

The authors would like to thank the Reviewer#2 for his careful review of our manuscript. We addressed each comment individually in the following electronic supplement, and have revised the manuscript accordingly.

General comments :

RC : This work investigates the transport processes of black carbon (BC) from Siberia to the Arctic and the effects of wet removal over mid- and high-latitudes during the ACCESS aircraft campaign using a regional scale chemical transport model (WRF-Chem) and a Lagrangian particle dispersion model (FLEXPART-WRF). The authors demonstrate that the BC emitted from Siberian fires were transported by low-pressure system and reached to the upper troposphere over the Arctic. They separately evaluate effects of large-scale and convective precipitation on wet removal and spatial distribution of BC. This study is interesting and scientifically important. The subject is of great interest to ACP. However, the interpretations of the transport mechanisms and analysis for wet removal of BC are not satisfactory, which are cause for concern (see Major comments). Another concern is that the manuscript is too long and the authors should attempt to shorten the manuscript (see Specific comments). Most other comments listed below are minor clarifications. Once these points are addressed satisfactory, the paper should in my opinion be suitable for publication in ACP.

AC : We thank the anonymous reviewer for his/her careful reading of the paper and for providing helpful comments, which help to improve the quality of the manuscript.

Major comments :

1. *RC : Section 4.1 and Figure 7a, horizontal poleward eddy heat flux It seems to me that the author's approach could not represent the activity of the migratory cyclones (WCBs). The authors define the horizontal poleward eddy heat flux ($\overline{v'\theta'}$), where the overbar denotes time-averaging over the ACCESS period (17 days?) and primes are instantaneous deviations from the means (17 days?). In general, a time scale of the migratory cyclones is about a week and it is shorter than the ACCESS period. Application of the long averaging period to the deviations (primes) may not detect the activity of migratory cyclones (WCBs) and anticyclones (i.e., the deviations from the 17-day means would not be adequate for the migratory cyclones). For example, the authors can use the 5-day running means for estimate the instantaneous deviations (v' and θ') instead of the 17-day means, and then calculate time average (overbar) for the ACCESS period.*

AC : In the original version of the manuscript, the time-averaging was indeed performed over the full ACCESS period (17 days) and the primes were instantaneous deviations from the 17-days means. The goal was to illustrate the persistence of the cyclones over northern Russia and Siberia where they are formed before reaching the Arctic region. We agree with the reviewer that the proposed approach is a better way to highlight the activity of migratory cyclones due to their shorter lifetimes. We therefore follow this advice and use the 5-day running means to assess the instantaneous deviations and then calculate time average for the ACCESS period (17 days). The result is shown on updated Fig. 7a. The main outflow regions remain exactly the same. The advantage of this method is that it underlines that values of the poleward eddy flux were larger over the Sakha (Yakutia) Republic region than in other outflow areas, illustrating the crucial role of this region in exporting fire plumes during summer.

2. *RC : Section 4.1 and Figure 7, upward BC flux and divergence of horizontal BC flux The authors state that "a strong ascent of BC mass fluxes are also co-located with areas presenting large values for the divergence of horizontal BC flux in the PBL" (P16L29-30). This would be misleading.*

According to Oshima et al. (2013), the horizontal convergence represents the upward BC transport from the PBL to the free troposphere, not the horizontal divergence. Considering air flow at the surface, the convergence can uplift air parcels from the surface to the free troposphere (like WCBs), but the opposite divergence cannot. For example, it seems to me that the upward BC region (red color around 90-100E, 50-60N in Fig. 7b) is slightly on the south of the divergence region (Fig. 7c) and corresponds to the light blue convergence region (Fig. 7c), although it is somewhat difficult to read the exact regions from these figures. The authors estimate upward BC mass flux at the 850-hPa level, but it seems to me that 850-hPa level is too low and 700-hPa is better. For example, Fig. 7b shows that there are large values over north China, but 850-hPa level over this region is close to the ground-level (largely influenced by BC mass concentration, rather than vertical velocity). In addition, because the authors define the PBL as the 700-1000 hPa layer for the horizontal BC flux, it is consistent to use the same 700-hPa level for the vertical BC flux to discuss the horizontal BC transport and the subsequent uplifting from the PBL to the free troposphere.

