18 September 2010

Reply to Reviewer #1 Comments
"*Aircraft observations of enhancement and depletion of black carbon mass in the springtime Arctic*," by Spackman et al.

Reviewer comments have been italicized.
Replies have been indented in normal type.

All page and line numbers refer to the original ACPD-formatted manuscript at:
http://www.atmos-chem-phys-discuss.net/10/15167/2010/acpd-10-15167-2010.pdf

---

Reviewer #1 (19 July 2010):

This manuscript presents in-situ airborne BC measurements in the springtime Arctic in Alaska. The authors discuss briefly the occurrence of BC enhancement in the free troposphere due to long-range pollution transport and the depletion of BC in the Arctic boundary layer. The latter issue is evaluated more thoroughly by a detailed analysis of the measurement data and the application of a box model. The authors thus elaborate a very important aspect by relating advected BC to its deposition on snow and ice and compare their results to surface measurements. Understanding the behavior of BC in the Arctic is of high relevance for understanding Arctic climate change. This work is clearly structured and the figures support the key findings. I recommend publication of this manuscript in ACP after consideration of the comments below.

General comments

In the introduction, p. 15170, lines 11ff, there is a brief discussion of the different characteristics of sulfate and BC aerosol. I understand that your main goal of argumentation is the possible importance of dry deposition of BC. However, a more detailed consideration, including references, of the mixing state and therefrom resulting characteristics of BC aerosol advected to the Arctic and a clarification of your mixing state assumption for this paper will improve the introduction (e.g. Schwarz et al., 2008; Subramanian et al., 2010; Moffet and Prather, 2009, Shiraiwa et. al, 2007). BC emission is usually accompanied by emission of various types of trace gases (SO2 - coal combustion, organic vapors - anthropogenic and BB etc.) that might condense on it in the course of transport (aging). So BC aerosol might not be as pure as I understand from your introduction. Especially, during Arctic springtime, when weather conditions are relatively stable and little wash-out happens, even BC coated with more hydrophilic compounds might survive transport to the Arctic.

We agree that more attention to mixing state in the introduction would provide more context for the observed BC enhancements and depletions. The relevant paragraph in the introduction has been edited to emphasize the importance of dry deposition of BC mass in the Arctic in the context of BC mixing state. We now cite Schwarz et al. (2008), Shiraiwa et al. (2007), and Jacobson (2001) as general references for BC

mixing state and McNaughton et al. (2010) and Brock et al. (2010) as more specific references for BC mixing state and BC removal during ARCPAC/ARCTAS:

"BC mass is observed in snow at sites throughout northern high latitudes (Hegg et al., 2010; 2009; Doherty et al., 2010). The seasonally enhanced BC mass loadings in the Arctic troposphere may lead to increased wet and dry deposition of BC aerosol to the snow or ice through precipitation scavenging and direct contact with the snow or ice surface, respectively. The mixing state of BC (e.g., Schwarz et al., 2008, Shiraiwa et al., 2007; Jacobson, 2001) may be an important aspect in the wet removal of BC particles because they are aged and mixed with hydrophilic materials such as sulfate in the Arctic (McNaughton et al., 2010). However, dry removal may play a more important role than wet removal in depositing BC particles to the snow because of limited precipitation scavenging in the Arctic in winter and spring (Brock et al., 2010). Atmospheric perturbations that facilitate the mixing of BC from the free troposphere into the Arctic boundary layer (ABL), such as through open leads (Serreze et al., 1992; Andreas and Murphy, 1986) or enhanced vertical wind shear, might lead to increased dry deposition of BC aerosol to the snow (Strunin et al., 1997)."

*The use of the box model complements your measurements very well and makes them comparable to surface station observations of BC loadings in snow. The general functioning of the model is explained sufficiently in the paper, however, there are a few details that deserve elaboration: Did you develop this box model especially for this study? If so, you should state it, if not, then a reference should be given. After the introduction of the model you present the results right away with little discussion on how you derived them. The paragraph on p. 15180, lines 4ff, is not clear to me: How many values did you try between 1 and 8 hrs? I would expect to see the same number of lines as selected kbl's (or an area comprising all assumed values) in Fig. 10. It is not clear which value the red curve depicts (single assumption for kbl or an average?). line 6: "The model *better* reproduces the . . .", here I don't understand what you com- pare the removal efficiency factors between 0.1 and 1 to. To which kbl value(s) does this range correspond? You should also formulate an argument why you focus on 5 % removal efficiency for the following calculation. By displaying the model results in a graph you can give the readers the chance to see for themselves how robust the model output is and include much of the discussion in a single figure.*

