wet removal scheme can improve the aerosol simulation, but it should not be listed as the new method since that the scheme was introduced in this model in Grythe et al. (2017). The authors need to highlight the unique findings in the manuscript.

Response:
Taking the comments, we added quantitative results about estimated major source regions/sectors of BC in the Arctic by previous studies along with model types, settings, meteorology, emission inventories, etc., placed them in Table 1, and revised the context accordingly.

Regarding the simulations of BC in the Arctic, the treatment of wet-scavenging parameterizations is a key factor affecting the model performance. The new wet removal scheme of Flexpart v10 introduced by Grythe et al. (2017) was a big improvement of the model in this context comparing to its previous versions. To apply the new version model to study BC in the Arctic, is the new point of the study. Comparing to Arctic BC studies using Flexpart v9, such as by Stohl et al. (2013) and Winiget et al. (2019), we found that the contribution of gas flaring to Arctic BC simulated by Flexpart v10 is 22% higher using the same emission inventory. We also used an updated emission inventory about gas flaring for Russia by Huang et al. (2015), which was reported to be closer to the real situation. We have revised the manuscript to make such a purpose clearer, as follows.

In L87-89, we revised as “The treatment of wet-scavenging parameterizations is a key factor affecting the model performance, which determines the uncertainties related to BC particle removal (Kipling et al., 2013; Schacht et al., 2019; Q. Wang et al., 2014).”

In L96-108, the difference of Flexpart versions was placed as “The FLEXible PARTicle dispersion model (Flexpart) had been used to investigate the transport pathways and source contributions of BC in the Arctic (Stohl et al., 1998, 2006, 2013). Of Flexpart model up to version 9, wet removal was treated considering below-cloud and within-cloud scavenging processes (Hertel et al., 1995; McMahon and Denison, 1979), which depends on cloud liquid water content, precipitation rate and the depth of the cloud. However, clouds were parameterized based on relative humidity, clouds frequently extended to the surface and at times no clouds could be found in grid cells, with unrealistic precipitation (Grythe et al., 2017). Recently, version 10 of Flexpart had been developed in which cloud is differentiated into liquid, solid, and mixed phase, the cloud distribution is more consistent with the precipitation data (Grythe et al., 2017). This improvement in the cloud distribution and phase leads to a more realistic distribution of below-cloud and in-cloud scavenging events. In this study, we quantified region-separated sources of BC in the Arctic in 2010 by using Flexpart v10.1.”

Regarding the difference of current findings with those by previous versions of Flepxart, we newly evaluated the sectorial contributions (open biomass burning, residential burning, gas flaring, and others) to Arctic BC in Figure 5. Our results indicated that residential combustions are important sources of Arctic surface BC (36% in October-March), supporting those by Stohl et al. (2013). Meanwhile, we estimated a higher value of gas flaring source BC (annually 17.5 ng m^{-3}, 36% of total) than those by Stohl et al. (2013). Such a difference could be attributed to the model treatment of BC removal as well as the inventory differences.

Such comparisons were added in L306-323, as “On an annual basis, anthropogenic sources and open biomass burning emissions accounted for 82 % and 18 %, respectively, of total Arctic surface BC. In which, gas flaring and residential burning (including burning of fossil fuels and biofuels) are accounting for 36 % (28–57 % in October–March) and 15 % (13–25 % in October–March), respectively (Fig. 5a-b). Our results support Stohl et al. (2013) that residential combustion emissions, especially in winter are important sources of Arctic BC (Table 1). We estimated a contribution of gas flaring to Arctic surface BC of 17.5 ng m^{-3} (36% of total). In comparison, the value was estimated as 11.8 ng m^{-3} using an average Arctic surface BC of 28 ng m^{-3} and a fraction from gas flaring of 42 % evaluated by earlier versions of Flexpart (Stohl et al., 2013; Winiger et al., 2019). The different contribution could be partly attributed to the difference in gas flaring emission inventory. BC emission from gas flaring in Russia by Huang et al. (2015) was used in the current study, where total BC emission from gas flaring in Russia in 2010 was ca. 81.1 kilotonne, which was larger than the estimate of ca. 64.9 kilotonne by GAINS inventory (Klimont et al., 2017) used by Stohl et al. (2013). Moreover, Adopting ECLIPSEv5 inventory as was used by Winiger et al. (2019), we estimated that gas flaring was contributing 14.4 ng m^{-3} to Arctic surface BC using Flexpart v10.1, a value 22 % higher than those obtained using Flexpart v9. This difference could be attributed to the improvement of the wet-scavenging scheme by Flexpart v10.1”.