AC : The text was really misleading. We had written "PBL" instead of "FT", which was very confusing for the reader. We apologize for that. The strong ascent of BC mass fluxes are indeed co-located with areas presenting large values for the divergence of horizontal BC flux in the FT (Fig. 7d) and with areas presenting large values for the convergence of horizontal BC flux in the PBL (Fig. 7c). Convergence of BC flux in the lowest layers can uplift air parcels from the surface to the free troposphere, which are then exported to outflow regions (divergence of BC flux in the FT). We also agree with the reviewer that it is more convenient to plot the upward BC flux at 700 hPa (top of the so-called PBL) rather than at 850 hPa to avoid the contamination by the emissions. It has been done in the new version. We have also changed the color scale of Fig. 7c and 7d to better highlight the convergence areas in the PBL (blue). The result is much clearer with a very good spatial correlation between the regions with large upward BC flux, convergence (resp. divergence) of the BC horizontal flux in the PBL (resp. FT). The results remain unchanged.

3. *RC :* Section 5.3.1, transport efficiency of BC particles (TEBC) I could not understand the advantage of the method (P29L3-6) and why the authors use CO for estimate of TEBC by model. The TEBC values estimated by using modeled CO in Eq. (1) would include uncertainties in CO calculations in the model, as described in section 4.2. The authors state different transport patterns and diffusion during transport (between the BASE and NoWetAll simulations?), but the authors have already used the ratios of two model simulations in Figures 9 and 11 (e.g., NoDry/BASE). I am not sure why the authors do not define TEBC as $BC_{BASE}/BC_{NoWetAll}$, similar as Figures 9 and 11. The authors define background values of CO in Eq. (1) using CO measurements. I could not understand why the CO background values obtained over the ACCESS flight regions could be applied to the all model domain. The background values of CO would be different over the Arctic and East Asia. This would cause the uncertainty in estimate of TEBC. If the authors use modeled CO in Eq. (1), the use of model background CO values would be better. In my opinion, the estimation of TEBC using BC/CO ratios would be conducted by observation studies, because they could not estimate BC concentrations not influenced by wet removal (e.g., such as $BC_{NoWetAll}$) from the observation. The use of BC/CO ratios for TEBC estimation in Eq. (1) assumes BC-CO correlation over the source regions (similar emission sources for BC and CO). However, the anthropogenic plumes in Figure 8 show some enhancements of BC but little enhancements of CO (no BC-CO correlation?). I am not sure that Eq. (1) could be applied to these plumes, although the CO values would be canceled out in numerator and denominator in Eq.(1).

AC : As noted by the reviewer, the estimation of TEBC using BC/CO ratios has also

been conducted by a few studies in other regions. Using this proxy that has already been used was clearly a motivation to propose a comparison of values obtained in this study (we compared our results to Matsui's values for example). Notwithstanding, we understand that this proxy has been defined for observation studies, not modelling ones. We have therefore preferred in the new manuscript the definition of TEBC defined by Oshima et al. (2013), which enables to get rid of errors linked to CO uncertainties. Those errors were however limited by the fact that CO values in Eq. (1) were almost canceled out in numerator and denominator. The background CO is not necessary anylonger. Figures 13 and 14 have been modified accordingly. The results are identical to our previous results, except when APT values are very small : TEBC were not equal to 100%, but close to 95%, in the original manuscript due to errors ascribed to CO uncertainties. This artefact has vanished in the updated paper.