To improve the box model discussion, the system of coupled linear differential equations developed in this work has been added to Section 4.2 to add transparency to the model description. Figure 11 (formerly 10) now includes several curves of varying removal efficiency to show how the removal efficiency affects the distribution of BC mass loadings in the model and how the 5% value was determined from the BC profiles. The text describing the model results has been rewritten for clarity and now emphasizes the model was developed for this work and could be used by others for similar applications:

"A simple box model is developed to estimate the deposition flux of BC aerosol to the snow. Fig. 10 shows a schematic of this 4-box system. A system of coupled

linear differential equations is formulated to quantify the flux of BC from the free troposphere into the ABL and removal by contact with the surface:

$$\frac{dC_4}{dt} = 0 \tag{1}$$

$$\frac{dC_3}{dt} = k_{FT}(C_4 - C_3) - k_{BL}(C_3 - C_2) \tag{2}$$

$$\frac{dC_2}{dt} = k_{BL}(C_3 - C_2) - k_{BL}(C_2 - C_1) \tag{3}$$

$$\frac{dC_1}{dt} = k_{BL}(C_2 - C_1) - fk_{BL}C_1 \tag{4}$$

The BC mass loading in the aged Arctic air mass in the free troposphere is represented by C_4. The BC mass loadings in the ABL are partitioned into 3 boxes each representing 100 m altitude depth and given by C_3, C_2, and C_1. The free troposphere-to-boundary layer exchange coefficient, k_{FT}, is inversely proportional to the e-folding timescale for mixing between the free troposphere and boundary layer. Similarly, the ABL exchange coefficient, k_{BL}, is inversely proportional to the e-folding timescale for mixing between 2 boxes in the ABL and is considered constant throughout the depth of the boundary layer. The e-folding timescale for mixing through the full depth of the ABL is then expressed as $3k_{BL}^{-1}$. A removal efficiency factor, f, of BC particles is used in the model to scale the deposition velocity of BC mass from the lowest altitude box to the snow. The removal efficiency is an effective removal efficiency that represents the fraction of particles that actually come into direct contact with the surface within the boundary layer turnover timescale. The removal efficiency factor is constrained by the observations.

This 4-box box is the simplest system to model the flux of BC mass from the free troposphere into the ABL and to the surface. The 3 boxes in the ABL are sufficient to represent the BC vertical gradient in the ABL while more boxes would not add more value to the model results. The variables k_{FT}, k_{BL}, and f are interrelated and not fully independent in the model. For example, the exchange coefficient k_{FT} is expressed as a small fraction of k_{BL} (i.e., $k_{FT}/k_{BL} < 0.15$) in the model because the observations indicate the timescale for mixing between the free troposphere and ABL is longer than the ABL turnover timescale. This ratio controls the BC vertical gradient at the boundary layer transition at ~300 m altitude as illustrated schematically in Fig. 9. Larger values of f lead to the result in the second panel of Fig. 9 with near complete removal near the immediate surface. The choice of k_{BL} (and hence k_{FT}) does not affect the steady-state concentrations of BC mass in $C_4 - C_1$ but does affect how quickly the model distributes BC. We later prescribe a value for k_{BL} to estimate the deposition flux of BC mass to the surface.

In theory, the system of Eqs. (1)–(4) can be solved uniquely for k_{FT}/k_{BL} and f. However, the variability in the BC vertical profiles and the limited data at the boundary layer transition (~300 to 750 m) do not provide the tight constraints necessary to uniquely determine k_{FT}/k_{BL} and f. Thus, a range of solutions bounding the observations with various values of k_{FT}/k_{BL} and f is shown in Fig. 11. These solutions are determined for the mean profile shown here but may not generally apply to BC depletion events in the ABL, such as the profile in Fig. 6C. Panels A and B in Fig. 11 show the sensitivity of the model to f and k_{FT}/k_{BL}, respectively. As shown by the solid red and blue lines, the model better reproduces the BC vertical profiles in the ABL for k_{FT}/k_{BL} values between 0.15 and 0.02. This range of smaller values of k_{FT}/k_{BL} confines more of the BC vertical gradient to the boundary layer transition. These solutions are consistent with the change in BC mass along the O_3–BC mixing line at the boundary layer transition shown in Fig. 8B. To maintain ~20 ng kg^{-1} of BC at the lowest altitude in the observations shown in Fig. 11, 1.5 to 10% removal efficiency is required for the k_{FT}/k_{BL} values of 0.02 and 0.15, respectively. In general, a larger inflection point grows into the profiles in Fig. 11 at 250 m altitude as both k_{FT}/k_{BL} and f decrease.

