Dear Federico Fierli,

Thank you for your efforts to help with the editorial process with our manuscript. We have organized our response as follows. First, we include the responses to the referees uploaded on ACPD as part of the interactive discussion. Then, we include a track changes version of the manuscript with all removed text in red and new text in blue. The goal was to answer each reviewer comment and integrate the related changes into the revised manuscript (attached). We note that there are some small differences in wording in the revised manuscript compared to the online responses, which have been modified primarily for readability.

Thanks in advance for your continued work as editor of this manuscript and I look forward to hearing from you in the coming weeks.

Best regards,

Jean-Christophe Raut

The authors would like to thank the Reviewer#1 for his careful review of our manuscript. We addressed each comment individually in the following electronic supplement, and have revised the manuscript accordingly.

RC : This paper presents a thorough and detailed model investigation of the factors affecting transport of several plumes of BC and CO to the Arctic, comparing the influences of different pollution sources and the spatial distribution, amount and types of precipitation. There is a limited comparison to aircraft measurements that shows the model is doing a reasonable job of capturing the general features of the observations, though not doing a perfect job of simulating the exact concentrations or resolving the finer structure of the plumes (and I would not expect it to). Overall I think the work is of a very high scientific quality, and the main issue I have is that it is too long. However, I think it is probably not suitable to be split into 2 papers, nor are there any sections that could easily be cut without detracting from the rigour of the analysis, so I think it will just have to remain long. Possibly, parts of Section 2 might work as an appendix/supplement. Additionally, some of the written English is phrased in strange ways and some sentences do not quite make sense, so it could do with some work. I recommend publication in ACP subject to the following minor revisions.

AC : We thank the anonymous reviewer for providing helpful comments. As mentioned by the reviewer himself/herself, it is not suitable to be split into 2 papers as it would compromise the second paper. We have tried to reduce parts of the paper, as suggested by Reviewer#2, but the length is still important.

— *RC : P1L13-15 The way this is phrased makes it sound like the source determines the APT. I think this is because you have said "is" rather than "was", which makes it sound more like a general statement rather than a specific statement about the plumes studied in this paper. This type of error occurs throughout the paper and is mostly benign but can sometimes be confusing, such as in the example above. As a general rule of thumb, the work you have done is in the past (e.g. "the campaign took place" or "we ran the model") but things you do in the paper itself should be in the present (e.g. "in this study we describe. . . " or "Figure 5 shows . . . "). It gets more tricky, for example when you come to the conclusions- the specific plumes you studied were affected by precipitation but plumes in general are affected by precipitation.*

AC : We agree with the reviewer that using present may give the impression of a general comment and may be confusing for the reader. We put the phrase in the past tense. More generally, we have decided to apply the reviewer's suggestion along the full manuscript. We use present when it refers to the work we have performed and the results obtained, and we use the past when statements refer to conclusions.

— *RC : P3L8-9 "Schwarz et al showed. . . ." I'm not sure what you are trying to say with this sentence, other than to mention the study by Schwarz et al. It seems like you do a better job of saying the same thing in the next sentence, so this one isn't really necessary*

AC : We removed that sentence.

— *RC : P3L15 What results from Koch and Hansen ? Do they confirm them or do they just agree with them ? (i.e. both could be wrong)*

AC : Koch and Hansen (2005) highlighted an overestimation of BC concentrations in the upper Arctic troposphere. The modeling study of Breider et al. (2014) found similar results. We modify the word "confirms" by "agrees with".

— *RC : P5L6-10 Please give details of how the SP2 was calibrated. Actually, you should also say how the CO box was calibrated. Just one sentence for each would probably be sufficient if a standard method was used.*

AC : The SP2 was calibrated using the recommended calibration material (Fullerene soot, as produced, Lot#F12SO11, Alfa Aesar Inc., Ward Hill, MA). CO was measured every second from VUV fluorescence with an accuracy of 10% (Gerbig et al., 1999).

— *RC : P5L14-15 "Absolute uncertainty of BC particle mass is within 10%, the uncertainty of the derived total BC mass mixing ratio is about 30%." I am not sure I follow the logic here. Also 10% is not an absolute uncertainty, it is a relative uncertainty- a percentage is relative by definition. Given you do not mention particle size in your analysis, the only relevant errors are A) The systematic uncertainty in your BC calibration and B) The statistical uncertainty in the derived BC mass concentrations. A) is down to a combination of the sampling time, concentrations and flowrate, but is easy as you can just pick a time when you think your concentrations are constant and look at the variation in your time series. You could express this as a relative error (e.g. +/- 10%) or absolute (e.g. ± 2 ng/kg) B) is more difficult as you have two factors- firstly the random variation in your calibration slope (in other words how accurately does your particular slope recreate the mass of the calibration material, if you repeated your calibration exactly how much would the slopes differ ?) (see Laborde et al. (2012b)) and secondly how well does your calibration material represent the instrument response to the BC you measure in the atmosphere ? As Laborde et al. (2012a) showed, the SP2 responds differently to different BC types. I understand the observations present a fairly minor part of your paper but if you are going to present them and quote an uncertainty it should be done correctly, which at present I don't think it is.*

AC : This was indeed a mistake. The instrument provides acumulation model refractory BC mass mixing ratio with a total relative uncertainty of 30% **(Laborde et al., 2012, Schwarz et al., 2013). The recent paper by Schwarz et al. (2017) details how the instrument had been calibrated and how the measurements had been corrected for the ACCESS campaign.**

— *RC : P7L23 You note that the height of the emissions injection is very important- how good a job does the plume rise model do ? How does it work out the buoyancy of a particular fire ? Please provide a brief summary*

AC : During the ACCESS airborne campaign, flights were only performed in the remote Arctic region. Validating the fire injection heights would require measurements of vertical profiles of BC over the source regions. The performance of the plume rise model has nevertheless been studied in detail in some papers (Grell et al., 2011 ; Sessions et al., 2011). They have shown that the plume-rise model embedded in WRF-Chem improves the injection heights when compared to the satellite-observed ones. These two papers are quoted in the manuscript. Appropriate fire properties are obtained from a synergy between remote sensing observations, land use and carbon fuel datasets to determine in which columns the fires are located and the plume rise is simulated explicitly (Grell et al., 2011).

— *RC : Figure 2a is there a reason why potential temperature is more useful than just temperature ?*

AC : The potential temperature is constantly increasing with altitude, suggesting strong atmospheric stability in this study. It has also been used to underline in

Fig. 10d and Fig. 14 that the transport of BC in biomass buring plumes followed isentropes. Validating this parameter is therefore useful.

— *RC : P10L20 wrong OH and transport- the way this is written it is not quite clear what you mean. Wrong transport ? Vertical or horizontal ? Or just transport ? Do these factors and the studies you reference explain why CO is underestimated specifically between 6 – 9km ?*

AC : This sentence was unclear. It was not referring to WRF-Chem. 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 : Section 3.2 I think you are overselling the agreement between model and measurements. For example "The two profiles are well correlated with maximum CO values of 200 ppbv at* $7-8$ *km, associated with elevated BC values reaching 25 ng* kg^{-1}*." Actually the maximum in the model CO is* 150 *ppb at 6.5km. Compare to P32L10 : this is a better way of describing the model/measurement comparison. The general features were well captured.*

AC : The rewiewer might have been confused about that sentence. We just compare here the two measured profiles of CO vs BC, not model vs observations. This comparison is done later, e.g. P11L8. We have added the word "the two measured profiles" to avoid confusion.

— *RC : P11 L 1 "the influence of flaring emissions in this area is insignificant". Insignificant for what ? At what scale ? Later on in the paper you talk about some flaring plumes so it can't have been insignificant within those plumes.*

AC : The term "in this area" was not clear. The influence of flaring emissions on the vertical profiles of CO and BC sampled by the aircraft is insignificant. It has however a small influence on the background pollution off the coast of Scandanavia, as simulated by the model but not in the plumes sampled by the aircraft. According to a comment of Reviewer#2, we have decided to removed this sentence.

— *RC : P11L7-8 "The model shows appreciable skill in capturing the vertical profile of BC, but overestimates the BC mixing ratio between 2 and 3 km of altitude." Looking at the median BC concentrations, the 40km model overestimates between 1.5 – 7.5km then underestimates higher than 7.5km. Additionally, if you just went with a flat BC concentration of around 6 or 7 ng/kg the medians would probably show similar agreement. Now, I am not saying that the agreement is terrible- actually it OK, perhaps reasonably good, and it seems to capture the general features of the observations without doing a perfect job. But you read the text and it sounds like the model is doing an amazingly good job, which figures 3 and 8 show it isn't, it's just doing a reasonable job. So I think you should just tone down how well you claim the model and observations agree.*

AC : We modified the sentence as follows : "The model shows appreciable skill in capturing the general structure of the vertical profile of BC, but overestimates the BC mixing ratio in the mid-troposphere".

— *RC : P11L9 The* 30% *error in the SP2- please relate this back to the previous comment on statistical vs systematic error. Also please give numbers for the model biases.*

AC : The resulting mean bias on BC is 1.5 ng kg^{-1} and the corresponding normalized mean bias if 27%. It is much lower that biases reported for most models in the Arctic region (AMAP, 2015). Eckhardt et al. (2015) indeed reported that BC concentrations in July-September are overestimated in the mean of intercompared models by 88%.

— *RC : P11L13 Here you say the CO between 6-9km is due to biomass burning emissions, but previously you said the underestimation in CO was due to wrong OH concentrations and transport. Could it not be that the model is underestimating the biomass burning CO at this altitude ? Is that what you mean by transport ? Please clarify*

AC : The underestimation in CO mentioned previously was a general features in CTM. Here we wanted to mention the origin of the plumes we sampled in this ACCESS study. For sake of clarity we modified the last sentence : "In Sect. 4, we discuss the origin and transport of plumes leading to this noticeable increases of CO and BC between 6 and 9 km of altitude, associated with higher ozone mixing ratios."

— *RC : P13L11 You say the AOD underestimation is due to simplified SOA, but you haven't given any details of how the AOD is calculated. I don't think you can say this is due to missing SOA when you don't have a good handle on even the size distribution, let alone composition or refractive index.*

AC : This is correct. 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 strongly influences the result. The simplified SOA mechanism is a potential cause, but we can't say it is the main one. We removed that comment.

— *RC : P18L13 are the BC enhancements you are talking about in the model or in the observations ? I assume model but it's not clear*

AC : We added the word "modeled".

— *RC : P18L23 The agreement between model and measured BC is "very good"- Again I would say it's reasonable but I wouldn't say it's "very good". The model plumes in figure 5 are too diffuse and some are missing, such as the smaller amounts of BC associated with the CO plume at 8km at 0915. You get the general features. For me, "very good" would be if you could plot model vs measurement for each grid box as the aircraft passed through it and get something approaching a 1 :1 line, though I doubt that could happen in a study like this.*

AC : We corrected the sentence, replacing "very good" by "reasonable".

— *RC : Figure 8 You could do with a longer time average of the measurement data as currently it's difficult to see the structure when the markers all overlap. I also wonder if there is a way to make the aircraft easier to see as the parts that stand out are the parts where it disagrees with the model. Also it might help if the x-axis was north/south or east/west as several points in the discussion relate to spatial location and this is difficult to see in this and subsequent plots*

AC : We follow the reviewers's suggestion in applying a sliding window to calculate longer time average of the measurement data. In situ measurements are averaged

using a 2-min running mean. We have added larger white dots behind the measurements to make them easier to see versus the modeled cross-sections. We have also highlighted by magenta circles the eight airmasses discussed in the Sect. 4.4 to facilitate the reading.

— *RC : P21L18 If the aerosol from flaring had been removed by precipitation, wouldn't you still see the CO ?*

AC : We thank the reviewer for pointing this. Indeed, if the decrease of BC in flaring plumes was only due to precipitation, the CO should have remained unchanged. This is not the case, suggesting that transport of plumes from flaring sources is not only directed towards the northern coast of Norway. We have decided to remove this last sentence.

— *RC : P21L25 Didn't you say in the previous section that the flaring plumes didn't exist ?*

AC : No, we had said previously that the aircraft did not sample any flaring plume. But flares have a small influence in the area of the study (between northern Norway and Svalbard archipelago). Those flaring airmasses have been identified in Fig. 8 and 9 and highlighted by circles in the new version of the manuscript.

— *RC : Section 4.4 The discussion may be easier to follow here if you circled the plumes on one of the figures*

AC : We agree. This has been done in Fig. 8.

— *RC : P23L2 you say the fire injection lofted the plumes to 6km, but doesn't fig 10d shows that the emissions from the 8th initially remained below 4km. I'm not really sure I follow what figure 10d adds to the analysis*

AC : This is correct, thanks for pointing this. The rapid uplift up to 6 km is due to WCBs not pyroconvection. The sentence has been removed. Fig. 10d illustrates the merge of the two BB over eastrn Siberia on 12 july and their transport to the Arctic following isentropes.

— *RC : P24L20 How does figure 9 show European influence ?*

AC : Below 4 **km, the BC concentrations are dominated by the anthropogenic contribution (Fig. 9). As discussed in Sect. 4.4, this area is influenced by European emissions. To clarify the sentence, we have written "European anthropogenic influence".**

— *RC : P24L24 Please define KFCuP in the text. You refer several times to KFCuP as if it is a process itself. In the real atmosphere it is actually "convective clouds" that do things that the model is trying to represent.*

AC : We had defined KFCuP in Table 1. We replaced "KFCuP" occurences by "convective clouds" when the manuscript speaks about processes and keep "KFCup" only when the cumulus scheme is described.

— *RC : P26L4 It's frowned upon these days to refer to it as a coating, as that may not represent the actual morphology of the particle. Maybe say BC in BB plumes is more internally mixed or*

something like that. I also saw another point where you referred to coatings, please also change this.

AC : This has been corrected P24L31, P28L3 and P33L16.

— *RC : P26L11-14 I don't know what these last 2 sentences add to the analysis*

AC : We have noted that the wet deposition processes were efficient in removing BC-containing particles during transport. But in Fig. 6, we had shown that clouds in this study are mostly mixed-phased or ice clouds. The last two sentences have been kept to quote studies underlying the role of impaction scavenging in removing BC in mixed-phased or ice clouds.

— *RC : P27L19 The removal efficiencies may be low for large rain drops but not for drizzle*

AC : Here we are talking about cumulus parameterized clouds. Drizzle mostly occurs in low-level stratocumulus clouds that are resolved by the model and do not depend on KFCuP mechanism.

— *RC : P29L6 The 5th percentile of the measured or the modelled CO concentration ? Also why not do this for the BC as well ? The average lifetime of aerosols is of the order of a week because of deposition processes, mostly wet deposition. So the lifetime of aerosol that escapes wet deposition is longer. If you mean you looked at figure 3 and saw that the minimum BC in the observations was basically zero at all levels then say that. But if that was the case I still don't see the harm in taking the 5th percentile like you do with the CO.*

AC : This is correct. But according to a major comment of Reviewer#2, we have modified the calculation of the transport efficiency of BC using the ratio of BC in the base run on BC in the NoWetAll run. This is more appropriate for a model estimate. Therefore, the background concentrations are not used anylonger.

— *RC : P29L22 Perhaps this is not the case in the model but in the real atmosphere BC would only be lost if the cloud precipitated. If it evaporated the BC would still be there.*

AC : We agree with the reviewer. However we do not consider losses for the *atmosphere* 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 : P32L16 Can you suggest how the discrepancy/difference might be resolved ? Is it just because there is less precipitation in this study ?*

AC : We have suggested three reasons to explain the differences between the two papers : different year (BC emissions differ), less precipitation and a difference in the methodology. Matsui's study is only based on observations. The BC-to-CO ratios are therefore normalized by BC-to-CO ratios over the sources. If a plume emitted from a specific source diverges before reaching the receptor area, the transport efficiency calculated by Matsui's method will decrease. In our modelling study, the transport efficiencies are only influenced by precipitation and therefore higher. They illustrate the role of deposition during transport rather than the contribution of the transport itself.

— *RC : P33L30 "found to be more important" It is not clear what you mean by this. If you mean the cumulus clouds remove more of the low-level aerosol by scavenging or washout than by uplifting it then say that. It's better to say what is actually happening.*

AC : Yes, the reviewer exactly understood the message. We modified the sentence accordingly.

— *RC : P34L1 This last sentence is an odd way to end a paper. It sounds like you are using the model to validate the measurements when actually it was the other way round. Also this wasn't the main focus of your analysis. It seems to me that the main important conclusions were 1) BC is transported more efficiently into arctic from high-latitude BB sources than from east-Asian anthropogenic sources because it rains less at higher latitudes 2) The ways in which the large-scale vs subgrid convective clouds affected the BC distributions differently. It would be good if you could highlight these more in the abstract/conclusions in terms of the physical processes your results suggest are actually taking place in the real atmosphere, rather than abstract terms like grid-scale and APT*

AC : The last sentence of the paper has been removed. We have reformulated the abstract and conclusions in focusing on the physical processes : we have for instance replaced "grid scale precipitation" by "large scale coulds" and "subgrid parameterized clouds" by "convective updrafts".

— *RC : Finally, given you have actual observations it seems like there is a missed opportunity to calculate TEBC based on the measured values of BC and CO. You have hinted at your reasons for not doing so in the comparison to previous studies calculating TEBC but I'm not sure it's clear exactly why not.*

AC : We have only observations over northern Norway. Calculations of TEBC based on measurements would require also measurements of CO and BC over the different emission sources (anthropogenic, fires and flaring).

Technical corrections

— *P3L25 "lead to in the" remove 'in'*

AC : Done.

— *P3L27-31 This is a very long sentence, please split it into at least 2.*

AC : Done.

— *P8L4 add "resolution" before "to adequately"*

AC : Done.

— *P11L12 remove "but"*

AC : Done.

— *Figure 3 please make the plots slightly taller to show the detail better*

AC : The vertical bars for the median values have been replaced by diamonds of

the same color as the mean values. This makes the plot clearer.