4. *RC : Sections 5.3.1 and 5.3.2, APT and ACWT calculations and interpretations It seems to me that the author's APT and ACWT approaches include below-cloud scavenging and in-cloud scavenging (nucleation scavenging with subsequent removal by precipitation, rainout) processes and could not distinguish these two processes, although these approaches could indicate the importance of precipitation on BC removal. It is expected that the both approaches would give the similar results, because the sum of the rain, ice, snow and graupel precipitation rates (used for APT) and the sum of cloud liquid water, ice, snow, rain and graupel contents (used for ACWT) would correlate. The authors should state that the similar results (Fig. 13) obtained from two different approaches suggest the validity of the importance of precipitation on BC removal, rather than effects of nucleation scavenging (P30L3).*

AC : Both APT and ACWT approaches include below-cloud and in-cloud scavenging. The difference between the two approaches refers to the nucleation scavenging process. To compute APT and ACWT, precipitation rates for the different hydrometeors are summed *only* in WRF-Chem grid boxes crossed by the FLEXPART-WRF trajectories. If activation occurs, particles are transferred from interstitial aerosol phase to cloud-borne aerosol phase (nucleation scavenging). As far as APT is concerned, this is not a deposition process if cloud droplets do not grow enough to produce rain drops that precipitate. But it can be considered as a deposition process for the plume if the droplets containing aerosols reach the sizes of precipitating rain drops in other cells (for example above or below the considered grid box crossed by the trajectory). In that case, the sum of cloud liquid water and ice crystals mixing ratios in such grid boxes is relevant. This is the purpose of ACWT. The sum of the rain, ice, snow and graupel precipitation rates (used for APT) and the sum of cloud liquid water, ice, snow, rain and graupel contents (used for ACWT) do not correlate. This correlation strongly depends on the size distribution of hydrometeors. Nevertheless, we agree that the two approaches almost give similar results, except for the flaring plumes, as indicated in the paper.

Specific comments :

- *RC : P1L13, P13L6, P16L2, P18L13, P18L24, P31L20, P31L21, P33L4, P33L23, Remove "very".*

AC : Done.

- *RC : P3L26, "ACCESS campaign", Please spell out it and add a brief description about the campaign here.*

AC : ACCESS stands for Arctic Climate Change, Economy and Society. This has been included in the new version in addition to the location and time of the airborne campaign.

- *RC : P7L1, "size bins (8 in this study)", Please show the minimum and maximum size ranges.*

In MOSAIC, aerosol size distribution is between 39 nm and 10 μ m.

- *RC : P7L18, Please show the horizontal and time resolutions of the fire inventory. Section 2.2.2, There is no description about the flaring emissions. Please add a brief description.*

AC : FINN provides daily, 1 km resolution emissions. The description of flaring emissions was indeed missing : flaring emissions are from the ECLIPSE (Evaluating the CLimate and Air Quality ImPacts of Short-livEd Pollutants) inventory (Stohl et al., 2015).

- *RC : P8L19-20, "using the meteorological fields from the WRF-Chem simulation." Is it BASE calculation ?*

AC : yes, it is the BASE run. It has been specified.

- *RC : Figures 2 and 3, It is difficult to find the vertical bars (median values). Please make the median values more visible. Please clarify in the figure captions that all flight data are used in these figures.*

AC : To represent the median values, the vertical bars have been replaced by diamonds of the same color as the mean values. The captions of Fig. 2 and Fig. 3 mention that data have been interpolated along all the 14 Falcon flight tracks.

- *RC : P10L19-20, "wrong OH and transport", This is not clear what you mean. Do you mean that OH calculation in the CBM-Z scheme has a problem for 6-9 km range but it is OK for other altitude range ? I cannot understand this. Please clarify.*

AC : This sentence does not refer to the slight underestimation of the CO in this study by WRF-Chem model via CBM-Z scheme. It was a general sentence explaining the general underestimation of CTM in the Arctic. We have re-written it to be clearer : The small underestimation in CO between 6 and 9 km is a common feature observed by most models (Emmons et al., 2015 ; AMAP, 2015). Variability in models, run with the same emissions, appears to be driven by differences in chemical schemes influencing modelled OH and/or differences in modelled vertical export efficiency of CO from mid-latitude source regions to the Arctic (Monks et al., 2015).