The box model results are used to estimate a dry deposition flux of BC aerosol to the surface. The most significant terms in the flux calculation are the timescales for mixing across the boundary layer transition, given by k_{FT}^{-1}, and the removal efficiency, f. The flux is calculated with the expression formulated here:

$$\text{Dry Deposition Flux of BC} = \frac{\Delta BC \cdot f\left[1-(1/e)\right] \cdot H}{k_{FT}^{-1}} \quad (5)$$

The total BC removal for the e-folding timescale, k_{FT}^{-1}, over the depth H (sometimes corresponding to the depth of the ABL) is given by the expression, $\Delta BC \cdot f\left[1-(1/e)\right]$. The BC depletion, ΔBC, is estimated at 20 ng kg^{-1} over the 300 m depth of the ABL based on the underlying data (gray points) in Fig. 11."

p. 15169, paragraph lines 15-24: From this explanation a non Arctic-specialized reader might not fully understand why there is an enhancement of BC (and other aerosol types) during Arctic winter and spring. You should elaborate this paragraph by answering the following questions: What are the origins of the Arctic aerosol and where are the sources located (within the Arctic dome)? Why is there build-up of Arctic haze (stable weather conditions, little wet deposition)? Line 22ff: "Together, these phenomena lead to an increase in BC aerosol . . ." increase compared to what? ". . .we refer to as aged Arctic air in this work." You should clarify that you exclude influence from (fresh) pollution plumes in your definition of aged Arctic air.

The general idea was to discuss the processes controlling BC aerosol—sources,

transport, and removal—in the springtime Arctic in paragraphs 2 and 3 of the introduction. The conditions governing the removal processes were introduced in paragraph 2 and the source regions and long-range transport were discussed in paragraph 3. We have maintained this general structure and the text describing the Arctic haze phenomenon (paragraph 2) has been expanded to elaborate on the removal processes in the springtime Arctic and add clarifying discussion:

"Enhancements of BC and other tracers of pollution have been observed for decades in the Arctic troposphere in the winter and early spring (Sturges, 1991; Hansen and Novakov, 1989; Sharma et al., 2006; Law and Stohl, 2007; Koch et al., 2009). These enhancements emerge from the chemistry and dynamics unique to high latitudes at this time of year. In the winter and early spring, the colder temperatures and weaker solar insolation lead to stratification in the Arctic troposphere that inhibits precipitating cloud formation and, hence, the wet removal of aerosols. The weaker insolation and colder temperatures also slow down the photochemistry and reaction rates that govern the chemical tracer abundances. As a result, aerosols and tracers are generally longer lived in the Arctic in winter and early spring than at other times of the year."

"Aged Arctic air" is now defined later in the paper at p. 15172, line 22:

"We use the phrase, aged Arctic air, in a qualitative sense to exclude fresh pollution from sources inside and outside the Arctic, such as biomass-burning plumes."

Specific Comments

p.15168, line 11: "Maximum average BC mass loadings..."It is not clear what you mean by this term. (Do you mean maximum average per altitude bin?)

We refer to the highest average, altitude-binned BC mass loading given by the blue line (aged Arctic air mass) in Figure 2. We have clarified and expanded this sentence to include a remark about biomass-burning plumes given by the red line in Figure 2:

"Average BC mass mixing ratios peaked at 150 ng BC (kg dry air)$^{-1}$ near 5.5 km altitude in the aged Arctic air mass and 250 ng kg^{-1} at 4.5 km in biomass-burning influenced air. BC mass loadings were enhanced by up to a factor of 6 from near the top of the Arctic boundary layer (ABL) to 5.5 km in biomass-burning influenced air compared to the aged Arctic air mass."

p.15168, line 16ff: ". . .across the boundary layer transition in the ABL. . ." This formulation is unclear: Does the gradient increase within the ABL or within the transition between ABL and the lower troposphere?