— *P11L19 This is a very long sentence, please split it into at least 2.*

AC : P11L19 is very short. Maybe the reviewer wanted to point P11L13-16. This long sentence has been removed according to a previous comment.

— *P12 L4 remove "that"*

AC : Done.

— *Figure 4 the scale is between 0 – 1 but the text mentions values up to 2.5*

AC : This is true. But we have chosen this color scale on purpose to hightlight the contrast between the source regions and the rest. The colorbar contains arrows on the top and bottom of it to show that data exist above (resp. below) the highest (resp. lowest) contour level.

— *Figure 5 are (a) and (b) really the same grid ? Panel (a) looks much more blocky. Also label (%) on panel (c) colorscale*

AC : Fig. 5a is provided by GPCP data at 1° resolution. They have been interpolated on the WRF grid, which gives this blocky feature. We have removed "same grid" to avoid confusion. We have also added the missing label.

— *P15L1 "than in altitude", do you mean "that at higher altitudes" ?*

AC : Done.

— *P18L6 "plumes transported in altitude" do you mean "at altitude" ?*

AC : We wanted to say "in the upper troposphere". This has been corrected.

— *P18L20 "shifted a bit"- how far ? Saying "a bit" is probably a bit too colloquial for a paper (but not a review !)*

AC : To be more quantitative, the plume enriched in CO is shifted towards the north by one pixel in the model simulation, which means 40 km. It has been precised in the new version.

— *P21L17 "not due to" do you mean "did not lead to" ?*

AC : Done.

— *P27L34-35 This sentence doesn't make sense*

AC : The new sentence is the following : "The combination of low-level scavenging in the Arctic region and transport decrease from mid-latitudes is the cause of the low summertime BC concentrations."

— *Figure 13 caption Maybe rephrase to "The points are colored by the time (in hours) before the release of the trajectories in FLEXPART-WRF)"*

AC : Done.

— *P30L25 You may consider not using the word "septenrional" as it is not a commonly used word.*

AC : We say that BB sources are located far north.

— *P32L4 Again change "a bit" to "slightly"*

AC : Done.

— *P33L4 Remove "very"*

AC : Done.

— *Finally, thankyou for your interesting (but long!) paper*

AC : We also thank the anonymous reviewer for providing helpful comments.

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 ouflow 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 10μm.

— *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 dimaonds 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-trosposphere 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 Andoya (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 (Fl1, Fl2). 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, te 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 precipiatation 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 ar 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 TEBC is not smaller over these regions. The TEBC 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.

Cross-polar transport and scavenging of Siberian aerosols containing black carbon during the 2012 ACCESS summer campaign

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Abstract. During the ACCESS airborne campaign in July 2012, extensive boreal forest fires resulted in significant aerosol transport to the Arctic. A 10 day episode combining intense biomass burning over Siberia and low-pressure systems over the Arctic Ocean resulted in efficient transport of plumes containing black carbon (BC) towards the Arctic, mostly in the upper troposphere ($6-8$ km). A combination of in situ observations (DLR Falcon aircraft), satellite analysis and WRF-Chem simulations are used to understand the vertical and horizontal transport mechanisms of BC with a focus on the role of wet removal. Between the northwestern Norwegian coast and the Svalbard archipelago, the Falcon aircraft sampled plumes with enhanced CO concentrations up to 200 ppbv and BC mixing ratios up to 25 ng kg^{-1}. During transport to the Arctic region, a large fraction of BC particles are scavenged by two wet deposition processes, namely wet removal by large-scale precipitation and removal in wet convective updrafts, with both processes contributing almost equally to the total accumulated deposition of BC. Our results underline that applying a finer horizontal resolution (40 instead of 100 km) improves the model performance, as it significantly reduces the overestimation of BC levels observed at a coarser resolution in the mid-troposphere. According to the simulations at 40 km, the transport efficiency of BC (TE_{BC}) in biomass burning plumes ~~is about 60%, which is~~ was larger (60%), because it was impacted by small accumulated precipitation along trajectory (~~APT) (~~1 mm). In contrast ~~TE_{BC} is~~ it was very small ($< 30\%$) and ~~APT is~~ precipitation amounts were larger ($5-10$ mm) in plumes influenced by urban anthropogenic sources and flaring activities in Northern Russia, resulting in transport to lower altitudes. TE_{BC} due to ~~grid~~ large scale precipitation is responsible for a sharp meridional gradient in the distribution of BC concentrations. Wet removal in ~~subgrid parameterized clouds (cumuli)~~ cumulus clouds is the cause of modeled vertical gradient of TE_{BC}, especially in the mid-latitudes, reflecting the distribution of convective precipitation, but is dominated in the Arctic region by the ~~grid-scale~~ large scale wet removal associated with the formation of stratocumulus clouds in the PBL that ~~produced~~ produce frequent drizzle.

1 Introduction

The Arctic region is particularly sensitive to environmental change, as it is predicted to warm faster than any other region (Manabe et al., 1992). ~~The summertime extent of~~ In the past decades, a significant loss of the summertime sea-ice ~~has decreased significantly in recent decades~~ extent has been detected (e.g., Lindsay et al., 2009). A ~~warmer Arctic may lead to a substantial expansion of resource extraction and seasonal shipping traffic due to improved sea access (Corbett et al., 2010; IPCC, 2013). In addition to long-lived greenhouse gas-induced warming and feedbacks, Arctic warming may also be caused by~~ substantial increase in oil and gas extraction activities as well as in shipping may result from a warmer Arctic (Corbett et al., 2010; IPCC, 2013). Recently, studies have highlighted the role of shorter-lived climate ~~forcing agents (Tomasi et al., 2007; Law and Stohl, 2007; Quinn et al., 2~~ ~~since~~ forcers (SLCFs), e.g. aerosols and ozone ~~perturb the radiative balance of the Arctic~~ in this warming (Tomasi et al., 2007; Law and Stoh perturbing the polar radiative balance directly by absorbing and scattering radiation (Shindell and Faluvegi, 2009), and indirectly due to aerosol effects on clouds properties leading to increases in shortwave scattering efficiency and IR emissivity of Arctic clouds (Garrett and Zhao, 2006; Lubin and Vogelmann, 2006; Zhao and Garrett, 2015). Black carbon (BC) is only a minor contributor to aerosol mass but is clearly a significant ~~short-lived climate forcing agent~~ SLCF in the atmosphere and ~~when it is~~ can be deposited onto snow and ice surfaces, reducing their albedo due to multiple scattering in the snowpack and the much larger absorption coefficient of BC than ice (Warren and Wiscombe, 1980; Hansen and Nazarenko, 2004; Jacobson, 2004), modifying snow grain size and ~~driving changes in snow meltand~~ facilitating snow melt, which drives changes in surface temperature (Flanner et al., 2007, 2009; Jacobson, 2010).

Winter and spring have been the most favored seasons for Arctic aerosol research ~~has predominantly focused on winter and spring while fewer measurement campaigns have been performed during summer when concentrations~~ while fewer summertime measurement studies have been carried out when aerosol mixing ratios are generally smaller (Law and Stohl, 2007). More attention has been given to surface measurements, whereas only very few ~~summertime airborne studies~~ airborne campaigns (e.g. POLARCAT, ARCTAS) of aerosol chemical composition have been ~~carried out~~ performed in summer (Schmale et al., 2011). During summer, inefficient long-range transport reveals that summertime aerosol originates from the marginal Arctic (sea-ice boundary and boreal forest regions (Warneke et al., 2010)) and from episodic ~~transport events, caused by rapid transport from heavily polluted but varied source types and regions, and producing~~ and rapid transport events from heavily areas, and leading to dense haze layers aloft ~~that rarely reach surface monitoring sites~~ (Browse et al., 2012; Pierro et al., 2013; Ancellet et al., 2014; Law et al., 2014). Some studies (Brock et al., 1989; Stohl, 2006; Paris et al., 2009) identified boreal and temperate forest fires, especially Siberian fires, as a ~~significant source of BC during the summer. Boreal forest fires may be the~~ dominant source of BC ~~for the Arctic during intense fire years. Smoke from biomass burning tends to be highly stratified but mostly present at higher altitudes (3 – 10~~ at this season, especially above the Planetary Boundary Layer (PBL) at altitudes between 3 and 10 km~~) because forest fire emissions can be injected well above the boundary layer during the flaming phase~~ , and even into the lower stratosphere by injection in pyrocumulus clouds (Lavoué et al., 2000). Air masses can also be uplifted ~~during transport to the Arctic~~ to the polar tropopause during transport in Warm Conveyor

Belt (WCBBelts (WCBs) of synoptic low pressure systems, which have been found to be a dominant mechanism in vertically redistributing pollution (Browning and Monk, 1982; Cooper et al., 2002). Climatological trajectory analyses have shown that . Eckhardt et al. (2004); Madonna et al. (2014) have highlighted frequent WCBs over northeastern China and Russia have a large frequency of WCB events (Eckhardt et al., 2004; Madonna et al., 2014)based on climatological analyses. During the IPY (International Polar Year(IPY) in 2008, different studies using backward trajectory and lagrangian particle dispersion model analyses have highlighted WCB as an important pathway to transport pollution into the polar tropopause region during summer (Roiger et al., 2011; Sodemann et al., 2011; Matsui et al., 2011). These studies lagrangian studies (e.g., Roiger et al., 2011; Sodemann et al
several Asian pollution plumes transported to the Arctic after a strong uplift in WCBs located over the Russian east-coast and subsequently transported to the Arctic. Franklin et al. (2014) and Taylor et al. (2014) have documented the impact of wet removal in Canadian biomass burning plumes and confirmed that wet deposition is the dominant mechanism for BC removal from the atmosphere and consequently determining its lifetime, its atmospheric burden, and affecting vertical profiles of number and mass concentration.

State-of-the-art global chemical transport models have large discrepancies and spread in the prediction of Arctic BC , including incorrect seasonality and order-of-magnitude errors in modeling Arctic BC mixing ratios and seasonal variability (Shindell et al., 2008; Koch et al., 2009; Lee et al., 2013; Schwarz et al., 2013; AMAP, 2015). Schwarz et al. (2010) showed that uncertainties in modeling of aerosol physical and chemical processing are extremely important, with lesser roles for emissions and dry transport. One of the main causes of these differences is large uncertainties in the Vignati et al. (2010), Bourgeois and Bey (2011) and Browse et al. (2012) identified the representation of wet removal of BC adopted in aerosol models (Vignati et al., 2010; Bourgeois and Bey, 2011), but a quantitative understanding of this process is still limited (Browse et al., 2012) one of the main reasons of these differences. More recently, Breider et al. (2014) compared GEOS-Chem model results with mean vertical profiles of BC modeled BC with observations from the ARCTAS summer campaign. The observations in July were less mean vertical profile of BC measurements in July was always lower than $25\ \mathrm{ng\ m^{-3}}$throughout the vertical profile, but the model strongly overestimated the concentrations at high altitudes due to influenced by long-range transport of deep convective outflow from Asian pollution (summer monsoon) and Siberian fire plumes. This overestimation in remote regions confirms previous results from Koch and Hansen (2005). Wang et al. (2014) used a more recent version of this model, increasing the result agrees with the overestimation of BC concentrations in the upper Arctic troposphere found by Koch and Hansen (2005). Despite an enhanced scavenging efficiency of BC in deep convection, but they also Wang et al. (2014) noticed a strong overestimation of BC in the mid-troposphere. The over prediction of BC concentrations during the summer might be due to the inability models to produce rain by convective clouds/fog that readily form during the Sharma et al. (2013) explained this overprediction of BC by an incorrect production of convective precipitation in summer in the Arctic(Sharma et al., 2013). These results reinforce the fact that aerosol and cloud physical and chemical processing (removal, oxidation and microphysics) is the primary source of uncertainty in modelling modeling aerosol distributions in the Arctic (Eckhardt et al., 2015).

During the summer of 2012, extensive boreal forest fires occurred both in western and in eastern Siberia (in the Yakutsk region)~~, influencing the atmospheric composition in the Arctic~~. These fires were associated with a hottest summer on record over Siberia, leading to ~~very high emission rates of a number of trace species and aerosols~~ elevated emissions of trace gases and particles (Ponomarev, 2013). In this paper, we focus on a case of pollutant transport from Asia and Russia across the Arctic because they may ultimately lead to ~~in~~ the transport of pollution to Europe and North America. During the ACCESS ~~campaign, the~~ (Arctic Climate Change, Economy and Society) campaign over northern Norway in July 2012, the DLR-Falcon 20 aircraft sampled plumes representative of cross-polar transport reaching the Norwegian coasts. Combining an analysis of aircraft measurements, satellite observations, lagrangian trajectories and simulations from a regional online chemistry transport model (CTM), we investigate the horizontal spatial and vertical structure of BC-containing air masses~~,~~. We focuse on the factors affecting this transport, including the meteorological situation~~and~~ , the dry and wet deposition processes and we assess the relative contribution of anthropogenic, flaring and biomass burning sources to the BC concentrations. ~~This synergy of methods gives good accuracy on short time scales for source regions with close proximity to the Arctic and enables us to treat pathway-dependent aerosol removal and quantify contributions from distant sources.~~ Section 2 describes the ACCESS campaign, as well as the modeling tools used in this study. Model output is validated against ACCESS observations in terms of meteorological and chemistry variables, aerosol optical depth (AOD), precipitation and cloud structures (Sect. 3). The export processes, the identification of plumes and the transport pathways are described in Sect. 4. Finally, Sect. 5 investigates the contribution of the various ~~deposition~~ pathway-dependent aerosols removal processes and the transport efficiency of BC reaching the Norwegian coasts.

2 Methodology

2.1 ACCESS campaign

Roiger et al. (2015) have recently given a detailed overview of the ACCESS aircraft campaign. Therefore we only describe it briefly here. This campaign was based out of Andenes, Norway (69.3°N, 16.1°~~W~~E), in July 2012 and included 14 flights over the northwestern coast of Norway using the DLR Falcon-20 aircraft (Fig. 1). Most of the flights were devoted to studying the impacts of local pollution sources (ships, oil and gas extraction) on Arctic atmospheric composition (Marelle et al., 2016), but pollution from remote sources was also measured during some specific flights in the middle and upper troposphere (Roiger et al., 2015). In this ~~paper~~study, we focus on all flights but describe in particular the two flights that both occurred on 17 July (namely flights 17a and 17b), which were specifically dedicated to probe father into the Arctic in order to study plumes transported from boreal and Asian sources (Fig. 1). ~~IASI (~~Infrared Atmospheric Sounding Interferometer (IASI))-retrieved CO total columns as well as global trace gas forecasts from the Monitoring Atmospheric Composition and Climate (MACC) were used during the campaign to target such flights in order to identify polluted air masses in the mid and upper troposphere (Roiger et al., 2015). In addition to the suite of meteorological instruments, the Falcon-20 was equipped with trace gas (NO_x, SO_2, O_3, and CO) and aerosol instrumentation including BC. CO was measured every second from VUV fluorescence with an accuracy

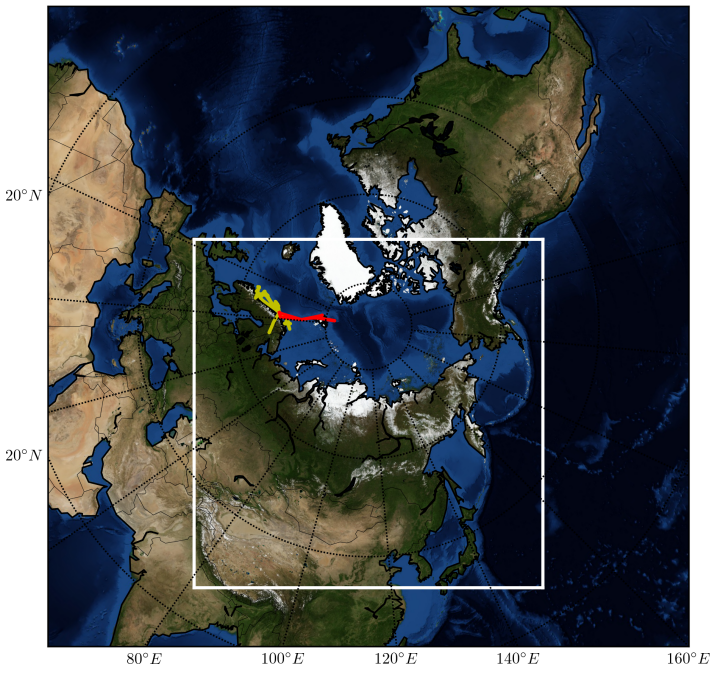

Figure 1. Map of the WRF-Chem domain (white) and the flights conducted as part of ACCESS used to evaluate the model. The Falcon-20 flights are in yellow. Flights of the 17 July are highlighted in red.

of 10%.

BC mass mixing ratios were measured using a single-particle soot photometer (SP2, Droplet Measurement Technologies, Boulder, CO, USA) based on laser-induced incandescence. The instrument measures the refractory black carbon (rBC; hereafter referred to as BC) on a single-particle basis (Schwarz et al., 2006). Particles are drawn through a high intensity 1064 nm Nd:YAG laser which heats BC-containing particles to incandescence. The instrument has been calibrated using the recommended calibration material (Fullerene soot). The individual particle masses of the BC cores of BC-containing particles were averaged over 10 s intervals to obtain mass mixing ratios of BC. Since the SP2 instrument detected BC cores only in the size range of $80 - 470$ nm, assuming a density of $1.8\ \mathrm{g\,cm^{-3}}$ (Moteki et al., 2010), the derived mass mixing ratios were scaled by a factor to account for particles outside the detection range (as in e.g. Schwarz et al. (2006)) to obtain the total mass mixing ratio. The factor was derived from the average mass size distribution of BC cores separately for each flight and ranged from 1.1 to 1.55, depending on the encountered aerosol type. ~~Absolute uncertainty of BC particle mass is within 10%, the uncertainty of the~~ Total relative uncertainty of the derived total BC mass mixing ratio is about 30% (??).

