Response to comments by reviewer#2

This manuscript presents the source apportionment of black carbon at surface (0-500 m) and high altitudes (4750-5250 m) over the Arctic region using the latest version of Flexpart model. This study provides interesting data specifying the contributions of anthropogenic and biomass burning sources from different source regions to the surface and high altitude Arctic BC that significantly contribute to the Arctic aerosol community. However, discussion needs to be improved further before the MS appears in ACP.

Response:
We appreciate the reviewer for the positive evaluations on our work. Taking the comments, we added new analyses and revised discussions. Each of the comments is responded as follows.

Specific comments: Lines 24-25: "— with a focus on — 2010." It is not clear in the abstract and in the whole text as well that whether the two source categories (i) anthropogenic activities include only the fossil fuel combustion (or human made such as domestic and agricultural biomass burning as well) and (ii) biomass burning includes only the forest fires (or human made biomass burning as well)!

Response: We apologize for the unclear description of such an important point. In this study, (i) anthropogenic activities include not only fossil fuel combustion, but also human made domestic burning of biofuels; while (ii) the term of biomass burning was intended to denote open biomass burning in the field, includes forest fires, peat fires, open burnings of agricultural waste, grassland and savanna, woodland, deforestation and degradation. For clarity, we changed the term from "biomass burning" to "open biomass burning" throughout the text and added descriptions in the method section.

In the Abstract, the general introduction was revised in L24-26 as, "Geospatial sources of Arctic BC were quantified, with a focus on emissions from anthropogenic activities (including domestic biofuel burning) and open biomass burning (including agricultural burning in the open field) in 2010".

Descriptions about anthropogenic BC emissions were revised in L160-164 as "The Hemispheric Transport of Air Pollution version 2 inventory (HTAP2) for 2010 was used for monthly anthropogenic BC emissions (Janssens-Maenhout et al., 2015), which include sectors from energy, industry, residential and transport. It is worth noting that the residential sector includes not only combustions of fossil fuels, but also biofuels".

Descriptions about open biomass burning BC emissions were revised in L170-173 as "The term "open biomass burning" here indicates burning of biomass in the open field as is determined by the remote sensing measurement basis, including forest, agricultural waste, peat fires, grassland and savanna, woodland, deforestation and degradation, where biofuel burning for residential use is not included".

Lines 58-92: The review of literature of Arctic BC and its sources appears very brief. It is necessary to describe it fully by detailing the previous studies and thus addressing the necessity of the present study.

Response:
Taking the comments, we newly summarized current progress about Arctic BC source estimation in Table 1, where major BC source regions/sectors, model types, settings, meteorology, emission inventories, etc. were included. The description in the context was revised accordingly.

For the simulation of Arctic BC, the treatment of wet-scavenging parameterizations is a key factor affecting the model performance. With the new wet-scavenging scheme, Flexpart v10 cloud water treatment is much improved comparing with its previous versions. We therefore applied Flexpart v10 to investigate Arctic BC source, a first study of its kind as far as we know. We now revised the context to addressing the necessity of the study clearer.

In L79-81, we added "With these large disagreements among studies, it is thus necessary to unveil BC sources in the Arctic with high precision simulations".

In L87-89, we revised as "… (Table 1). 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, we 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.".

Lines 188~: "Flexpart generally reproduced the seasonal variations —-" Lines 200~: Flexpart v10.1 underestimated observed BC —-" I suggest the authors to compare the ground based and estimated concentrations, rather than just correlations, which are medium (r = 0.53-0.80) only, and statements, in order to make the extent of differences / uncertainties' clear.

Response:
Taking the comments, we added the comparisons of the concentrations, in L221-225 as "From January to May at Barrow and Alert, the mean BC simulated by Flexpart v10.1 were 32.2 ng m^{-3} and 31.2 ng m^{-3}, respectively. Which was 23 % and 46 % lower than the

observations (42.0 ng m^{-3} and 58.2 ng m^{-3}, respectively). This is probably related to the inadequate BC emission in the inventory, although seasonal variations in residential heating are included in HTAP2, which would reduce the simulation bias (Xu et al., 2017)".

In L231-232, we revised as "At Zeppelin, the Flexpart-simulated BC (39.1 ng m^{-3} for annual mean) was 85 % higher than the observed value (21.1 ng m^{-3} for annual mean), especially in winter (112% higher).".