The BC-altitude gradient actually occurs within the ABL and across the ABL transition. In general, factors of 2–3 and 4–6 generally apply to the aged Arctic and biomass-burning influenced air masses, respectively. The change in BC mass at the boundary layer transition was often larger or more variable than the change in the ABL but this was often associated with the biomass-burning plumes in the free

troposphere. For these latter cases, CO did not remain constant across the boundary layer transition as shown in Figure 3. The text has been edited to properly reflect the BC gradient for invariant CO in the aged Arctic air mass:

"In the aged Arctic air mass, BC mass loadings more than doubled with increasing altitude within the ABL and across the boundary layer transition while carbon monoxide (CO) remained constant. This is evidence for depletion of BC mass in the ABL."

p.15168, Line 18-22: The logic of this sentence is unclear. The relation between the release of Br2 and the dry deposition of BC is not mentioned.

We have restructured this part of the abstract describing the BC–O_3 correlation to convey the idea more clearly:

"BC mass loadings were positively correlated with O_3 in ozone depletion events (ODEs) for all the observations in the ABL. Since bromine catalytically destroys ozone in the ABL after being released as molecular bromine in regions of new sea-ice formation at the surface, the BC–O_3 correlation suggests that BC particles were removed by a surface process such as dry deposition."

p.15170, paragraph lines 8-17: do you have a reference for these statements?

In response to the general comments from this reviewer, we have rewritten this paragraph that discusses wet and dry removal of BC particles in the Arctic to add mixing state context. The paragraph is reproduced here:

"BC mass is observed in snow at sites throughout northern high latitudes (Hegg et al., 2010; 2009; Doherty et al., 2010). The seasonally enhanced BC mass loadings in the Arctic troposphere may lead to increased wet and dry deposition of BC aerosol to the snow or ice through precipitation scavenging and direct contact with the snow or ice surface, respectively. The mixing state of BC (e.g., Schwarz et al., 2008, Shiraiwa et al., 2007; Jacobson, 2001) may be an important aspect in the wet removal of BC particles because they are aged and mixed with hydrophilic materials such as sulfate in the Arctic (McNaughton et al., 2010). However, dry removal may play a more important role than wet removal in depositing BC particles to the snow because of limited precipitation scavenging in the Arctic in winter and spring (Brock et al., 2010). Atmospheric perturbations that facilitate the mixing of BC from the free troposphere into the Arctic boundary layer (ABL), such as through open leads (Serreze et al., 1992; Andreas and Murphy, 1986) or enhanced vertical wind shear, might lead to increased dry deposition of BC aerosol to the snow (Strunin et al., 1997)."

p.15171, line 10ff: It is unclear whether 7 hrs of measurement in the ABL took place or if only 7 hrs of these data are discussed.

The sentence has been edited for clarification:

"The aircraft probed the ABL mostly over the ice and open leads north of Alaska for a total ~7 h in duration."

p.15171, line 20: 2 g/cm^3 density, is Schwarz et al., 2006 also meant to be the reference for this value or is this assumption based on different considerations?

This assumed density for ambient BC of 2 g/cm^3 has been used for all NOAA SP2 datasets and is based on the known density of graphite, 1.9–2.3 g/cm^3, from the CRC Handbook. Note that the assumed density has a small impact on the computed size because the diameter goes as the third root of the volume of a BC particle. In instantaneous heating rate calculations, the assumed density is important because the index of refraction is directly related to the BC particle density. We have added a comment to the text noting that the assumed value for the density does not affect the results in the paper:

"This assumed density does not affect any results in this work except the VED in the constructed size distributions."

p.15171, line 25ff: What do you expect to be the losses due to your inlet system? Is there a reference for the low-turbulent inlet? How long was your inlet system tubing, what kind of tubes did you use, how large is your sample flow?