Table 1. ~~Parameterizations~~ Schemes and ~~options~~ parameterizations used for the WRF-Chem simulations

Physical process	WRF-Chem option
Planetary boundary layer	Mellor-Yamada-Janjic (Janjic, 1994)
Surface layer	Monin-Obukhov
Land surface	Noah Land Surface model (Ek et al., 2003)
Microphysics	Morrison 2-moment (Morrison et al., 2009)
Shortwave radiation	RRTMG (Iacono et al., 2008)
Longwave radiation	RRTMG (Iacono et al., 2008)
Cumulus parameterization	Kain-Fritsch-CuP (KFCuP) (Berg et al., 2013, 2015)
Photolysis	Fast-J (Wild et al., 2000)
Gas phase chemistry	CBM-Z (Zaveri and Peters, 1999)
Aerosol model	MOSAIC 8 bins with aqueous chemistry (Zaveri et al., 2008)

2.2 Modeling strategy

2.2.1 WRF-Chem model set-up

In order to study the transport and processing of aerosols, regional chemical transport simulations are performed using the fully compressible mesoscale meteorological Weather Research and Forecasting model ~~with chemistry~~ (Skamarock and Klemp, 2008) with chemistry modules (WRF-Chem Version 3.5.1) (Grell et al., 2005). ~~WRF model is a fully compressible meso-scale meteorological model designed for research experiments as well as operational weather forecasts (Skamarock and Klemp, 2008).~~ The chemistry component ~~(Grell et al., 2005) contains a number of modules for simulating aerosol direct and indirect effects and chemistry processes, which~~ is fully consistent with the meteorological ~~component~~ part (Fast et al., 2006). The different ~~parameterizations and options used for the WRF-Chem simulations~~ schemes and parameterizations used in this work are summarized in Table 1. Land surface processes are resolved using the Noah Land Surface model scheme with 4 soil layers (Ek et al., 2003). ~~Planetary Boundary Layer (PBL)~~ PBL processes are parameterized according to the Mellor-Yamada-Janjic local scheme (Janjic, 1994). It is coupled to a surface layer based on Monin-Obukhov with Zilitinkevich thermal roughness length and standard similarity functions from look-up tables. We use the Morrison 2-moment ~~(Morrison et al., 2009) microphysics scheme , which contains hydrometeor classes for water vapor, cloud water, rain, cloud ice, snow, and graupel~~microphysics scheme (Morrison et al., 2009). To represent the effects of subgrid-scale convective processes on the grid variables, we use the Kain-Fritsch convective implicit parameterization (Kain, 2004), which has been recently improved by Berg et al. (2013) (details below). The shortwave and longwave radiation schemes are from the RRTMG model (Iacono et al., 2008). The Fast-J photolysis scheme (Wild et al., 2000) is coupled with hydrometeors as well as aerosols.

The model domain ~~, shown in Figure 1, has a 40 horizontal grid resolution~~ is set up on a polar stereographic grid ~~and a distance of 8800~~ of 8800×8800 km^2 (Fig. 1). It has a horizontal resolution of 40 km ~~in both horizontal directions. There are~~ and 50 vertical levels ~~, and the~~ up to 50 hPa. The lowest vertical grid spacing is about 25 m. The ~~model top is set to 50 using~~ simulation uses a model time step of 180 s~~. The simulation~~ , begins on 00 UTC 4 July 2012 (after 3 days spinup) and ends on 00 UTC 21 July 2012. Initial conditions and lateral boundaries of the meteorological fields are provided by ~~NCEP (~~National Centers for Environmental Prediction (NCEP) Global Forecast System (GFS) Final Analysis (FNL) 1° resolution data, which has 26 pressure levels and is updated every 6 hours. The simulations are also initialized with NCEP-archived 0.5° sea-surface temperatures fields (updated every 6 hours) and sea ice data (updated every day). The modeled horizontal wind components, atmospheric temperature and humidity are nudged towards the 6-hourly NCEP/FNL reanalysis data (Stauffer and Seaman, 1990) above the PBL. The ~~chemistry and aerosol species are initialized with reanalysis data from the~~ offline global chemical transport model MOZART-4 (the Model for Ozone and Related Chemical Tracers) ~~(Emmons et al., 2010). Spatially and~~ provides initial and spatially and temporally (6-hourly) varying ~~chemical boundary conditions are also provided by MOZART-4. MOZART-4 data has a 2.5° horizontal resolution and uses regional and global anthropogenic and natural emissions inventories~~ boundary conditions for chemistry and aerosol species ~~. The MOZART-4 reanalysis fields, provided by NCAR/NESL, contain 28 vertical levels with a maximum height at the 2 pressure level.~~(Emmons et al., 2010).

In this study, the MOSAIC (Model for Simulating Aerosol Interactions and Chemistry) aerosol model (Zaveri et al., 2008) coupled with the CBM-Z (Carbon Bond Mechanism) photochemical mechanism (Zaveri and Peters, 1999) is used. MOSAIC ~~simulates a wide variety of aerosol species (sulfates, nitrates, ammonium,~~ employs a sectional approach to simulate size distributions of a wide number of internally mixed aerosol components (BC, organics, sea salt, sulfate, nitrate, ammonium, dust, ~~BC, organics,~~ and others)~~using a sectional approach to represent aerosol size distributions by dividing up the size~~ . The size distribution for each species ~~into several size bins (8 in this study) and assumes that the aerosols are internally mixed in each bin~~is divided into eight size bins between 39 nm and 10 µm. The MOSAIC aerosol scheme includes physical and chemical processes of nucleation, condensation, coagulation, aqueous phase chemistry, and water uptake by aerosols. It is also coupled to the microphysics and cumulus schemes, and to the shortwave and longwave radiation schemes.

Aerosol dry deposition is modeled using the "Resistance in Series Parameterization" (Wesely and Hicks, 2000)~~, following the approach of Binkowski and Shankar (1995)~~. Wet removal of aerosols by grid-resolved stratiform clouds/precipitation includes in-cloud ~~removal (rainout)~~ and below-cloud removal ~~(washout)~~ by rain, snow, graupel by Brownian diffusion, interception, and impaction mechanisms, following Easter et al. (2004) and Chapman et al. (2009). ~~Aerosol-cloud interactions were included~~ Gustafson et al. (2007) included aerosol-cloud interactions in the model ~~by Gustafson et al. (2007) for calculating~~ to calculate the activation and ~~re-suspension~~ resuspension between dry aerosols and cloud droplets. Furthermore, ~~Berg et al. (2015) have modified~~ WRF-Chem ~~to include treatments of a number of factors and processes important for accurately representing~~ model is now able to represent aerosol and trace gases within ~~sub-grid-scale convective clouds, including~~ subgrid scale convective clouds. Berg et al. (2015) indeed included fractional coverage of active and passive clouds, vertical transport, activation and

resuspension, wet removal, and aqueous chemistry for cloud-borne particles. This new treatment, coupled to MOSAIC aerosol scheme, is important ~~for including additional realism in regional-scale modeling studies that require the use of cumulus parameterizations when investigating the~~ in our Arctic-scale modeling study to investigate the cloud effects on aerosol within parameterized shallow and deep ~~convection, and aerosol effects on cloud droplet number~~convective clouds.

2.2.2 Emissions

~~The fire emissions inventory has been estimated using~~ Our simulations use daily, 1 km horizontal resolution fire emissions from the Fire INventory from NCAR (FINN-v1) (Wiedinmyer et al., 2011)~~. FINN provides emissions on a per fire basis~~ , based on fire count information from the MODIS (Moderate Resolution Imaging Spectrometer) instrument. ~~One of the most important aspects of simulating wildfire plume transport is the height at which emissions are injected. WRF-Chem contains an~~ An integrated one-dimensional plume rise model ~~to determine the appropriate injection layer (Freitas et al., 2007; Grell et al., 2011; Sessions et al., The plume rise model accounts~~ is embedded in WRF-Chem, accounting for thermal buoyancy associated with fires and local atmospheric stability~~, and is applied to~~ . It helps to determine the appropriate injection layer and distribute the fire emissions vertically (Freitas et al., 2007; Grell et al., 2011; Sessions et al., 2011). Anthropogenic emissions ~~were~~ are taken from the HTAPv2 (Hemispheric Transport of Air Pollution version 2~~(HTAPv2)~~ $0.1^\circ \times 0.1^\circ$ inventory (http://edgar.jrc.ec.europa.eu/htapv2/index.php?SECURI Volatile organic compounds (VOCs) are given as a bulk VOC mass and distributed into CBM-Z emission categories assuming the speciation reported by Murrells et al. (2010). ~~Time profiles are applied to anthropogenic emissions to account for the daily and weekly cycle of~~ Hourly and weekly variability of anthropogenic emissions is applied for each emission sector (van der Gon et al., 2011). ~~Soil-derived (dust) and sea salt~~ Flaring emissions are from the ECLIPSE (Evaluating the CLimate and Air Quality ImPacts of Short-livEd Pollutants) inventory (Stohl et al., 2015). Sea salt and soil-derived (dust) aerosol emissions are incorporated into WRF-Chem (Shaw et al., 2008)~~, and biogenic~~ . Biogenic emissions are calculated online following the MEGAN (Model of Emissions of Gases and Aerosols from Nature~~(MEGAN)~~ (Guenther, 2006).

2.2.3 Model simulations

The fine-scale structure of the Arctic atmosphere (e.g. due to the thermal stratification) poses a major problem for atmospheric transport model simulations. Here we run the WRF-Chem on a polar-stereographic projection at relatively high horizontal (40 km) and vertical (50 levels up to 50 hPa) resolution to adequately represent the actual structure of the Arctic atmosphere, avoiding an overly rapid diffusion and decay along the boundaries of advected plumes (Rastigejev et al., 2010). A total of nine simulations are conducted: a baseline WRF-Chem simulation (BASE) and eight sensitivity tests to investigate the effect of several processes and emission sources on BC transport to the Arctic: a run without biomass burning emissions (NoFire), a run without anthropogenic emissions (NoAnthro), a run without emissions from flaring activities (NoFlr), a run without dry deposition (NoDry), a run without wet deposition in grid scale clouds (NoWet), a run without wet deposition in parameterized cumulus clouds (NoWetCu), and a run without wet removal in any clouds (NoWetAll). An additional simulation (Run100)

Table 2. List of WRF-Chem simulations performed

Name	Description
BASE	Baseline simulation at 40 km
NoFire	Run at 40 km without biomass burning emissions
NoAnthro	Run at 40 km without anthropogenic emissions
NoFlr	Run at 40 km without flaring emissions
NoDry	Run at 40 km without dry deposition
NoWet	Run at 40 km without wet deposition in grid scale clouds
NoWetCu	Run at 40 km without wet deposition in parameterized cumulus clouds
NoWetAll	Run at 40 km without wet removal in any clouds
Run100	Run at 100 km with the same vertical resolution as in the BASE run

similar to the BASE simulation, but at a coarser resolution (100 km) is also performed to study the influence of horizontal resolution on the transport and deposition of carbonaceous aerosols. The list of the WRF-Chem runs performed is summarized in Table 2.

2.2.4 FLEXPART-WRF

We use the Lagrangian particle dispersion model FLEXPART-WRF (Fast and Easter, 2006; Brioude et al., 2013) to assess pollution transport and dispersion from individual sources, and to identify the origins of measured pollution plumes during the ACCESS campaign. FLEXPART-WRF is derived from the FLEXPART dispersion model (Stohl et al., 2005), ~~but is~~ driven by meteorological fields simulated by WRF. ~~In this study,~~ We use FLEXPART-WRF ~~is run~~ in backward mode to study source-receptor relationships: 10000 particles are released at a receptor point and are transported backward in time using the meteorological fields from the ~~WRF-Chem simulation~~BASE run. Source-receptor relationships are quantified by calculating the potential emission sensitivities (PES), which represent the amount of time spent by the particles in every grid cell. In this paper, meteorological variables are interpolated in time and space over trajectories from the WRF runs. FLEXPART-WRF is therefore useful to identify the origins and transport pathways of pollution plumes observed during the ACCESS campaign and to derive precipitation and deposition efficiencies along those transport pathways (Sect. 5).

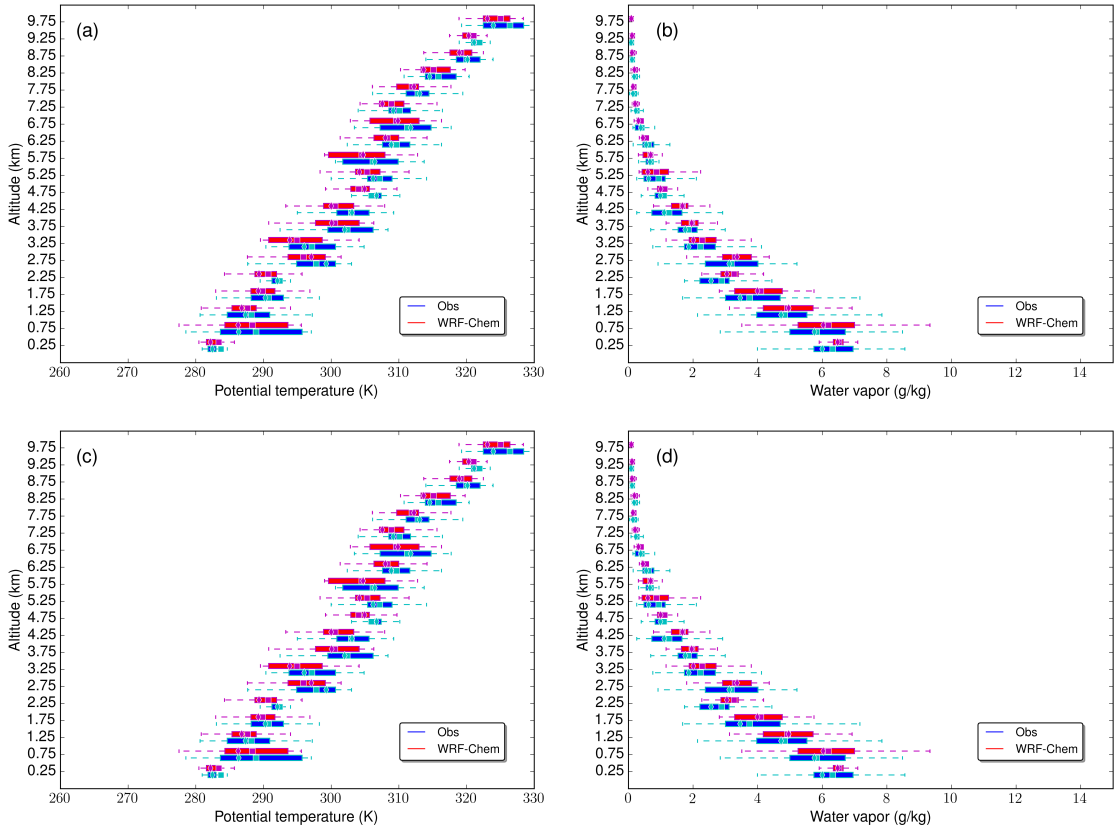

Figure 2. Vertical profiles of observed (blue) and ~~modelled~~ modeled (red) (a) potential temperature (K), (b) water vapor mixing ratio ($\mathrm{g\,kg^{-1}}$), (c) wind speed ($\mathrm{m\,s^{-1}}$) and (d) wind direction ($^{\circ}$) interpolated along all the 14 Falcon flight tracks. Boxes represent the interquartile range, ~~vertical bars~~ diamonds are the median values, squares show the means and whiskers show to the mininum and maximum of the data.

3 Model validation

3.1 Meteorological parameters

To evaluate the skill in the ~~modelled~~ modeled weather and transport patterns predictions, ~~we first compare ACCESS observations of~~ model results are first compared to ACCESS observations in terms of potential temperature, relative humidity (RH), wind speed and wind direction~~to model results. The simulated meteorology is extracted every minute along the flight track using linear interpolation at the exact~~ . The model outputs available at hourly intervals are linearly interpolated in latitude, longitude, altitude and time~~using the model output available at hourly intervals~~, every minute along the flight track. The ensemble of key observed and simulated ~~data~~ meteorological parameters interpolated along the 14 flight tracks are then gathered to compute vertical profiles~~of key meteorological parameters influencing the long range chemical transport~~. The comparison with Falcon observations is shown in ~~Figure~~Fig. 2. The predicted temperature is in very close agreement with airborne measurements,

Table 3. Performance statistics for meteorological variables and chemical species (BC, CO). For all flights, R^2, MB, $RMSE$ and NMB represent the Pearson correlation coefficient, the mean bias, the root-mean-square error, and the normalized mean bias, respectively.

Variable (unit)	R^2	MB	$RMSE$	$NMB(\%)$
Pressure (hPa)	0.999	10.14	13.61	1.37
Potential temperature (K)	0.997	-0.96	1.60	-0.32
Relative humidity (%)	0.709	1.29	14.60	1.73
Wind speed ($\mathrm{m\,s^{-1}}$)	0.866	-0.27	2.39	-3.33
Water vapor mixing ratio ($\mathrm{g\,kg^{-1}}$)	0.924	0.15	0.61	3.67
BC ($\mathrm{ng\,kg^{-1}}$)	0.468	1.58	7.44	27.30
CO (ppbv)	0.524	1.47	17.63	1.49

with only a small negative bias (about -1 K) at high altitudes. The modeled wind direction also matches the observations closely, with a slight positive normalized mean bias ($NMB < 6\%$) in the boundary layer. We note however that statistical performances for wind direction should be taken with caution as they are derived from a vector quantity. The simulated wind speed shows good agreement with a correlation coefficient of $R^2 \simeq 0.866$, although a slightly negative bias (lower than 3%) at altitudes above 6 km. The water vapor mixing ratio and the resulting RH are also well reproduced by the model with high correlation coefficients (Table 3). Overall ~~the WRF model~~ WRF-Chem shows appreciable skill in capturing the variability seen in the observations, suggesting that the large scale transport patterns and major aspects of flow conditions are well represented in the WRF simulations for this campaign.