In L237-242, we revised as "At Tiksi, Flexpart underestimated BC (74.4 ng m^{-3} for annual mean) in comparison with observation (104.2 ng m^{-3} for annual mean). Other than the hydrophilic BC assumption and underestimated BC emission in the simulation as the cases for Barrow and Alert, the observations at Tiksi by an aethalometer could containing light-absorbing particles other than BC, resulting in higher observed concentrations if compared with those obtained by SP2, EC and PSAP".

Lines 255~: "This seasonality —-" It is not at all clear that how good the results obtained in the present study are in agreement with the previous reports and how advanced /differed the source assessment of Arctic BC obtained from this study compared to the previous reports. For example, Stohl et al. 2013 reported that gas flaring and domestic biomass burning are the major sources of Arctic BC. This study also showed that the gas flaring is a major source, but the role of domestic biomass burning is not clear.

Response: Taking the comments, we newly evaluated the sectorial contributions to Arctic BC in Figure 5. We found that residential combustions are important sources of Arctic surface BC (36% in October-March), results being similar with 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 caused by the model treatment of BC removal as well as the inventory differences.

We added such comparisons 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:

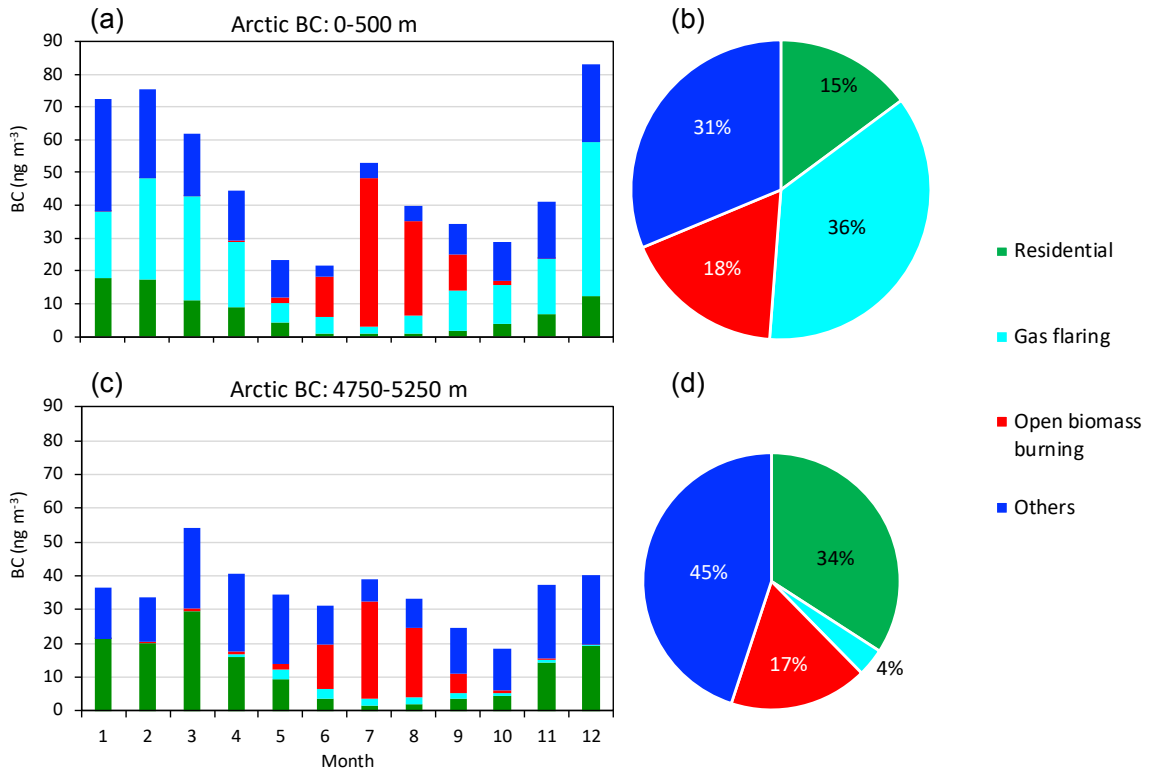

Figure 5. Sectorial contributions from residential combustion (including fossil fuel and biofuel combustions), gas flaring, open biomass burning and others (energy other than gas flaring, industry and transport) to (a) seasonal variations in Arctic surface BC, (b) annual mean Arctic surface BC, (c) seasonal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