- *RC : P11L19-20, The authors show the overestimation of BC in the mid-troposphere for the Run100 simulation in Figure 3. Please specify the altitude ranges of the overestimation.*

AC : The overestimation is between 1.5 and 5 km.

- *RC : P11L21, " This suggests that, at a coarser resolution, the model is unable to resolve the fine structure of plumes transported in altitude", If the authors state this, it is better to check the CO concentrations. Is there overestimation of CO in the mid-troposphere for the Run100*

simulation?

AC : Yes, we have noted a corresponding overestimation of CO in the mid-troposphere in the Run100 simulation. There is an enhancement of 4 to 5 ppbv as compared to the BASE run.

- *RC : P12L5, "Global models always overestimated BC mass", I do not think "always". Some models underestimate BC mass concentrations at upper troposphere during the ARCTAS campaign (Koch et al., 2009).*

AC : We replaced "always" by "generally".

- *RC : P13L12, The authors state that the AOD underestimation is due to the simplified SOA calculation. If so, please explain this in more detail.*

AC : We don't know the exact reason of the model underestimation in this region. AOD is computed through a Mie code embedded in the model. The representation of the size distribution and complex refractive index also strongly influences the result. The simplified SOA mechanism is a potential cause, but we can't say it is the main one. According to a comment of Reviewer#1, this sentence has been removed.

- *RC : P16L6-11, "The major objectives of this section ... towards the Arctic.", The authors need not to describe objectives in each section. Please remove this paragraph to shorten the manuscript.*

AC : It has been removed, which helps to shorten a bit the paper.

- *RC : P16L15, " the overbar denotes time-averaging over the ACCESS period", Please specify the time-averaging period (i.e., the ACCESS period). From 4 July to 21 July in 2012 (17 days) ?*

AC : The information about the time-averaging has been added.

- *RC : P18L11, "Andøya and Spitsbergen", It is better to write the longitude and latitude of these locations.*

AC : We have specified the coordinates of Andøya (69.1°N, 15.7°E) and Spitsbergen (78.9°N, 18.0°E).

- *RC : P18L19-20, "This is mostly due to numerical diffusion in the model", Please explain the numerical diffusion in more detail.*

AC : Numerical diffusion is caused by the finite difference method applied to the advection equation on the model grid. In the model, the gradients are simply taken along coordinate surfaces, hence are imperfectly described. A 6th-order numerical diffusion is used in our simulations.

- *RC : P18L33 and Figure 9, "relative contributions", BC or CO ? Please clarify in text and figure captions.*

AC : This had been forgotten. Fig. 9 shows the relative contributions of BC concentrations due to the different emission sources.

- *RC : Section 4.4, "four plumes (boreal fires), two plumes (anthropogenic), two plumes (flaring)". It is better to mark (e.g., by circles) these plumes in some of Figure 8. It seems to me that some plumes were observed by the aircraft, but others (e.g., flaring) were not observed (not along the flight tracks). Please clarify these differences in text, because the FLEXPART calculations were conducted for these eight plumes and the reliability of interpretation will be different whether the plumes were observed ones or not (only modeled ones).*

AC : The eight airmasses selected for further analysis have been marked by magenta circles on Fig. 8. We have given them a name for further discussion, distinguishing biomass burning plumes (BB1, BB2, BB3, BB4), anthropogenic airmasses (An1, An2) and airmasses from flaring sources (F11, F12). We also clarified that only the fire plumes were observed by the aircraft. The anthropogenic and flaring ones have been detected on the model cross-sections.