3.2 Trace gases and aerosols

Figure 3 shows the vertical profiles of the CO and BC mixing ratios measured during the ACCESS campaign and simulated by WRF-Chem (BASE and Run100 simulations). On average, the ACCESS measurements show enhanced BC and CO concentrations between 6 and 9 km of altitude. The two measured profiles are well correlated with maximum CO values of 200 ppbv at $7-8$ km, associated with elevated BC values reaching 25 $\mathrm{ng\,kg^{-1}}$. The corresponding mean values for CO and BC in this altitude range are 135 ppbv and 12 $\mathrm{ng\,kg^{-1}}$, respectively. The small underestimation in CO between 6 and 9 km is a common feature observed by most models (Emmons et al., 2015; AMAP, 2015)~~and is caused by chemical schemes leading to wrong OH andtransport (Monks et al., 2015). A sensitivity test (not shown) based on HTAPv2 emissions versus ECLIPSE (Evaluating the CLimate and Air Quality ImPacts of Short-livEd Pollutants) inventory with much larger flaring emissions in northern Russia (Stohl et al., 2015) has suggested that the influence of flaring emissions in this area is insignificant~~. Variability in models, run with the same emissions, appears to be driven by differences in chemical schemes influencing modeled OH and/or differences in modeled vertical export efficiency of CO (via frontal or convective transport) from mid-latitude source

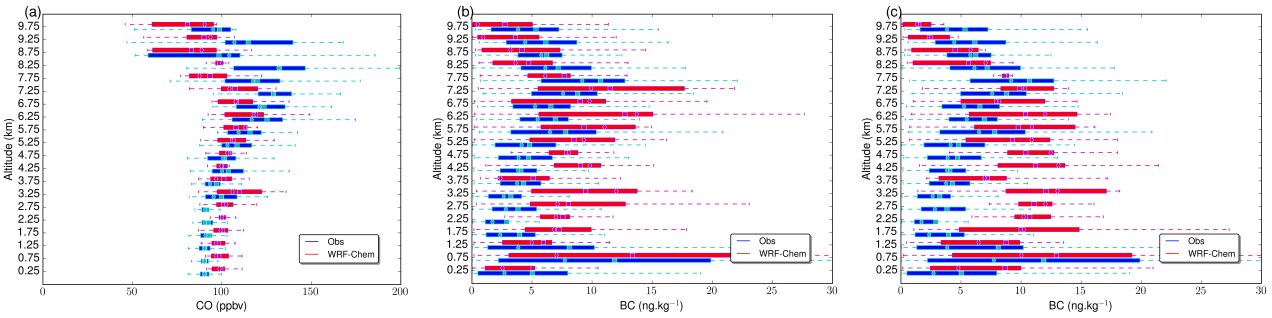

Figure 3. Vertical profiles of observed (blue) and ~~modelled~~ modeled (red) mixing ratios of (a) CO (ppbv), (b) BC ($\mathrm{ng\,kg^{-1}}$) at a horizontal resolution 40 km (BASE), and (c) BC ($\mathrm{ng\,kg^{-1}}$) at a horizontal resolution 100 km (Run100) interpolated along all the 14 Falcon flight tracks. Boxes represent the interquartile range, ~~vertical bars~~ diamonds are the median values, squares show the means and whiskers show to the mininum and maximum of the data.

regions to the Arctic (Monks et al., 2015). A noticeable increase in ozone concentrations is also clearly seen at 7 km, with a mean value of 135 ppbv (not shown). The vertical profiles of CO, BC and ozone ~~demonstrate~~ suggest that, during summer 2012, ~~transported pollution~~ pollution transported to the Arctic increased trace gases and aerosol ~~concentrations~~ mixing ratios in the middle and upper troposphere compared to typical clean background values (Roiger et al., 2015). The statistical performance of the model in regards to the representation of key chemical species extracted along flight tracks are given in Table 3. The comparison of simulated CO ($NMB < 1.5\%$) against the Falcon flight ~~observations showed~~ measurements proves that the model predictions are able to capture the magnitude and temporal variability seen in observed values. ~~The model shows appreciable~~ The model demonstrates reasonable skill in capturing the general structure of the vertical profile of BC, but overestimates the BC mixing ratio ~~between 2 and 3 of altitude~~in the mid-troposphere. The resulting ~~normalized~~ mean bias on BC is 1.5 $\mathrm{ng\,kg^{-1}}$ and the corresponding normalized mean bias (27.3%) is lower than the error on the SP2 instrument (30%) and much lower than biases reported for most models ~~on~~ in the Arctic region~~(Eckhardt et al., 2015; AMAP, 2015).~~ . Eckhardt et al. (2015); AMAP (2015) indeed reported that BC concentrations in July-September are overestimated in the mean of intercompared models by 88%. The mean profile of BC sampled during the ACCESS campaign is of the same order of magnitude as those reported by Breider et al. (2014) (25 $\mathrm{ng\,m^{-3}}$ in July 2008) and by Schwarz et al. (2013) during the HIPPO (HIAPER Pole-to-Pole Observations) campaign at similar latitudes (~~between 60° and 80°~~60 – 80°N), ~~but~~ in the remote Pacific region ($10 - 20$ $\mathrm{ng\,kg^{-1}}$). ~~The noticeable increase of CO , an incomplete combustion product,~~ In Sect. 4, we discuss the origin and transport of plumes leading to this noticeable increases of CO and BC between 6 and 9 km of altitude~~in this study~~, associated with higher ozone mixing ~~rations, points towards a regular transport of forest fire/biomass burning emissions to northern Norway, providing us with the opportunity to study to chemical and aerosol composition in these plumes.This is discussed further in Sect. 4 (source attribution).~~ratios.

3.3 Model resolution

The result of the Run100 simulation is also shown in Fig. 3. The corresponding BC vertical profile shows the same enhancement between 6 and 9 km as the BASE run, but it also clearly highlights a strong overestimation (×3) in the mid-troposphere ~~.~~ (1.5 – 5 km). The CO concentrations are also enhanced in the Run100 simulation by 4 – 5 ppbv as compared to the BASE run. This suggests that, at a coarser resolution, the model is unable to resolve the fine structure of plumes transported ~~in altitude~~aloft, illustrates the difficulty to represent the cloud and precipitation structures and points for the need for improved representation of BC processing in global models~~and the necessity to compare models with measurements in Arctic regions~~. More generally, global models are commonly run at horizontal and vertical resolutions that are inadequate for representing the actual structure of the Arctic atmosphere, mostly because of computational limited resources. Schwarz et al. (2013) studied ~~that~~ the bias between BC measurements and the AeroCOM model suite in remote regions. Global models ~~always~~ generally overestimated BC mass concentration, especially in the Arctic upper troposphere where the overestimation was about a factor of 13, suggesting that the aerosol lifetime and removal was not correctly in models. Model features that govern the vertical distribution and lifetimes of ~~short-lived climate forcers~~ SLCFs in the Arctic atmosphere must still be improved (AMAP, 2015). Ma et al. (2013) showed that BC was better simulated with higher spatial resolution, and described that there is likely less wet removal at higher spatial resolution since aerosols and clouds do not overlap as much. In our study, at a horizontal resolution of 40 km, the model-to-observations comparison indicates that the ~~summertime~~ WRF-Chem model correctly represents the transport pathways of pollution into the Arctic ~~including the mid-latitude pollution transport and forest fire~~in summer from mid-latitudes and wildfire/biomass burning~~are well represented in the WRF-Chem model~~. At a finer resolution, the model is also able to better represent mean vertical motions associated with large-scale synoptic systems, i.e. conveyor belts, that lift PBL aerosols into the free troposphere. Resolution can therefore affect long-range transport, since it affects how high aerosols are lifted into the free troposphere.

3.4 Aerosol optical depth

To evaluate the total aerosol column simulated by WRF-Chem during the ACCESS campaign, we calculate the aerosol optical depth (AOD) at 550 nm, defined as the integrated extinction coefficient over a vertical column of unit cross section, taking into account the attenuation of the radiance by aerosol scattering and absorption. This is compared to the AOD retrieved at the same wavelength by the MODIS instrument aboard Aqua satellite passing over the equator in the afternoon. ~~The MODIS level 3 products used in this study are described by Hubanks et al. (2008). MODIS AOD fields at 550 are retrieved from daily observations averaged~~ We average, over the period of the ACCESS campaign ~~and taking into account~~ (4-21 July), daily observations of AOD obtained from MODIS level 3 products (Hubanks et al., 2008), accounting for missing observations primarily due to the presence of clouds within the column ~~.~~ (Fig. 4~~shows average maps of observed and simulated AOD at 550~~). Note that the AOD retrieved from MODIS above Terra satellite is ~~very~~ similar to that obtained from Aqua (not shown). In general, the model correctly represents the main features of the spatial ~~AOD distribution~~ distribution of AOD, including the

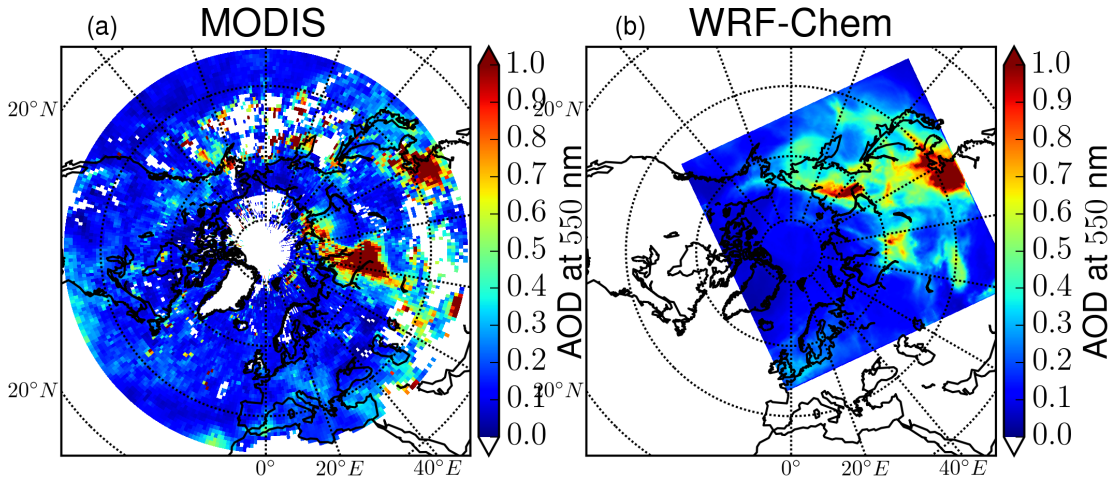

Figure 4. Aerosol optical depth (AOD) at 550 nm (a) measured by MODIS instrument aboard Aqua and (b) simulated by a Mie code in the WRF-Chem model, averaged between 4 and 21 July 2012. White areas in (a) indicate the presence of clouds, preventing the AOD retrieval.

large values over northeastern China and western Siberia. In China, high AODs reaching 2.3 (1.8 in the model) are linked to high pollution levels in the PBL combined with strong dust episodes. Over western Siberia, the extended area with substantially enhanced values of AOD (up to 2.5) is due to large fires (Ponomarev, 2013) and with a potential contribution from intense flaring activities where oil and gas resources are exploited (Stohl et al., 2013). The AOD is however underestimated by the model by 25% over this latter region~~, due to the fact that the secondary organic formation is simplified in this current version of the model~~. The model also highlights a strong signal over the Yakutsk region (maximum AOD of 2.2), which is less visible on the MODIS images due to the continuous presence of clouds in July 2012. This is due to intense biomass burning plumes at this period (Ponomarev, 2013). The plumes extend towards the East and above the Bering Strait. Other significant enhancements of ~~modelled~~ modeled AOD ($0.4 - 0.5$) are also seen above northern India and above the Pacific Ocean, corresponding to the outflow area of China. The good performance to represent the main features of AOD distribution, especially in eastern Asia and western Siberia, gives a good confidence in the model to transport plumes to the Arctic. The underestimation above northern Russia indicates that the transport of plumes from that area should be considered with caution. Notwithstanding, due to the large amount of precipitation in this area (Sect. 3.5), the potential impact of this source area is reduced.

3.5 Precipitation

The lofting of BC can occur isentropically at low RH or can be caused by rapid ascent and heavy precipitation (Matsui et al., 2011). Studying precipitation during transport is therefore crucial and it needs to be modeled correctly to understand the role of wet removal of aerosols. ~~The reproducibility of precipitation in the WRF-Chem BASE simulation is assessed using daily~~ Daily GPCP (Global Precipitation Climatology Project) precipitation data are used to assess the reproducibility of precipitation

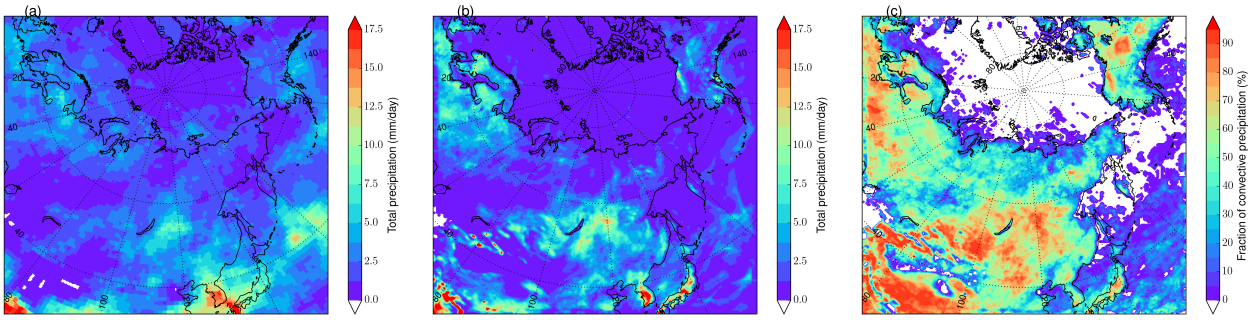

Figure 5. Average precipitation from period 4-21 July (a) obtained from 1° daily GPCP data and (b) simulated by WRF-Chem~~on the same grid~~. The fraction of convective precipitation is shown in panel (c).

in the BASE simulation. Here we use GPCP data (Version 1.2) ~~available on a latitude-longitude grid with a resolution of~~ at 1° ~~, which is based~~ resolution, retrieved from on a combination of satellite observations and rain gauge measurements (Huffman et al., 2001). Model estimates of rainfall are interpolated every day to the GPCP 1° grid and then averaged between 4 and 21 July to give an assessment of the mean precipitation during the ACCESS campaign. The WRF-Chem simulation overall reproduces the spatial distribution of the observed precipitation reasonably well (Fig. 5), with highest precipitation intensity over South-East Asia (South Korea and Japan) with about 17 $\mathrm{mm\,day^{-1}}$, high values over northern China, Mongolia and in the vicinity of Lake Baikal ($9-11\ \mathrm{mm\,day^{-1}}$) or over the Pacific Ocean due to storm tracks and associated WCBs ($10-13\ \mathrm{mm\,day^{-1}}$). Modest values are detected over Europe with $5-8\ \mathrm{mm\,day^{-1}}$. The model nevertheless overestimates the precipitation intensity over southern Russia close to Lake Baikal and over Europe (by 20%) and underestimates values over the Pacific Ocean. Over Siberia and central Arctic the precipitation intensity is low ($1-3\ \mathrm{mm\,day^{-1}}$). The highest values of rainfall are correlated with high convective precipitation fractions (Fig 5), reaching 60 to 90% over land. Over the oceans and northern Russia the main contribution is from non-convective precipitation (e.g. drizzle).

3.6 Cloud vertical distributions

Prediction of Arctic clouds (regional extent and heights) remains a challenge but is also crucial, since the vertical distribution of clouds as well as the state of cloud microphysics is important to understand how models assess the wet removal of aerosols. To evaluate the vertical distributions of clouds in our BASE simulation, we use the DARDAR-MASK v1 data set, which employs a combination of the CloudSat ~~,~~ and CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations) ~~, and MODIS~~ products (Delanoë and Hogan, 2010). To retrieve cloud phase properties, the algorithm uses the 94 GHz radar reflectivity from CloudSat, the lidar backscatter coefficient at 532 nm and vertical feature mask from CALIPSO, ~~three MODIS infrared channels 8.55 , 11 , 12 from MODIS,~~ as well as ECMWF (European Centre for Medium-Range Weather Forecasts) thermodynamic variables. A range of categories is returned from the DARDAR-MASK: clear, ground, stratospheric features, aerosols, rain, supercooled liquid water, liquid warm, mixed-phase and ice. The algorithm also includes an uncertain classifi-

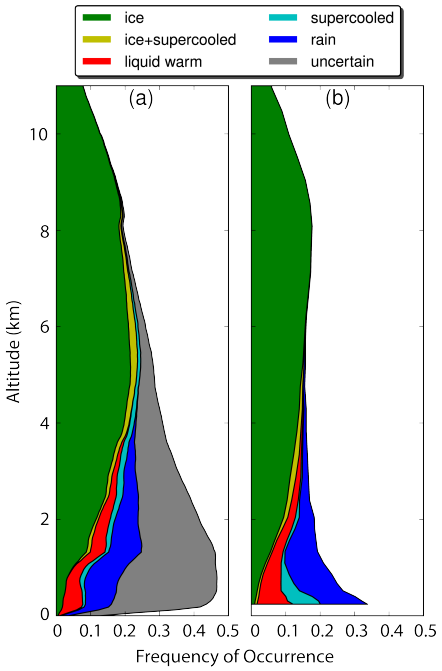

Figure 6. Mean vertical profiles of the different cloud phase categories (a) returned from the DARDAR-MASK and (b) simulated by the WRF-Chem model between 4 and 21 July.

cation, in regions where the ~~radar and lidar signal are~~ lidar signal is heavily attenuated or ~~are~~ is missing. In this study, we use a simple DARDAR-MASK simulator for WRF-Chem enabling a clear comparison between the model and satellite observations. Mass mixing ratios of liquid water, ice crystals, rain, graupel and snow, as well as the temperature, are interpolated in time and space from the model along each CALIPSO or CloudSat track passing through the WRF-Chem domain during the simulation (4-21 July 2012). The observed and simulated masks are then temporally averaged at each altitude bin for each cloud phase category: uncertain, ice, mixed-phase (ice and supercooled water), liquid warm, supercooled water and rain. The result is a mean vertical profile for each category and is represented in Fig. 6. Not surprisingly, the cloud fraction is more important in the lowest layers than ~~in altitude~~at higher altitudes, as the source of water in clouds is the evaporation from the Arctic Ocean and the humid soil. In the upper troposphere, the fraction of occurrence is 20% both in DARDAR and WRF-Chem, and clouds are composed only of ice crystals. This is directly liked to the temperature of clouds, below $-40°$C at those altitudes. The ability of the model to represent those high clouds is excellent. In the PBL, the cloud fraction is larger (between 20% and 35% if the uncertain category is not taken into account) and is dominated by liquid warm and rain. The main discrepancy between the model and the satellite observations can be ascribed to the uncertain cloud type. When the cloud type is classified as uncertain because the lidar signal is extinguished or too attenuated below optically thick clouds, the model generally predicts no cloud. The model slightly overestimates the fraction of clouds containing supercooled water in the PBL (by 2%) whereas this fraction is underestimated in the mid-troposphere. The model also slightly underestimates the fraction of ice and supercooled droplets

in the upper troposphere. If we do not consider the uncertain class, the result of the WRF-Chem model shows appreciable skill in capturing the average vertical distribution of cloud phases. In particular, the vertical distribution of the liquid warm and rain categories is ~~very~~ well reproduced. This is very promising in simulating the wet deposition efficiency of aerosols in plumes transported to the Arctic region.