Although we do not mention the details in the manuscript, this is an important point. For accumulation-mode BC mass reported here, we expect less than 10% losses through the inlet and sample line (Gao et al., JGR, 2008). To verify this, we performed a detailed intercomparison among aerosol instruments sampling off the low-turbulence inlet (LTI) on the WP-3D. We prepared various test aerosols (e.g., polystyrene latex spheres, ammonium sulfate, etc.), size-selected the polydisperse aerosol with a DMA, and then sampled the monodisperse aerosol through the installed sample lines. Good agreement between the instruments at their different sampling points along the turbulent line indicated minimal aerosol losses during transport along the sample tubing. We have expanded the text where the sampling is discussed to provide the details for this test:

"The SP2 sampled ambient aerosol behind a 1 μm particle impactor through a low-turbulence inlet (LTI) (Brock et al., 2004; Wilson et al., 2004). The sampling inlet was designed and mounted to the aircraft in a manner to sample the ambient air stream without contamination from the boundary layer along the skin of the aircraft. To ensure no particle losses during sampling, an intercomparison was performed between aerosol instruments sampling off the LTI. For various test aerosols introduced, good agreement between the instruments at their different sampling points along the sample line indicated minimal aerosol losses during transport along the sample tubing."

The work of Wilson et al. (2004) has been added to the text as a reference for the LTI. Other SP2 instrument details are generally described by Schwarz et al. (2006).

p.15172, line 18: "BC mass" should be named "BC mass concentration" (here and throughout the paper, also in the figure captions)

We prefer "BC mass loadings" or "BC mass mixing ratios" and have now applied this consistently throughout the paper.

Figure 2: Not the lines but the round markers denote the 1 km mean values. For the black curve the markers seem to illustrate a 500 m average. One-sided error bars seem rather confusing to me.

We have chosen to keep the one-sided error bars because the BC mass loadings are shown on a log scale. Two-sided error bars overwhelm the plot for smaller BC mass mixing ratios. The figure caption has been rewritten for accuracy:

"**Figure 2**. Vertical profiles of BC mass mixing ratio for 5 flights in the Arctic during ARCPAC (blue and red data) and 16 flights at midlatitudes in eastern Texas during the Texas Air Quality Study (black curve). The data in blue and red are from 1 flight in the aged Arctic air mass and 4 flights in long-range biomass burning plumes, respectively. The small dots and line markers denote 30 s averages and 1 km mean values, respectively, of BC mass mixing ratio, except at midlatitudes where 0.5 km averages are shown. The horizontal bars represent one standard deviation on both sides of the mean but are only drawn on the positive side for visual clarity."

p.15172f, line 25ff: This sentence should be rephrased for more clarity. ("increase with altitude" and "with increasing altitude from the lower to upper troposphere" is redundant) It is not clear to me if you mean both, the blue and red curve given that the blue curve continues like the red one >5.5 km, or only the red curve.

The sentence has been restructured to clarify the descriptions of the Arctic BC vertical profiles:

"For both types of air masses, the mean BC mass loadings increase with altitude in the springtime Arctic. Average BC mass mixing ratios peak at 150 ng kg^{-1} near 5.5 km altitude in the aged Arctic air mass and over 250 ng kg^{-1} in biomass-burning influenced air at 4.5 km. BC mass loadings are enhanced by up to a factor of 6 from the top of the Arctic boundary layer to 5.5 km in biomass-burning influenced air compared to the aged Arctic air mass."

These edits have also been incorporated into the abstract.

p.15173, line 4ff: Do you refer to the Texas Air Quality Study shown in Fig. 2? If yes, you should mention the black curve and reference already here. If not, then you should give another reference.

This sentence refers to the midlatitude vertical profile from Texas in Figure 2. The text is edited to include the citation:

"The BC mass loadings in the middle troposphere are comparable to those observed in the planetary boundary layer in a polluted urban environment in the United States (Spackman et al., 2008)."

p.15173, line 6ff: This paragraph needs some restructuring for the reader: "Although one would not necessarily expect agreement. . ." It is unclear whether you refer to the comparison of midlatitude boundary layer and Arctic free troposphere BC concentrations or to the comparison of SP2 and aethalometer measurements. If the latter is the case you should start a new paragraph and start with one introductory sentence that puts this paragraph into context. You should also state that both campaigns took place in the same geographical area.