- *RC : Section 4.4, BC and CO concentrations in the anthropogenic and flaring plumes, It seems to me that some enhancement of BC with little enhancement of CO was observed in the anthropogenic plumes. Could you explain this difference? Figures 8 and 9 also show that both BC and CO concentrations were low in the flaring plumes. Could you explain why CO concentrations were low for the anthropogenic and flaring plumes?*

AC : The color scales chosen in Fig. 8 might have given the impression that a small enhancement of BF was correlated with little enhancement of CO in flaring and anthropogenic plumes. This is actually not the case. The enhancements in BC and CO are on the same amount of magnitude (in %). If the decrease of BC in anthropogenic and flaring plumes was only due to precipitation, the CO should have remained unchanged. This is not the case, suggesting that transport of plumes from anthropogenic and flaring sources is not only directed towards the northern coast of Norway.

- *RC : P23L2, "(Fig 7)"?*

AC : This was a misprint. Thanks for noticing it.

- *RC : Figure 10d, The authors state that "Heating of large Siberian fires can inject CO and BC into the free troposphere (P21L9)", but Figure 10d shows that high BC concentrations initially appear at 0-2 km in altitude. Please clarify the injection height of the fire emission in text.*

AC : This was a mistake and has also been detected by Reviewer#1. The sentence has been removed. The rapid uplift to 6 km is due to WCBs over eastern Siberia.

- *RC : P24L8-9 "we use the normalized differences between the NoDry, NoWet, NoWetCu simulations and the BASE run." It is better to express the calculation method to estimate the relative contributions, specifically.*

AC : We have clarified this by giving the formula used for those calculations : $100 \times \left(\frac{NoX - BASE}{NoX} \right)$, where NoX represents the NoDry, NoWet or NoWetCu simulation.

- *RC : P24L20, "European influence", European anthropogenic influence?*

AC : Corrected.

- *RC : P24L28-29, "An understanding of the wet removal of BC ... to the Arctic.", This means a general importance of wet removal. Please remove this sentence to shorten the manuscript.*

AC : This has been removed.

- *RC : P24L31-P26L4 and P33L16-18, The authors state that BC particles were coated with sufficient water-soluble compounds, but they have not shown the coating information for the observed plumes. If the coating information will be available from the SP2 measurements, this information may be helpful for the interpretation, although a portion of thickly-coated BC particles in the observed plumes had been removed by precipitation during transport from the source regions to the Arctic.*

AC : This information would have been indeed very useful. Unfortunately, the coating thickness was not available from the SP2 measurements.

- *RC : P26L22, "map of the upward BC flux and the patterns of the divergence of horizontal BC flux in PBL (Fig.7)." Please see Major comments and remove "the upward BC flux and".*

AC : Figure 7 has been updated according to the answer to major comments. The term "PBL" has been replaced by "FT".

- *RC : P27L2-3, " BC particles are removed through the nucleation scavenging mechanism or through below-cloud scavenging", Nucleation scavenging of aerosols alone is not a deposition process, because if only nucleation scavenging takes place (aerosols become cloud droplets) and subsequent cloud evaporation takes place, the aerosols would remain there. Or do you mean in-cloud scavenging (rainout) ?*

AC : We never say that the nucleation scavenging mechanism was a deposition process, which would suppose that aerosols are removed from the atmosphere by this process. However we do not consider losses for the *atmosphere* here but rather losses for the *plumes*. When there is activation of clouds droplets, BC is not lost for the atmosphere but is transferred from interstitial aerosol to cloud-borne aerosol (nucleation scavenging). If the cloud droplets reach the sizes of precipitating rain drops, it will act as a deposition process from the plume.

- *RC : P27L18-19, "the below-cloud removal efficiencies are indeed very small." If the authors did not estimate the effects of below-cloud scavenging by the model, please add some references here. Below-cloud scavenging depends on size distributions of particles and precipitation intensity. Is intensity of convective precipitation small ?*

AC : These lines refer to below-cloud scavenging in parameterized cumulus clouds only. The model takes into account below-cloud scavenging in grid-resolved clouds. The intensity of convective precipitation can be seen in Fig. 5 combining the total precipitation (5b) and the fraction of convective precipitation (5c). The efficiency of below-cloud scavenging depends on the ratio of the sizes of particles and rain drops. This ratio is very small here as mostly all BC-containing particles are in the fine and accumulation modes (80 – 470 nm).