4 Source attribution of BC particles

~~The major objective of this section is to understand the vertical transport mechanisms of BC particles over Siberia and their transport pathways towards the Arctic in summer using the results from WRF-Chem calculations. First, we identify the major uplifting processes for BC particles and the major transport pathways for the export of BC from Siberia. In particular, we conduct analyses of the mass flux of BC and meteorological fields that play an important role in the transport and wet removal of BC. Second, we quantify the contributions of BC emitted from various Russian, Asian, and European regions to the outflow towards the Arctic.~~

4.1 Export processes

To identify the sources and mechanisms of pollution BC export to the Arctic~~and the mechanisms involved in the mean upward motion~~, we compute the horizontal poleward eddy heat flux at the 850 hPa level using the model (Fig. 7). Regions with large values of the horizontal poleward eddy heat flux generally illustrate the activity of migratory cyclones and may reflect strongly ascending moist airstreams (WCBs) in extratropical cyclones that rise from the PBL to the free troposphere (FT) on short time-scales (1 or 2 days) (Eckhardt et al., 2004). The horizontal poleward eddy heat flux is calculated as $\overline{v'\theta'}$, where the overbar denotes time-averaging over the ACCESS period (4-21 July 2012). Because the time scale of the migratory cyclones is about one week, v' and θ' are the instantaneous deviations from the 5-days running means of the meridian wind component and the potential temperature, respectively. This eddy heat flux is computed at each grid point of the WRF-Chem simulation. ~~Four regions with large~~ Large values of eddy heat transport ~~can be identified during the ACCESS period. The values were large in northern central Europe, particularly Germany and Poland ($50-63°$N, $10-40°$E),~~ are found over the West Siberian Plain ($60-70°$N, $40-80°$E) extending above the Barents Sea, over the Sakha (Yakutia) Republic ($65-70°$N, $110-150°$E) and finally over the Pacific storm track around northern Japan. ~~Regions with large values of the horizontal poleward eddy heat flux generally illustrate the activity of migratory cyclones and may reflect strongly ascending moist airstreams (WCBs) in extratropical cyclones that rise from the PBL to the free troposphere (FT) on short time-scales (1 or 2 days) (Eckhardt et al., 2004). If these regions characterized by large poleward eddy flux~~ If those regions are located in the vicinity of BC sources, they are likely to contribute to export pollution to the Norwegian Arctic, including areas sampled by the Falcon.

In Fig. 7, the ~~spatial~~ horizontal distribution of the mean upward BC mass fluxes averaged over the ACCESS period ($\overline{[BC]w}$, where w denotes the vertical velocity) is also represented at ~~the same altitude~~700 hPa. Three regions with strong upward BC

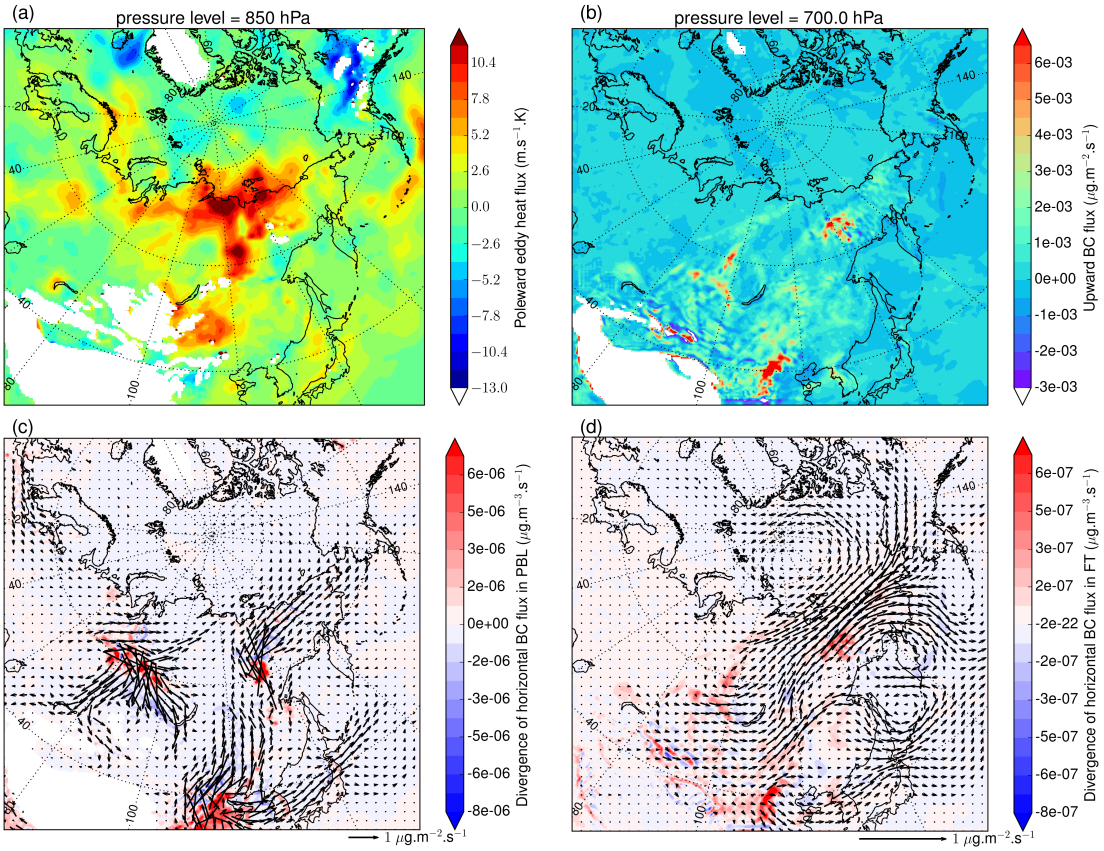

Figure 7. Time-averaged (a) horizontal poleward eddy heat flux $\overline{v'\theta'}$ at the 850 hPa level, (b) upward mass flux of BC $\overline{[BC]w}$ at the ~~850~~ 700 hPa level, and divergence of the horizontal mass flux of BC integrated over (c) the PBL and (d) the FT during the ACCESS period in the WRF-Chem BASE simulation. Regions ~~in (b) and (c)~~ without data in white ~~correspond to regions with high-altitude mountains~~ have a high orography (surface pressure below ~~850 hPa~~the plotted level). Vectors representing the time-averaged horizontal mass flux of BC integrated over the PBL and FT during the ACCESS period are shown in panels (c) and (d) respectively, as well as the scaling near the lower right corner.

fluxes can be identified: central and northeastern China ($30-45$°N, $110-120$°E), south of the Krasnoyarsk region ($50-60$°N, $80-100$°E) and Yakutia ($60-68$°N, $125-150$°E). Those regions characterized by a strong ascent of BC mass fluxes are also co-located with areas presenting large values for the ~~divergence~~ convergence of horizontal BC flux in the PBL, defined here as the $700-1000$ hPa layer and large values for the divergence of horizontal BC flux in the FT, defined here as the $700-200$ hPa layer (Fig. 7). ~~The strong divergence regions of the horizontal BC fluxes within the PBL generally correspond to the large BC source regions~~ They illustrate the locations of source regions for BC emissions and the divergences represent the horizontal transport of BC emitted from those source regions to the surrounding regions (Oshima et al., 2013). Convergence of BC flux in the lowest layers can indeed uplift air parcels from the surface to the FT, which are then exported to outflow regions.

A good spatial correlation is also observed with the spatial distribution of AOD (Fig. 4) illustrating that the main emission areas present a significant fraction of carbonaceous aerosols. BC emitted from central and northeastern China is mostly of anthropogenic origin, whereas the two other regions (south of the Krasnoyarsk region and Yakutia) are co-located to intense biomass burning sources, explaining the strong values of the upward BC flux. Aerosols and CO emitted from those source regions may potentially be exported towards the Arctic through the areas presenting large horizontal poleward eddy heat flux: the Sakha (Yakutia) Republic ($65-70°$N, $110-150°$E) and the Pacific storm track. ~~It is confirmed with the time-averaged horizontal mass flux of BC integrated over the FT during the ACCESS period (Fig. 7).~~ We also note that the source of BC identified in western Siberia is also partly located in the region where the Russian oil and gas flaring emissions are very high and along the main low-level pathway of air masses entering the Arctic (Stohl et al., 2013). They may be exported towards the Arctic through the region with high value of $\overline{v'\theta'}$, extending above the Barents Sea. Anthropogenic emissions in Europe may also be exported to the Arctic (large values of $\overline{v'\theta'}$ in northern Norway) but to a lower extent since the corresponding horizontal and vertical mass fluxes of BC are low in summer.

4.2 Plume identification

As indicated in Sect. 2.1, the two flights of 17 July (namely flight 17a in the morning and flight 17b in the afternoon) were specifically dedicated to plumes transported in ~~altitude~~ the upper troposphere from boreal and Asian sources to the Arctic region. Data obtained during these flights is thus the most relevant for the study of BC transport to the Arctic. ~~Figure 8 shows the~~ The vertical cross-sections of CO ~~and BC extracted~~ and BC interpolated from the WRF-Chem ~~model on~~ BASE simulation onto the two flight tracks of 17 July ~~.~~ are shown in Fig. 8. In situ measurements of these variables are also ~~shown in Fig. 8. The predicted time-altitude cross-sections along the selected flight tracks help in distinguishing the~~ represented. The horizontal and vertical ~~variability~~ variabilities of the CO and BC enhancements ~~due~~ ascribed to different source regions can be distinguished from the modeled time-altitude cross-sections along the selected flight tracks. The Falcon transected pollution plumes located between Andøya (69.1°N, 15.7°E) and Spitsbergen (78.9°N, 18.0°E) during flight 17a and ~~Spitsbergen during the first flight and~~ sampled the same plumes again during ~~the second flight~~ flight 17b before returning to Andøya. The model simulates various CO enhancements during each flight period. Increases in modeled BC mixing ratios are ~~very~~ well co-located with those CO enhancements. The PV (potential vorticity) vertical cross-section suggests that all those enhancements are in the troposphere: at the time of the flights, the stratospheric air masses are confined in regions with potential temperature larger than 310 K, with a fold between 09:30 and 13:30 UTC, bringing stratospheric intrusions down to 310 K. In the upper troposphere, the model predicts three periods of enhanced CO and BC during each flight, near the 310 K isentrope (altitude between 6 and 8 km). In this altitude range, when the Falcon crosses the plumes predicted by the model, the instruments also detect higher concentrations of CO and BC. WRF-Chem however underestimates the magnitude of the mixing ratios of carbon monoxide in the peaks, reaching 210 ppbv. This is mostly due to numerical diffusion ~~in the model, but also~~ 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 6^{th}-order numerical diffusion is used in our simulations. The CO underestimation can be also due to the fact that the Falcon sampled the southern edge of a larger and more intense plume located farther north

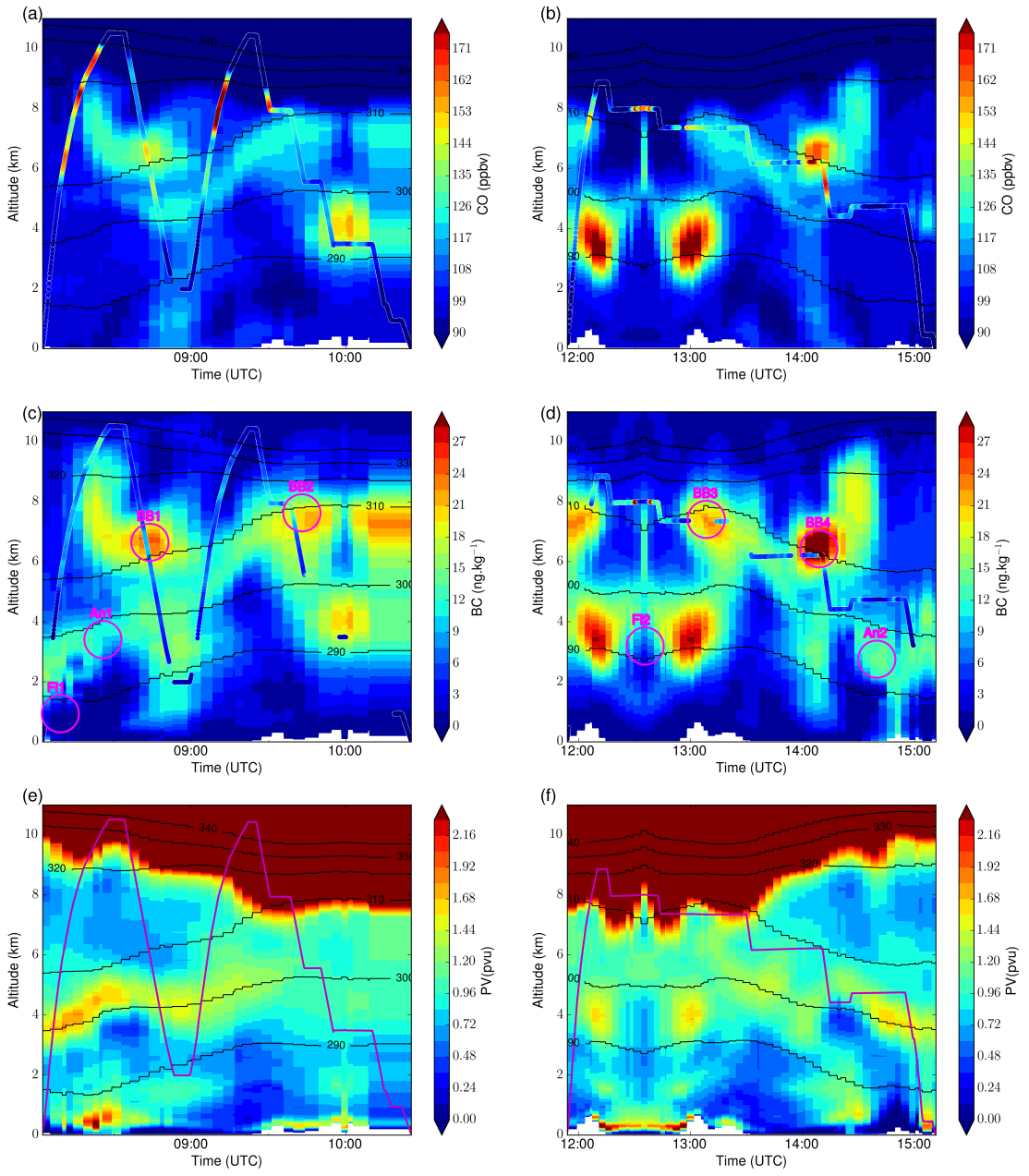

Figure 8. Vertical cross-sections of (a) and (b) CO mixing ratio, (c) and (d) BC mixing ratio, (e) and (f) PV along Falcon flight tracks on 17 July 2012 extracted from WRF-Chem between Andøya island and Svalbard archipelago. In situ measurements aboard the Falcon averaged using a 2-min running mean are shown in each panel with colored dots, using the same color scale. Black solid lines represent the dry isentropes between 290 K and 340 K. Magenta circles highlight the air masses discussed in Sect. 4.4.

before returning to Andenes (Roiger et al., 2015). The underestimation of CO can also partly be ascribed to too low emissions or problems in modeling OH. This plume is shifted ~~a bit~~ towards the north by 40 km in the model simulation, explaining the strong underestimation of the model at 09:15 UTC. In terms of BC, the agreement between WRF-Chem and the SP2 instrument is ~~very good~~reasonable. Inside the plumes, values of $20-25$ ng kg^{-1} are reported, with peak values larger than 30 ng kg^{-1}. In the mid-troposphere, CO and BC concentrations are also significantly increased sporadically in the band delimited by the 290 and 300 K isentropes (between 2 and 4 km) with values reaching 180 ppbv and 30 ng kg^{-1}, respectively.

