

**Review of "Cross-polar transport and scavenging of Siberian aerosols containing black carbon during the 2012 ACCESS summer campaign" by J.-C. Raut et al.**

**General comments:**

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.

**Major comments:**

1. 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  $\theta'$ ) instead of the 17-day means, and then calculate time average (overbar) for the ACCESS period.

## 2. 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.

## 3. Section 5.3.1, transport efficiency of BC particles ( $TE_{BC}$ )

I could not understand the advantage of the method (P29L3-6) and why the authors use CO for estimate of  $TE_{BC}$  by model. The  $TE_{BC}$  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  $TE_{BC}$  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  $TE_{BC}$ . 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  $TE_{BC}$  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  $TE_{BC}$  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).

#### 4. 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).

#### **Specific comments:**

P1L13, P13L6, P16L2, P18L13, P18L24, P31L20, P31L21, P33L4, P33L23, Remove "very".

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

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

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.

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

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.

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.

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.

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?

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).

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

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.

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)?

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

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

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

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).

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?

P23L2, "(Fig 7)"?

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.

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.

P24L20, "European influence", European anthropogenic influence?

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.

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.

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".

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)?

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?

P28L14, background values of BC mass mixing ratios "and CO concentrations", respectively.

Figure 13, Please mark or emphasis the starting points (release time = 0) of the eight

plumes, if possible.

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.

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.

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.

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

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