- *RC : P28L14, background values of BC mass mixing ratios "and CO concentrations", respectively.*

AC : This has been added. Thank you for pointing this.

- *RC : Figure 13, Please mark or emphasis the starting points (release time = 0) of the eight plumes, if possible.*

AC : We have added magenta circles in Fig. 13. They emphasis the starting points of the eright airmasses discussed in Sect. 4.4.

- *RC : P30L2-3, "as a function of ACWT, suggesting that BC can also be removed efficiently by nucleation scavenging when transported to the Arctic." Please see Major comments. I could not understand why this result indicates the BC removal by nucleation scavenging. Please explain interpretations of the ACWT results more clearly.*

AC : Please see our response to major comments. When there is some activation of BC-containing particles during the transport of a plume, there is a transfer of interstitial particles to cloud droplets. This suggests a loss of aerosol in the advected plume (but not from the atmosphere if subsequent cloud evaporation takes place). The difference between ACWT and APT is the sum of the integrated mixing ratios of cloud liquid water and ice crystals in clouds. The fact that the relation between TE_{BC} and ACWT is slightly different that the one between TE_{BC} and APT, especially in flaring plumes, indicates a role of cloud liquid water and ice crystals in clouds to remove BC during transport. This is caused by the nucleation scavenging mechanism. This has been explained more clearly in the text.

- *RC : P30L29 and Figure 14, "mean BC mass concentrations zonally averaged during the ACCESS period", The model domain shows that the longitude range used for the zonal averages is different depending on latitude, for example, all longitude range at high latitude but only Asian longitude range at mid latitude. This may contribute to the contrast between the mid-latitude and the Arctic. If so, please add descriptions about the possible effects for the zonal averages due to the different longitude ranges in text.*

AC : The reviewer is right but it has been done on purpose. Figure 9 had shown that the sum the relative contributions of anthropogenic, flaring and fire emissions to the total BC in the Arctic was higher than 98%. This confirms that the influence of the model boundary conditions on Arctic BC at this period is insignificant. The goal of Fig. 14 is to show the relation between BC mixing ratio and TE_{BC} at different latitudes for plumes that have been transported to the Arctic during the ACCESS period. Our study shows that our domain is appropriate to include all sources influencing the Arctic region in July 2012. Some details have been added to the text.

- *RC : P31L4, "illustrating the sharp meridional gradient in the distribution of moisture and precipitation", The authors have not shown any results or discussions of moisture. It seems to me that this description will be misleading, because effects of wet removal will be greater where moisture and precipitation are greater (e.g., Asian regions), but TE_{BC} is not smaller over these regions. The TE_{BC} will be smaller for air experiencing wet removal over Asia and that subsequently transported to the outflow regions (high latitude). Please clarify the interpretation.*

AC : We agree with the reviewer, this sentence was confusing. It has been replaced by "This sharp meridional gradient is due to the fact that TE_{BC} is indeed smaller for air experiencing wet removal over Asia or Siberia and that is subsequently transported to the outflow regions (high latitudes)."

RC : P31L19, "The interactions between aerosols and clouds", Please clarify what this means.

AC : "The interactions between aerosols and clouds" has been replaced by "Aerosol removal in cumulus clouds".

RC : P33L5-6, "the spatial distribution of the mean upward BC mass fluxes", Please see Major comments and this should be removed.

AC : Please see our answer to major comments. There was a mistake here : this is a good spatial correlation between AOD, the strong divergence regions of the BC horizontal fluxes in the FT (not PBL as written before) and the spatial distribution of the mean upward BC mass fluxes.