4.3 Contribution from the different sources

To understand the contributions of the different source emissions (anthropogenic, biomass burning and flaring) to the BC transported to the region of the flights, we use the difference between the BASE run and the NoAnthro, NoFire, NoFlr simulations. Model results from each run are interpolated in time and space to the position of the Falcon during the two flights on 17 July. The relative contribution of each source to total BC is obtained by dividing by the results derived from the BASE simulation. Figure 9 shows the time-altitude cross-sections of the relative contributions of BC concentrations due to the different emission sources. The two vertical cross-sections clearly underline that the mass mixing ratio of BC is strongly dominated by the fire contribution, which is generally larger than 80% at all altitudes and times, and often larger than 90%. We note two exceptions to the dominance of fires. In the vicinity of the Norwegian coast, i.e. just after take off from Andenes before 09:00 and on the way back to Norway after 14:00 UTC, a strong influence of anthropogenic sources (about 85%) is clearly identified between 2 and 4 km, and to a lesser extent in the PBL, where a weaker contribution of Norwegian flares is predicted ($40-70$%). A second zone where the influence of fires is low (below 15%) is detected in the mid-troposphere ($2-5$ km) between 12:30 and 13:00 UTC. In this region, the contribution of flaring emissions from Siberian oil exploration is dominant, about 80%.

In Sect. 4.2, we identified significant enhancements in CO and BC between 6 and 8 km. According to Fig. 9, these plumes originate from large biomass burning sources ~~. Heating of large Siberian fires can inject CO and BC into the free troposphere. The online plume-rise injection model in~~ in Siberia. In WRF-Chem~~is used to predict fire emission injection heights in our simulations (Grell et al., 2011). The convective motions that occur with such wild fires increase the likelihood that emissions will~~ , the fire emission heights are determined by the online plume rise injection model (Grell et al., 2011). These emissions may then be lofted to the faster winds of the free atmosphere. We should note that in this altitude range ($5-10$ km), the contribution of anthropogenic sources is weak (maximum 15%) but not zero. There is likely a small influence of the vertical transport by continental convective lifting over Asia in summer and frontal lifting ~~associated with cyclonesin the mid-latitudes~~due to mid-latitude cyclones. The transport pathways from the different source regions to the Arctic is discussed in Sect. 4.4. In the mid-troposphere ($2-4$ km), the CO and BC concentrations have also increased due to the influence of fires, except in the vicinity of the northern Norwegian coast, where the impact of anthropogenic sources is larger. The flaring emissions play a very localized role in the area of the flights and ~~are not due~~ do not lead to any noticeable increase in CO or BC. ~~This suggests that a large fraction of the aerosols emitted from flares has been removed by precipitation during transport, which is investigated in~~

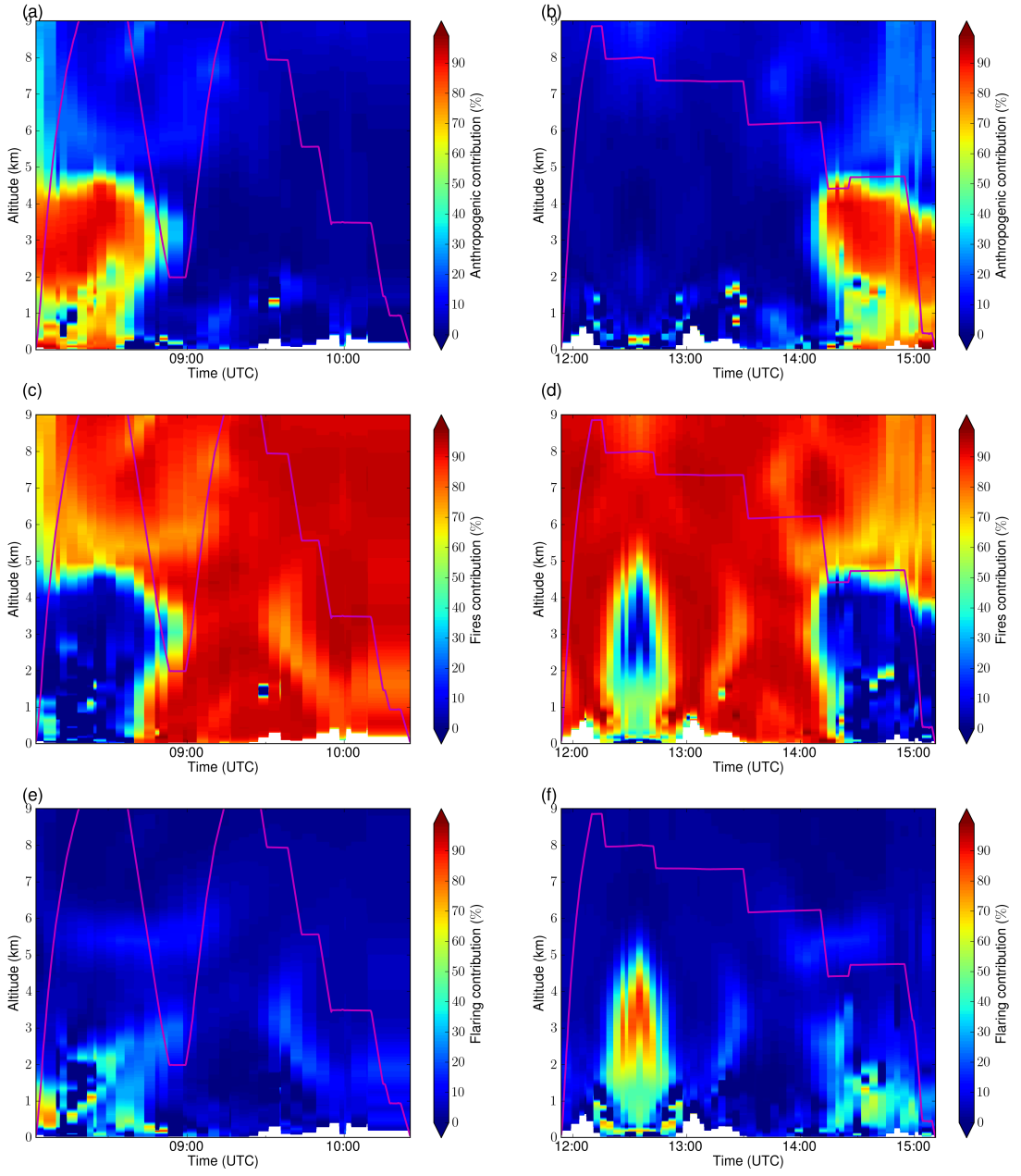

Figure 9. Time-altitude cross-sections of the relative contributions (%) of BC concentrations due to (a) and (b) anthropogenic, (c) and (d) biomass burning and (e) and (f) flaring emissions along flight tracks 17a (in panels (a), (c) and (e)) and 17b (in panels (b), (d) and (f)). The altitude of the two flights is indicated in magenta in each panel.

~~more detail in Sect. 5.~~

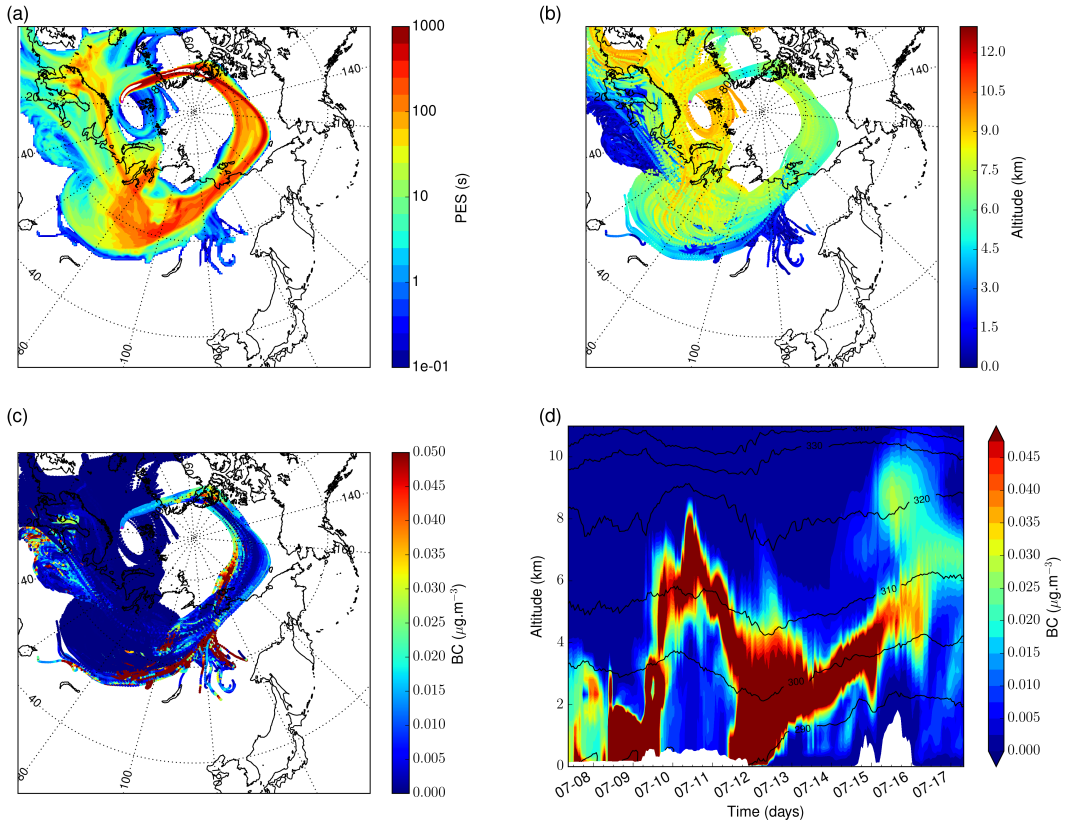

Figure 10. FLEXPART-WRF backward simulation from the biomass burning plume observed by the Falcon aircraft at 73.1°N, 18.2°E and 6.5 km between 08:37 and 08:47 UTC. (a) ~~:~~Column-integrated PES, (b) altitude and (c) BC mass concentration interpolated along the 10000 trajectories for 10 days prior to the release. In panel (d), the vertical cross-section of BC interpolated along the plume centroid location is shown, together with the dry isentropes (black lines) between 290 K and 340 K.

4.4 Transport pathways

~~We use FLEXPART-WRF in backward mode to study~~ To investigate the origin and transport pathways of ~~plumes and to~~ air masses and provide insight into the WRF-Chem representation of BC, the FLEXPART-WRF model is run in backward mode. We identify four plumes originating from boreal fires on 17 July from Fig. 8 in the upper troposphere between 6 and 8 km and confirm them by in situ measurements from the aircraft. The center of those plumes has been found at 08:42 (BB1), 09:39 (BB2), 13:06 (BB3), 14:10 (BB4) UTC. Their dominant biomass burning origin has been suggested by Fig. 9. We also consider two ~~plumes predicted~~ air masses predicted by the model to be influenced by flaring emissions, in the PBL (0.7 km) at 08:05 UTC (Fl1) and in the mid-troposphere (3.7 km) at 12:35 UTC (Fl2). Finally two anthropogenic ~~plumes~~ air masses have been selected in the mid-troposphere (3.2 – 3.5 km), in the proximity of Andøya island at 08:30 (An1) and 14:45 (An2) UTC. For

each selected ~~plume~~air mass, 10000 FLEXPART-WRF particles are released in a volume 40 km $\times$40 km (horizontally) and 1 km (vertically). Since transport ~~times are typically less~~ time scales are typically lower than 10 days, each ~~of the simulations~~ simulation is run backwards for 10 days to track the ~~air mass origin over the source regions of interest. Specifically, we use FLEXPART-WRF PES to study source-receptor relationships for~~ origin of air masses reported in Fig. ~~9~~8.

Figure 10 shows the $0-20$ km column of FLEXPART-WRF PES integrated for 10 days for the first fire plume (released at 08:42 UTC on 17 July), together with the altitude and BC concentration retrieved from the model and interpolated along each of the 10000 trajectories. A vertical cross-section of BC interpolated along the plume centroid location is also shown. We note that the results are very similar for the four backward simulations in biomass burning plumes (not shown). Figure 10 highlights a cross-polar transport of biomass burning emissions from Siberian fires into the Arctic~~. They were~~ , which reach the Svalbard archipelago thanks to a pronounced northerly flow. These plumes have been probed in detail by the ~~Falcon~~ aircraft on the two flights on 17 July~~thanks to a pronounced northerly flow over the Svalbard archipelago~~. During the ACCESS campaign in July 2012, this cross-polar transport of biomass burning pollution ~~across the Pole~~ was caused by Arctic low-pressure systems, regular phenomenons of the ~~polar circulation especially during summer~~ summertime Arctic circulation (Orsolini and Sorteberg, 2009). WRF-Chem meteorological analyses suggest that these Arctic transport events in the upper troposphere are due to WCBs ~~associated with~~ linked to two wave cyclones over extreme northern Russia and eastern Siberia. The first one was present at 72°N, 80°E between 11 and 12 July and reached 970 hPa. The second cyclone, responsible of the plume curl over North Pole (Fig. 10), moved northward and extended from 80°N, 140°E to 88°N, 180°E, progressively deepening to reach a minimum surface pressure of 975 hPa.

Two large plumes enriched in CO and BC form over the south of the Krasnoyarsk region on 8-9 July and over Yakutia on 11-12 July ~~(Fig. 7). Fire injection over those boreal sources facilitates a rapid uplift up to an altitude of 6 . These plumes~~ and merge over eastern Siberia into a large plume advected to higher latitudes. When the first low-pressure system forms at the surface, the plume intrudes into the Arctic atmosphere, where it elongates and becomes narrower (12 July). The second low-pressure system forms at 80°N, 140°E, which facilitates the progression of the eastern part of the plume further North ~~and in altitude~~aloft. Due to the progression of the cyclone to the North-East (88°N, 180°E), the plume elongates and moves almost over the North Pole (13 July). Along the plume boundaries, some mixing processes with the surrounding air occur, partly eroding its outer parts and leading to a filamentary structure. The ageing plume is split into two branches reaching northern Canada on 14 July and moving towards Svalbard. The relatively fine resolution of our simulations (40 km) can resolve the increased filamentation of the pollution plumes, an advantage over most global models with coarser grids (Sodemann et al., 2011; Ma et al., 2014). The concentrations of CO and BC interpolated along trajectories generally decrease with time due to mixing with the surrounding clean atmosphere and deposition processes along transport (Fig. 10), except on 14-15 July when two plume branches of the same initial plume merge together.

Figure 9 also demonstrates a small contribution (15%) of anthropogenic sources to the enhanced CO and BC detected by the Falcon observations. As suggested by Fig. 10, the anthropogenic emissions that arrive near Norway may originate from fossil-fuel burning in northern China where high levels of BC have modeled ~~observed~~ 10 days before the flights on 17 July (Fig. 7). These emissions undergo a strong and efficient uplift from the PBL to the middle and upper troposphere within WCBs and therefore enter the Arctic at high altitudes. Those Asian anthropogenic air masses are characterized by high water vapor mixing ratios ($14\ \mathrm{g\,kg^{-1}}$) and RH values larger than 80%, facilitating their uplift over cold, dense Arctic air masses. The fact that the contribution of fossil-fuel emissions is found to be weak over Scandinavia suggests that significant deposition of aerosols occurs during transport. Wet removal is due to cloud formation and a high amount of precipitation during uplift, mostly convective rain (Fig. 5). It is also ~~accompanied by an increase~~ characterized by an enhancement of potential temperature from 300 K to 315 K ~~due to~~ caused by latent heat release from water vapor condensation.

According to Serreze and Barrett (2008) and Orsolini and Sorteberg (2009), such pollution transport events characterized by the presence of low-pressure systems in summer could be common and effective transport mechanism to the Arctic atmosphere for Siberian forest fires and Asian industrial emissions. Such transport pathways reaching the Arctic have been observed by Sodemann et al. (2011) and Roiger et al. (2011) during July 2008. In our study, the low-pressure systems are more intense (surface pressures ranging from 970 to 975 hPa), suggesting an effective mechanism for intrusion of pollutants into northern Norway.

The pollution plumes exported from the flaring emission sources follow different pathways as a function of their altitude. The plume reaching Scandinavia in the PBL is exported above the West Siberian Plain where large values of the poleward eddy heat flux ~~is~~ are predicted (Fig. 7) and passes over the Barents Sea before progressing poleward at low level. On the contrary, another plume emitted from the flares in western Siberia is transported eastward along the Siberian coast, moves to the North over Yakutia, curls over North Pole before reaching Svalbard in the mid-troposphere. Finally, the two anthropogenic plumes identified in the mid-troposphere close to Andøya island are exported from Europe in about 6 days.