Newly added Figure 5:

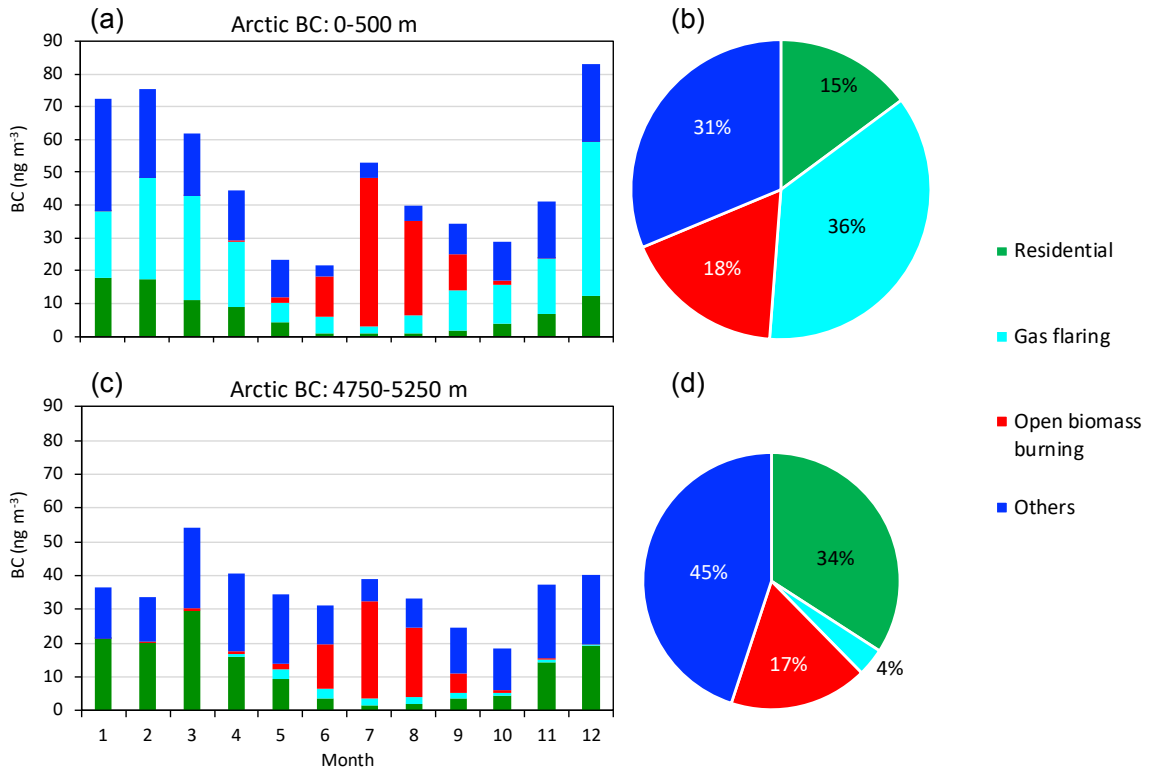

Figure 5. Sectorial contributions from residential combustion (including fossil fuel and biofuel combustions), gas flaring, open biomass burning and others (energy other than gas flaring, industry and transport) to (a) seasonal variations in Arctic surface BC, (b) annual mean Arctic surface BC, (c) seasonal variations in Arctic BC at high altitudes, and (d) annual mean of Arctic BC at high altitudes.

Specific comments:
Line 133: The Flexpart lagrangian transport model has some assumptions, like the hydrophilic BC. The uncertainties of the results related to the these assumption should be discussed.

Response:
Taking the comment, we added in the method section in L146-148 as “This may lead to an overestimation of BC removal and hence force underestimation of simulated BC concentration, especially of fossil fuel combustion sources where BC could be in the hydrophobic state for a few days.”, and in L227-230 as “The underestimation by Flexpart could also be partly contributed by the assumption that all particles are hydrophilic, where the BC scavenging could be overestimated. The corresponding uncertainties are larger in winter months, when there are more sources from fossil fuel combustion.”.

Lines 165-187: These two paragraphs should be in the ‘Materials and methods’ section.

Response:

Taking the comment, the two paragraphs is now placed in the 'Materials and methods' section in L185-208.

Line 253: The observations did not show 'a secondary peak in summer', as the authors presented in line 189 'Winter maxima and summer minima were observed'.

Response:
Taking the comment, the observed results are now described more accurately in L213-214 as "Winter maxima were observed for the four sites, while a secondary elevation was observed for Alert and Tiksi."

Descriptions on the simulated results were revised in L285-286 as "This seasonality agrees with observations and simulations at Alert, Tiksi, and Barrow if consider the unintentional data exclusion (Stohl et al., 2013),…".

Line 276-278: What caused the difference between this study (18%) and Winiget et al. (2019) (39%)? Can the different emissions (2010 vs 2011-2015) lead to the doubled contribution?

Response:
We appreciate for the comment. Winiger et al. (2019) included open field burning and residential biofuel burning as the total biomass burning. In the current study, residential burning of biofuels and fossil fuels, and open biomass burning altogether contributed 33% to Arctic surface BC. Such results indicated that the contribution of biomass burning (residential burning of biofuels and open burning) to Arctic surface BC in the current study is smaller than those by Winiger et al. (2019). Such a difference could be attributed the different treatment of BC removal by models (versions 9 and 10), as well as the different emission data.

These discussions were added in L324-333 as "A recent study based on isotope observations at the Arctic sites and Flexpart v9.2 simulation suggested that open biomass burning, including open field burning and residential biofuel burning, contributed 39 % of annual BC in 2011–2015 (Winiger et al., 2019) (Table 1). In comparison, we estimated that residential burning and open biomass burning together account for 33 % of total Arctic surface BC. As the residential burning in our study includes burning of both biofuels and fossil fuels, our results indicated that biomass burning has a relatively smaller contribution. Other than the differences in BC removal treatment between different versions of the model, the contribution difference could also be attributed to the different emission inventories and years (2010 versus 2011-2015)".