5 Deposition during transport

5.1 Contribution of deposition processes

To understand the contributions of the different deposition processes (dry deposition, wet removal in grid-resolved clouds and in parameterized convective clouds) on the mass of BC transported to the region of the flights, we use the normalized differences between the ~~NoDry, NoWet, NoWetCu~~ NoX simulations and the BASE run, where NoX represents the NoDry, NoWet or NoWetCu simulations : $100 \times \left(\frac{\mathrm{NoX} - \mathrm{BASE}}{\mathrm{NoX}}\right)$. Model results from those four runs are interpolated in time and latitude and longitude at the position of the Falcon during the two flights on 17 July. Figure 11 shows the time-altitude cross-sections

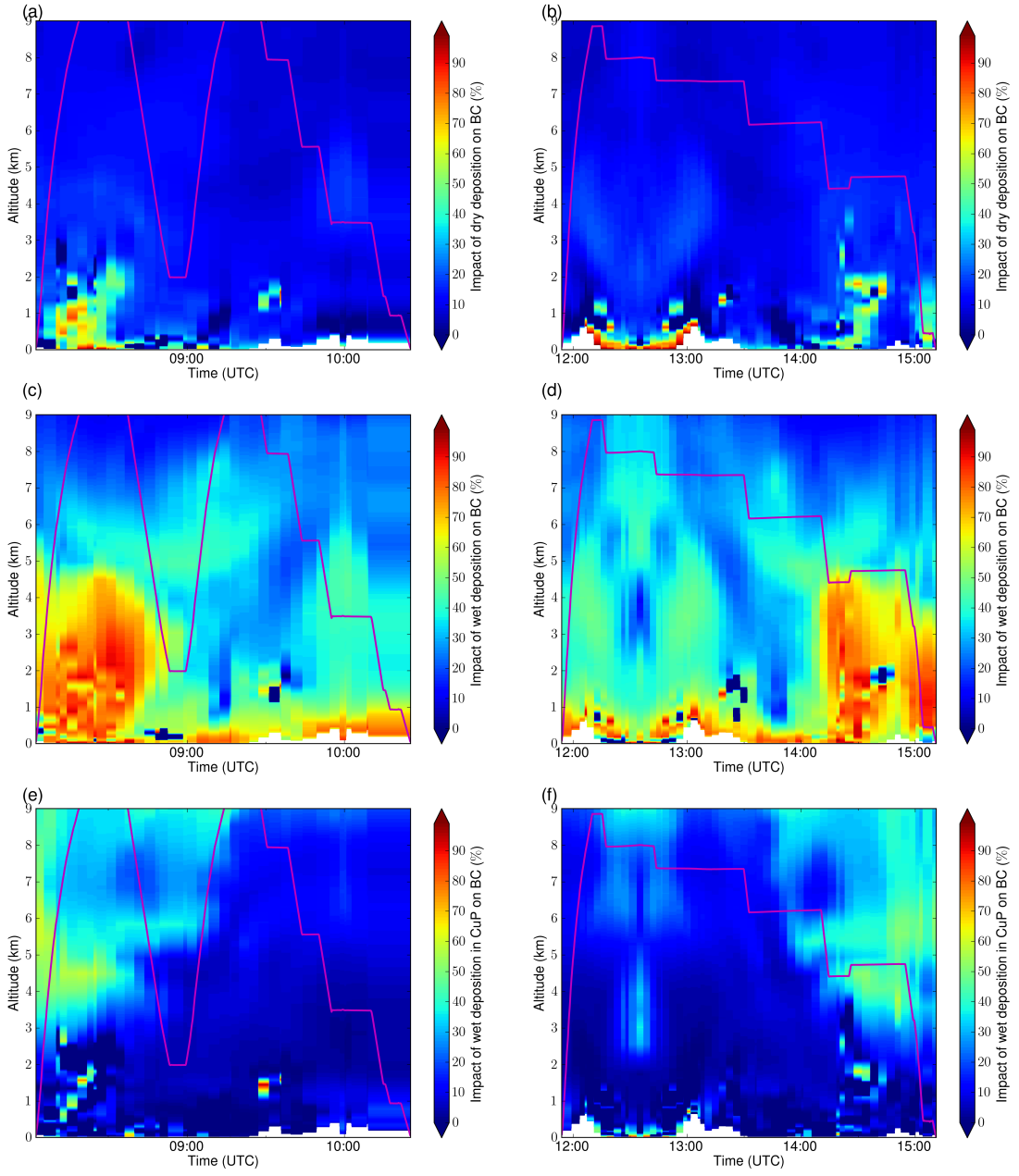

Figure 11. Time-altitude cross-sections of the relative contributions (%) of (a) and (b) dry deposition, (c) and (d) wet deposition in grid-resolved clouds and (e) and (f) wet deposition in parameterized cumulus clouds to the BC mass mixing ratio along flight tracks 17a (in panels (a), (c) and (e)) and 17b (in panels (b), (d) and (f)). The altitude of the two flights is indicated in magenta in each panel.

of the relative contributions of the different deposition processes.

The dry deposition is only significant (more than 50%) in the lowest layers, and close to the Norwegian coast. Due to the fact that BC particles predominantly occupy the Aitken and accumulation mode (90% of BC particles have a diameter lower than $2.5\ \mu\mathrm{m}$ in our simulation), the impact of the sedimentation process is very weak. Dry deposition of BC-containing particles therefore arise from the aerodynamic transport down through the atmospheric layer to a very thin layer of stagnant air just adjacent to the surface (by turbulent diffusion) and from the Brownian transport across this quasi-laminar sublayer to the surface itself. As a consequence, the role of dry deposition is limited to the lower troposphere (PBL). More important are the impacts of wet deposition processes in grid-scale and subgrid parameterized clouds. Grid-scale wet removal has the largest effect (80% BC removal) in the mid-troposphere, where European anthropogenic influence is also large (Fig. 9). Altitudes influenced by biomass burning plumes (Fig. 9) experience similar BC removal rates, $40-60\%$, from both grid scale and subgrid scale clouds. In the areas where the biomass burning plumes were more intense, as identified in Fig. 4.2, the impact of the deposition processes is generally smaller, almost negligible for dry deposition, about 30% for wet removal in grid scale clouds and 10% in ~~KFCuP~~convective clouds. The fact that the maximum in BC concentrations correspond to zones less affected by deposition processes illustrates the heterogeneous vertical cross-sections of BC (Fig. 8). The vertical profile of BC (Fig. 3) would have therefore peaked more intensively in the mid- and upper troposphere if wet removal had been less efficient. Between 4 and 6 km, the BC mixing ratio would have been multiplied by a factor of 3 without wet removal in subgrid convective clouds, or by a factor of 4 without wet removal in grid scale clouds. ~~An understanding of the wet removal of BC is critically important because it directly controls vertical profiles of BC and amounts of BC transported from source regions to the Arctic.~~

BC particles coated with ~~sufficient~~ non-refractory and water-soluble secondary compounds ~~are~~ can be CCN (cloud condensation nuclei) active in liquid clouds. BC-containing particles can also act as IN (ice nuclei). In the MOSAIC aerosol module used in WRF-Chem, aerosol components are assumed to undergo a instantaneous internal mixing in each size bin. Petters et al. (2009) showed that internally mixed BC particles can be more hygroscopic and easily removed by wet scavenging compared to externally mixed BC. This should not have a large effect on our results as BC particles sampled by the Falcon have principally been transported in biomass burning plumes from Siberia (Sect. 4.4). Kondo et al. (2011) and Sahu et al. (2012) observed that ~~the coating is often thicker~~ BC in biomass burning plumes is often more internally mixed than in fossil fuel emissions, and is thought to occur in the first few hours after emission (Abel et al., 2003; Akagi et al., 2012).

During the long-range transport of plumes towards the Arctic region, the WCBs observed in this study loft aerosols from the PBL to the free troposphere, or rapidly inject them into the upper troposphere, in the warm section of mid-latitude cyclones. Clouds form and a ~~large fraction of aerosols~~ significant portion of particles becoming efficient CCN is scavenged by cloud droplets or rain drops and potentially removed from the atmosphere via wet deposition in those vertical transport pathways. A fraction of aerosols may survive this deposition associated with the cyclones~~. Aerosols that survive these vertical transport pathways to higher altitudes can~~ , reach higher altitudes and travel to polar latitudes. But a small ~~fraction~~ portion of the remaining interstitial BC particles can be removed through impaction scavenging by collection of cloud or rain droplets. This process is said to be more effective for BC removal, both in mixed-phase (Twohy et al., 2010) and in ice clouds (Baumgardner et al.,

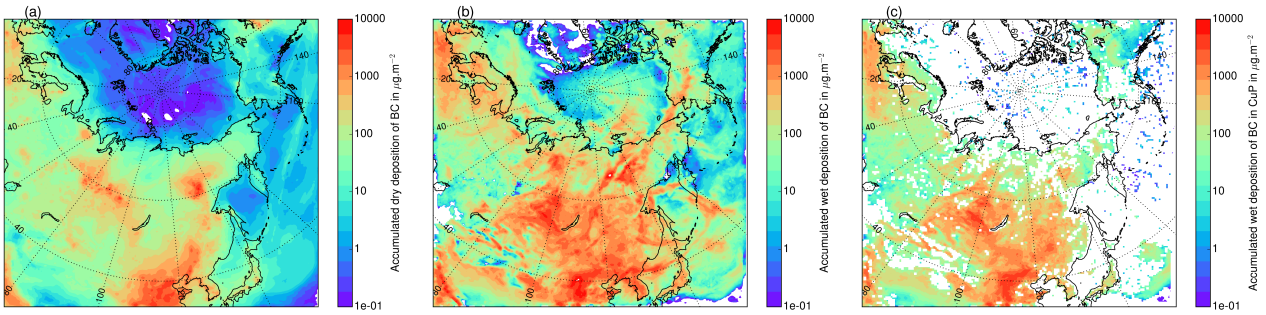

Figure 12. Accumulated deposition of BC due the (a) dry deposition, (b) wet removal in all clouds (grid-resolved and cumuli) and (c) wet removal in parameterized cumulus clouds between 7 and 21 July. Note the logarithmic color scale.

2008; Stith et al., 2011). This is particularly important in this study as Fig. 6 indicated that ice, supercooled and mixed-phase clouds are predominant in the mid-troposphere during the ACCESS period.

5.2 Impact of deposition

Fig. 12 shows the accumulated deposition of BC between the 7 and the 21 July, which is the period during which aerosols were transported from mid-latitudes to the Scandinavia. The three types of deposition processes are studied here: dry deposition, wet removal by large-scale precipitation (first-order rainout and washout) and scavenging in wet convective updrafts~~(KFCuP)~~. The deposition of BC is clearly dominated by wet removal from the middle to high latitudes. The impact of dry deposition can only been observed near emission sources: the spatial distribution of accumulated BC by dry deposition is co-located with the map of the upward BC flux and the patterns of the divergence of horizontal BC flux in ~~PBL~~ FT (Fig.7). It is therefore similar to the spatial distribution of emissions. Sedimentation and turbulent mix-out are indeed the ~~dominant sink of large~~ major sink of coarse particles containing BC near emission sources. The dry deposition flux of BC particles from the troposphere to the surface is moreover almost proportional to the local BC mass mixing ratio near the surface. In large scale clouds, BC particles are removed through the nucleation scavenging mechanism~~or through~~ , or through wet deposition processes (in-cloud scavenging called rainout and below-cloud scavenging referred to as washout), where particles are collected by falling hydrometeors (in stratiform precipitation) through Brownian motion, electrostatic forces, collision or impaction (Seinfeld and Pandis, 2006). Interstitial and cloud-borne BC-containing particles can finally be removed in the strong updraft core of deep convective clouds parameterized in KFCuP (Kain-Fritsch-CUmulus Parameterization).

The horizontal distribution of BC wet removal results from a combined effect of the precipitation amounts (Fig. 5) and the upward BC flux (Fig. 7). During the vertical transport of plumes containing carbonaceous aerosols, the wet deposition is indeed directly linked to the distribution of precipitation. The spatial distribution of BC scavenged by wet deposition in parameterized cumulus clouds matches well that of convective precipitation (Fig. 5). Aerosol wet removal from predicted deep and shallow

convective clouds is however lower than the removal from grid scale clouds. This is the case even where the cumulus scheme predicts greater amounts of precipitation than the microphysical scheme (Fig. 5). ~~KFCuP~~ This KFCuP scheme triggers almost everywhere in the mid-latitudes, but not so much over the oceans and in the Arctic or sub-Arctic (beyond $60°$N) where it is mostly dominated by the formation of low-level stratocumulus cloud decks in the boundary layer and lower troposphere, producing frequent drizzle (Browse et al., 2012). We have to note that the KFCuP ~~mechanisms~~ mechanism actually lacks below-cloud wet scavenging and scavenging by snow and ice. Adding the below-cloud scavenging by the convective precipitation would nevertheless have only a minor impact on the BC concentrations: assuming that nearly all the BC was in the $80-470$ nm diameter range, the below-cloud removal efficiencies are indeed very small for large rain drops below cumuli. Some of the enhanced wet removal in cumuli must be compensated by the enhanced vertical transport of aerosols from the PBL into the free troposphere during deep convection (Berg et al., 2015). It is also important to bear in mind that the cumulus cloud fraction within the model grid box (here 40 km) can be quite small, so that the wet removal occurs over a relatively small area and does not have a large impact on the total aerosol loading within the model grid box. While validating the KFCuP cumulus scheme, Berg et al. (2015) highlighted a reduction of low-altitude aerosol associated with the venting of aerosol from the PBL as well as changes in the vertical structure associated with the cumulus induced subsidence, but little change in the average aerosol aloft. This may be also consistent with the small amount of secondary activation in the simulations.

~~The fact that wet deposition is predominant~~ During the summer of 2012, the wet removal is the dominant process at all latitudes~~during summer 2012 suggests that a large portion of BC~~ , which illustrates that a substantial fraction of BC mass concentrations is removed in the regions south of $70°$N before reaching the Arctic. ~~Matsui et al. (2011) and Sharma et al. (2013) confirmed that the BC emissions in the East-Asia region ($30-40°$N) are subject to more efficient scavenging (90% wet deposition) than other temperate source regions, meaning the East-Asian influence on the Arctic is significantly inhibited compared to other regions. Due to stronger scavenging in East-Asia, this source region has less influence on the Arctic compared to biomass burning sources in Russia (Sect. 4.4), where precipitation rates are lower.~~ The influence of wet removal of BC scavenging is also predicted in the high latitudes in summer, underlining the role of drizzle from low-level clouds and fogs. ~~This scavenging at low level inside~~ The combination of low-level scavenging in the Arctic region ~~associated with a decrease in transport from outside the Arctic are responsible from~~ and transport decrease from mid-latitudes is the cause of the low summertime BC concentrations. We note the fact that the WRF-Chem performs worse at 100 km to reproduce the mean BC profile observed by the Falcon particularly in the mid-troposphere (Fig. 3) suggests that the coarse resolution and the increased role of ~~the KFCuP mechanism~~ convective clouds result in less efficient BC removal during transport. This is due to the presence of weakly precipitating shallow clouds at high latitudes, that do not scavenge aerosols adequately.

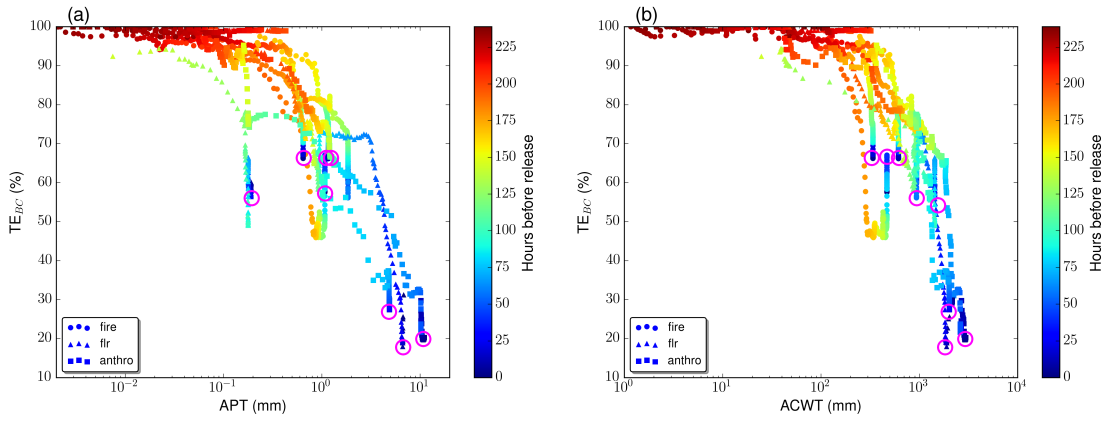

Figure 13. Transport efficiency of BC (TE_{BC}) for eight plumes identified from sampled air parcels by the Falcon (4 boreal fire plumes, 2 plumes from flaring activities in Northern Russia and 2 anthropogenic plumes from Europe) as a function of (a) the accumulated precipitation along trajectory (APT) and (b) the accumulated condensed water along trajectory (ACWT). The ~~parameter is~~ points are colored by the time (in hours) before the release of the trajectories in FLEXPART-WRF. Magenta circles emphasis the starting points (~~color scale~~release time = 0) of the eight air masses discussed in Sect. 4.4.

5.3 Transport efficiency of BC particles

5.3.1 Role of accumulated precipitation and condensed water along FLEXPART-WRF trajectories

To investigate the transport efficiency of BC particles ~~, we performed a simulation that did not consider aerosol wet deposition both in grid scale and in parameterized convective clouds (NoWetAll run), and the results of this simulation are compared with those of the BASE simulation to~~ and evaluate the effects of the wet removal of BC from the atmosphere by precipitation during transport~~. We use CO as an inert combustion tracer (lifetime of a few weeks) that is not generally impacted by precipitation during transport from Siberian and East Asia~~, we use the NoWetAll simulation, which does not consider aerosol wet deposition both in grid scale and in parameterized convective clouds. We define the transport efficiency of BC as a function of altitude z as:

$$\mathrm{TE_{BC}}(z) = \frac{[BC](z)_{\mathrm{BASE}}}{[BC](z)_{\mathrm{NoWetAll}}} \quad (1)$$

~~where $\Delta[BC]$ and $\Delta[CO]$ represent the differences between simulated and background values of BC mass mixing ratios, respectively.~~ Similar quantities have been used in previous studies (e.g., Koike et al., 2003; Matsui et al., 2011; Oshima et al., 2012), but the denominator was often chosen as the observed BC-to-CO ratios for the source regions rather than its value extracted from a simulation where wet removal has been turned off, as in Oshima et al. (2013).~~In the latter study however, the modeled TE_{BC} values did not take into account the CO concentrations. Our method has the advantage that the depletion of the BC-to-CO ratios is only due to wet removal (both in grid scale and parameterized clouds), and not to the fact that some plumes emitted from the source regions do not finally reach the receptor area (Scandinavia) due to different transport patterns~~

~~or diffusion during transport. The background value of CO has been determined at each height by the fifth percentile of all CO measurements (Fig. 3) in an altitude range 500 -thick centered around that height. The background value of BC has been assumed to be zero, due to its short lifetime (less than one week).~~

~~To evaluate the aerosol wet removal processes used in the model in more detail, we examined the dependence of TE_{BC} on the amount of precipitation that an air parcel had experienced during transport.~~ Following the methodology of Oshima et al. (2012) who investigated the role of wet removal of BC in Asian outflow towards the Pacific Ocean, we estimate the accumulated precipitation along trajectory (APT) and use it as a proxy for wet deposition. APT represents the amount of precipitation that an air parcel had experienced during transport. For every sampled air parcel by the Falcon and identified as a plume on 17 July in Sect. 4.4, the TE_{BC} and APT values are computed along each of the 10000 FLEXPART-WRF trajectories released at the time and location of the plume. Specifically, the APT is derived by integrating the WRF-Chem precipitation amounts from the BASE run along each Lagrangian 10 days backward trajectory of the corresponding uplifted air parcel. For the plume originating from Russian flaring sources (rapidly transported at a low altitude (Sect. 4.4)), the trajectory runs backwards for 6 days. At hourly intervals, the rain, ice, snow and graupel precipitation rates (including values in cumuli) are interpolated at the closest WRF-Chem grid box of the FLEXPART-WRF trajectory, summed and then integrated in time. This method is preferred to the one based on the WRF-Chem surface precipitation amounts that would not take into account the relative vertical distribution of precipitation occurrence and air parcels. Similarly, we also extract the accumulated condensed water along trajectory (ACWT) to account for the fact that BC particles are also removed from the plumes ~~through~~ during activation. This process, called the nucleation scavenging mechanism, refers to the transfer of particles from interstitial aerosol to cloud-borne aerosol. The ACWT is calculated using the method applied for APT but based on the mixing ratios of cloud liquid water, ice, snow, rain and graupel contents both in grid scale and convective parameterized clouds. The mixing ratios are summed and integrated vertically in each model grid box crossed by the trajectory and finally along this trajectory. The nucleation scavenging mechanism can be considered as a deposition process from the plume when cloud droplets containing aerosols reach the sizes of precipitating rain drops in other cells, e.g. those above or below the grid box crossed by the trajectory. TE_{BC}, APT and ACWT values extracted along each of the trajectories are then averaged to give a mean trajectory corresponding to an uplifted air parcel.~~As presented in Sect. 4.4, this method is applied to 4 boreal fire plumes, 2 plumes from flaring activities in Northern Russia and 2 anthropogenic plumes from Europe.~~

5.3.2 Results along trajectories

In Sect. 4.4, eight distinct plumes have been identified: 4 plumes originating from boreal fire sources in Russia (associated with a small influence of anthropogenic Asian air masses (Fig 9)), 2 plumes from flaring activities in Northern Russia and 2 anthropogenic plumes from Europe. Figure 13 shows the dependence of TE_{BC} ~~values~~ for these eight plumes on the APT and ACWT as a function of the ~~APT, ACWT and~~ FLEXPART-WRF transport time. TE_{BC} systematically decreases with increasing APT or ACWT, reflecting the crucial role of precipitation and nucleation scavenging in removing particles containing BC

from advected plumes along transport, and thereby in controlling the amount of BC reaching the Arctic. ~~A similar behavior is observed plotting TE_{BC} as a function of ACWT, suggesting that BC can also be removed efficiently by nucleation scavenging when transported to the Arctic.~~ An exception occurs on 14-15 July, when TE_{BC} slightly increases in some plumes, because two branches of the same initial plume merge over Northern Canada (Sect. 4.4). TE_{BC} decline is not linear with APT or ACWT (logarithmic scale in Fig. 13) but starts to significantly decrease $6-7$ days ($144-168$ hours) before reaching Scandinavia, corresponding to APT values of $0.2-0.4$ mm and ACWT between 200 and 400 mm.

When reaching Scandinavia (retroplume age very small), plumes present various values of TE_{BC} as a function of their origins. TE_{BC} is the greatest for Siberian biomass burning impacted air masses. As shown in Fig. 9, 85% of the BC sampled by the aircraft in such plumes originated from boreal fires and 15% from Asian anthropogenic masses. In such plumes, the TE_{BC} was as high as $56-68\%$ and was caused by low APT and ACWT values (1 mm and $300-1000$ mm, respectively). In contrast, TE_{BC} was low ($21-28\%$) and APT and ACWT amounts were high for European anthropogenic air parcels ($5-10$ mm and $2000-3000$ mm, respectively). Air parcels influenced by flaring emissions exhibit very different behavior as a function of their altitude: moderate TE_{BC} values (50%) were calculated in plumes reaching the middle Arctic troposphere (3.7 km), whereas TE_{BC} decreased to 20% in plumes transported in the PBL. ~~ACWT~~ The fact that the relation between TE_{BC} and ACWT is different from 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. ACWT appears to be a better proxy for characterizing the efficiency of deposition processes in such air parcels, since the values are of the same order of magnitude in both flaring impacted air masses ($1000-2000$ mm), while APT varies from 0.2 mm (in mid-tropospheric plumes) to 8 mm (in PBL plumes).

The magnitude of the differences in TE_{BC} values obtained during the same period due to various origins is particularly noticeable. European anthropogenic air parcels and flaring emissions are generally transported at lower altitudes than biomass burning air masses, according to Fig. 8. They encountered more ~~(~~non-convective ~~)~~ precipitation caused by frequent drizzle in low-level stratocumulus clouds, scavenging aerosols during transport. In contrast, Russian and to a lesser extent Asian air masses have exported BC more efficiently towards the Arctic. Most air parcels from those sources have experienced transport in WCBs, removing part of the BC loadings. But, the transport of aerosols has been facilitated thanks to the injection of fire plumes at high altitudes and to the fact that ~~the locations of biomass burning~~ biomass burning sources in Siberia are ~~very septentrional sources~~ located far north in summer ($45-65$°N). These results are in agreement with previous studies from Matsui et al. (2011) and Sharma et al. (2013). They underlined a significant inhibition of emissions from eastern Asia (90%) compared to biomass burning sources in Russia on the total Arctic BC, because of larger precipitation rates and stronger scavenging.

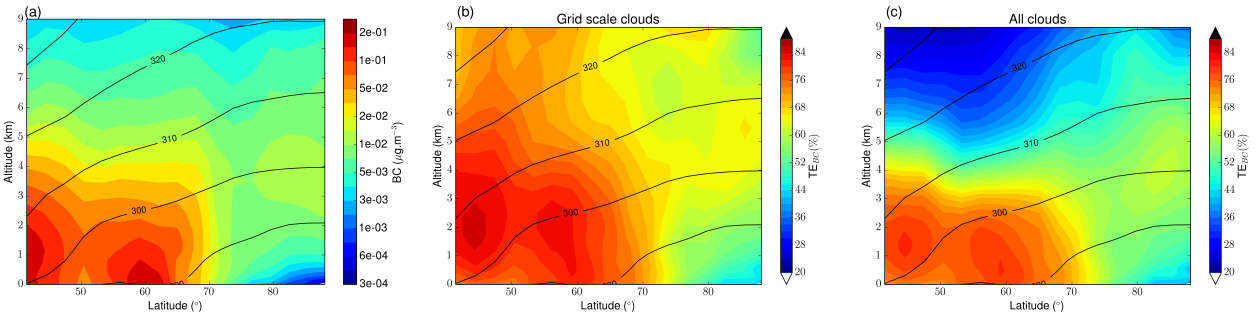

Figure 14. Latitude-altitude cross-sections of (a) the BC mass concentrations, and the corresponding TE_{BC} values due to (b) grid scale clouds and (c) all clouds, averaged zonally and temporally over the ACCESS campaign period in the BASE run. On each panel, black solid lines represent the dry isentropes between 290 K and 330 K.

5.3.3 Zonal mean of transport efficiency

The vertical distributions of the mean BC mass concentrations zonally averaged during the ACCESS period in the BASE run and the corresponding mean TE_{BC} values due to grid scale clouds and all clouds are shown in Fig. 14. The ~~BC~~ WRF-Chem domain (Fig.1) is appropriate for this calculation because all sources influencing the Arctic BC during the ACCESS period are in this domain : the sum of anthropogenic, flaring and fire contributions was found larger than 98% (Fig. 9). The BC mass concentrations are greatest below the altitude of $4\ \mathrm{km}$ on the mid-latitude region ($40-66°\mathrm{N}$) with values larger than $50\ \mathrm{ng\,m^{-3}}$. The highest concentrations ($>100\ \mathrm{ng\,m^{-3}}$) are found close to emission sources and they continuously decrease with height. A high contrast is observed between the mid-latitudes and the Arctic. In the mid-troposphere ($3-6\ \mathrm{km}$), the contrast is less pronounced as moderate BC concentrations are spread on a wide latitude range. As a consequence, above $80°\mathrm{N}$, lowest BC concentrations are found at the surface, and the maximum is detected between 2 and $4\ \mathrm{km}$ ($15\ \mathrm{ng\,m^{-3}}$).

In contrast, TE_{BC} due to only grid scale precipitation exhibits a very distinct dependency on latitude: above $70°\mathrm{N}$, TE_{BC} decreases from $70-85\%$ to ~~44 – 66%, illustrating the~~ 44 – 69%. This sharp meridional gradient ~~in the distribution of moisture and precipitation~~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). This explains the contrast between the BC concentrations in the mid-latitudes and in the Arctic region. Within the Arctic, the maximum of TE_{BC} (~~≃ 66%~~≃ 69%) is found at higher altitudes (4 and $6\ \mathrm{km}$) than the maximum in BC concentrations. This provides an evidence that the transport in WCBs partly, but not completely, removes carbonaceous aerosols by wet deposition. The low BC concentrations in the lowest polar layers are correlated with a weak transport efficiency (45%). This is due to frequent drizzle produced by the formation of low-level stratocumulus clouds in the Arctic PBL and lower troposphere, together with a slower transport in the lowest altitudes. Is is also interesting to note that in the mid-latitude regions, higher TE_{BC} are decoupled from the surface, reflecting the strong uplift of Siberian fires emissions above boreal forest. The associated strong ascent generally occurs at subgrid scale ($<40\ \mathrm{km}$), which

explains the inability of wet removal in large scale precipitation to clean the atmosphere above sources.

TE_{BC} due to wet removal in all clouds exhibits a totally different pattern. TE_{BC} values are correlated more closely with BC mass concentrations, suggesting that wet removal in subgrid parameterized clouds (cumuli) is also fundamental to understand the vertical distribution of BC mass. It also suggests that ~~, in cumuli , the effect of wet deposition~~ cumuli in the mid-latitudes ~~is more important than the enhanced vertical transport in reducing~~ removed more of the low-level ~~aerosols. The interactions between aerosols and clouds~~ aerosol by scavenging or washout than by uplifting it. Aerosol removal in cumulus clouds is responsible of the vertical gradient of TE_{BC}, especially in the mid-latitudes. Two noticeable differences between the zonal cross-sections of BC and TE_{BC} are observed. First, in mid-latitudes ($40-70°$N), TE_{BC} is ~~very~~ low above 6 km ($TE_{BC} < 20\%$), whereas BC concentrations reach moderate values in this altitude range: this reflects the ~~very~~ efficient BC wet removal due to convection and precipitation during the rapid ascent over the source regions. The BC mass concentrations observed above 6 km mainly arise from the boundaries. Second, in the Arctic, the maximum of TE_{BC} is located between 3.5 and 5.5 km (as high as 64%): it illustrates the fact that the transport of Siberian fires plumes closely followed isotropes, as also noticed by Matsui et al. (2011). ~~Aerosol removal strongly~~ Wet removal of aerosols tightly modulates long-range transport from Asia and Siberia to the Arctic, with a ~~strong~~ marked zonal component and transport at higher ~~elevations~~altitudes. It is important to note that the transport efficiency is probably ~~a bit~~ slightly overestimated in our study because the model slightly over predicts BC mixing ratios in the mid-troposphere (Fig. 3).

The Siberian plume has been sampled by the Falcon in the upper troposphere ($6-8$ km). In Fig. 14, we find that TE_{BC} is about 60%, which is in agreement with the value found by along trajectories (Fig. 13). The altitude ~~dependency of the BC mass concentrations~~ dependence of BC in the ACCESS aircraft measurements (Fig. 3 and Fig. 8) is consistent with the zonally and temporally averaged TE_{BC} distribution simulated in the BASE simulation during the ACCESS campaign period. This suggests that the general features of wet removal of BC transported in plumes from Siberia and Asia during summer 2012 were well captured in the ACCESS aircraft measurements. We finally note that the TE_{BC} values in this study are much ~~higher than in those~~ larger than the ones reported by Matsui et al. (2011). That discrepancy is partially due to the different year (2008 vs 2012) with different emissions and precipitation patterns (APT values much lower in our study). In addition, the definition of TE_{BC}, used by Matsui et al. (2011), is the transport efficiency of BC uplifted air masses sampled by the aircraft relatively to their corresponding sources. In our study, TE_{BC} represents the transport efficiency of BC due only to wet removal in grid scale and subgrid parameterized convective clouds. The fact that some plumes are exported from the same emission source to different destinations (e.g. over the Pacific Ocean, as in Oshima et al. (2012)) does not influence our results.

6 Summary and conclusions

During the ACCESS campaign in summer 2012, extensive boreal forest fires associated with temperatures record in both western and eastern Siberia strongly impacted the atmospheric composition of the Arctic atmosphere. The Falcon aircraft regularly sampled transported pollution. During two dedicated flights on 17 July, it measured plumes in the middle and upper troposphere that were representative of cross-polar transport from Siberia and Asia towards Scandinavia and Svalbard archipelago. Specifically, enhanced CO concentrations and BC mixing ratios up to 200 ppbv and 25 $\mathrm{ng\,kg^{-1}}$ are reported between ~~7~~ 6 and 8 km altitude, reflecting the influence of pollution transported from remote sources. The WRF-Chem model run at a horizontal resolution of 40 km on a polar stereographic projection shows good skill in capturing the variability ~~seen~~ observed in the vertical distribution of chemical profiles, with NMB of 1.5% and 27.3% for CO and BC respectively. BC increases in the middle and upper troposphere are mostly linked to biomass burning plumes transported from boreal sources (85%), weakly influenced by anthropogenic sources in northern China (15%).

Our results underline that a coarser horizontal resolution (100 km instead of 40 km) deteriorates the model performance, with a significant overestimation of BC levels in the mid-tropophere. This suggests that, at a coarser resolution, the model is unable to resolve the fine structure of plumes transported in altitude across the North Pole, illustrates the difficulty to represent the cloud and precipitation structures and scavenging processes at subgrid scale and points for the need for improved representation of BC processing in global models in the Arctic.

With a horizontal resolution of 40 km, a ~~very~~ good spatial correlation is found between the horizontal distribution of AOD, the strong divergence regions of the BC horizontal fluxes in the ~~PBL,~~ FT and the spatial distribution of the mean upward BC mass fluxes, underlying ideal conditions for BC poleward transport. This highlights two major biomass burning regions in Siberia, south of Krasnoyarsk region and Yakutia, and one source of anthropogenic pollution in central and northeastern China. Aerosols and CO emitted from the two boreal forest fires regions are exported towards the Arctic through the North of Yakutia in the region presenting a large horizontal poleward eddy heat flux. Their transport towards the North Pole is caused by low-pressure systems controlling the progression and path of the plumes. ~~The plume-rise injection model embedded into WRF-Chem high resolution simulation correctly represents the mixing processes at the edge of the plumes as well as plume filamentation.~~ In the lowest Arctic atmospheric layers, weak CO and BC enhancements are linked to the inefficient transport of European anthropogenic emissions and to a lesser extent to emissions from flaring activities in Northern Russia. These emissions are exported over the Barents Sea and transported at low altitudes into the Arctic.

During the ~~long-range~~ cross-polar transport of Siberian and Asian plumes to the Arctic region, a large fraction of BC particles ~~coated~~ mixed with sufficient water-soluble compounds become CCN active and are scavenged by cloud droplets or rain drops via wet deposition associated with the warm section of mid-latitude cyclones. The two wet deposition processes, namely the wet removal by large-scale precipitation and the scavenging in wet convective updrafts contribute almost similarly to the total

accumulated deposition of BC. The weak role of dry deposition is limited to the lower troposphere. The simulated transport efficiency of BC (TE_{BC}), accumulated precipitation along trajectory (APT) and accumulated condensed water along trajectory (ACWT) values provide insights for understanding the wet removal of aerosols transported to the Arctic. The TE_{BC} in biomass burning plumes is about 60% and is found to be due to low APT (1 mm). In contrast, TE_{BC} is ~~very~~ small ($< 30\%$) and APT are larger ($5 - 10$ mm) in plumes influenced by urban anthropogenic sources or flaring activities in Northern Russia and transported at lower altitudes.

The transport efficiency of BC due to ~~grid~~ large scale precipitation is responsible for the sharp meridional gradient in the distribution of BC concentrations. The transport in WCBs, rapidly injecting aerosols into the upper troposphere, removes some but not all carbonaceous aerosols by wet deposition. ~~The wet removal from subgrid parameterized clouds (cumuli)~~ Cumulus clouds in the mid-latitudes ~~is found to be more important than the enhanced vertical transport in reducing~~ removed more of the low-level ~~aerosols and is~~ aerosol by scavenging or washout than by uplifting it and are responsible for the vertical gradient of TE_{BC}, especially in the mid-latitudes. Wet deposition in ~~parameterized subgrid clouds~~ convective updrafts is essentially active below $60°$N, reflecting the distribution of convective precipitation, but is dominated in the Arctic region by the ~~grid-scale~~ large scale wet removal associated with the formation of stratocumulus clouds in the PBL producing frequent drizzle. An understanding of the respective contributions of the different wet ~~removal processes of BC~~ deposition processes is therefore critically important ~~because~~ since it drives the vertical profiles of BC and ~~strongly~~ tightly modulates the long-range transport of BC aerosol from Asia and Siberia to the Arctic, with a ~~strong~~ marked zonal component and transport at higher ~~elevations.The general features of wet removal of BC transported in plumes from Siberia and Asia during summer 2012 were well captured in the ACCESS aircraft measurements.~~ altitudes.

Author contributions. The first author performed the WRF-Chem simulations, the analyses and drafted the paper ; the co-authors contributed text, ideas and discussed the results. Most of the authors participated in the ACCESS campaign in July 2012.

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