Starting with this sentence, we are comparing SP2 and aethalometer measurements in the Arctic. The text has been moved to a new paragraph and expanded. We have also noted in the text that the AGASP campaign also took place in the Alaskan Arctic:

"The profiles obtained with the SP2 during ARCPAC generally show lower BC mass loadings compared to those from an aethalometer during the Arctic Gas and Aerosol Sampling Program (AGASP) conducted in the Alaskan Arctic (Schnell, 1984). Agreement between the AGASP and ARCPAC data should not necessarily be expected because of variability in long-range transport and changes in BC emissions over time. The ARCPAC profiles show distinct enhancements in BC mass in the middle and upper troposphere compared to the lower troposphere unlike the composite profiles from AGASP-I (March 1983) and -II (April 1986) that indicate a polluted lower troposphere and decreasing BC mass loadings with increasing altitude. In summary, BC mass mixing ratios decreased from approximately 150–400 ng kg^{-1} at lower altitudes to 20–75 ng kg^{-1} at 8-10 km (Hansen and Novakov, 1989)."

p.15173, line 26f: red points, do you include or exclude data points that might be influenced by the aircraft's own off-gas during take-off or landing (if your instrument was already acquiring data during that time)?

The red points in Figure 3 represent the ambient BC data on ascent from and descent into Fairbanks. The sampling inlet is designed and mounted to the aircraft in a manner to sample the ambient air stream without contamination from the boundary layer of the skin of the aircraft. The work of Brock et al. (2004), cited in the instrument section, provides details of the sampling. We have added a reference for the low-turbulence inlet (Wilson et al., 2004) and added a sentence to the text to clarify the sampling:

"The sampling inlet was designed and mounted to the aircraft in a manner to sample the ambient air stream without contamination from the boundary layer along the skin of the aircraft."

Figure 3: Minor tick marks for the vertical axis (100 m) would be helpful since you mention "several hundred", "700 m" etc. in the text. The captions say that the lines with

markers represent 1 km altitude bins: However, there are 5 markers per 1 km. Again, not the lines but the markers represent the averages. Neither the text (p.15174, line 3f) nor the caption state if the Fairbanks data is included in the black curve. Optionally, you might shade the ABL for quicker understanding by the reader.

One hundred meter minor tick marks have been added to the vertical axes. The mean vertical profiles include the Fairbanks data but these do not significantly affect the mean in the free troposphere because of the small number of data points on initial ascent and final descent. The figure caption has been updated.

p. 15174, paragraph line 1-12: I recommend stating clearly what you consider ABL, lower, free and upper troposphere as you mention these terms continuously throughout the text. In this paragraph it is not clear that you consider <300 m the ABL. This only becomes evident later.

The ABL height was specified at p. 15171, line 10. We have now defined the ABL in more detail in the text at p. 15171, line 11:

"The ABL is defined in this work as the surface layer, typically characterized by a temperature inversion, over the sea-ice and open leads up to about 300 m altitude based on the temperature profiles from the WP-3D. This layer generally appears consistent in depth and inversion strength with temperature soundings over the pack ice in the central Arctic north of Alaska (Tjernström and Graversen, 2009). The free troposphere refers to the region immediately above the ABL."

We use the terms lower and upper troposphere more loosely in the paper and do not define them explicitly in the text.

p. 15174, line 16: 160 ppb CO background value, is that your definition based on the observations or is it a literature value (if so, reference should be given)

The 160 ppb CO background is explicitly stated in Brock et al. (2010). We have now added a citation to this work.

Figure 4: Uppermost panel: You can either insert a legend or color the axes

Thank you for noting this. The axis labels are now color-coded in Figure 5 (formerly 4).

p. 15176, line 15f: The BC removal needs to be defined to more detail at this point. What time scale does 15 ng kg-1 refer to? It is unclear what exactly determines the top of the profile.

We estimate BC removal from the profile in Figure 6B (formerly 5B) from the difference between the BC mass loading at 300 m and at ~100 m, near the lowest altitude measurement. The timescales associated with the removal are unknown. We investigate the timescales for the removal processes (deposition flux) in Sect 4.2. In

the text we more clearly state over what altitude range we determined the BC removal:

"We use an average of the ascent and descent profiles in Fig. 6B (shown in Fig. 11) to estimate the difference in BC mixing ratio of ~20 ng kg^{-1} between the top of the boundary layer and ~100 m altitude, at the bottom of the profile. The mean profile from Fig. 6B is used to estimate the BC depletion because the BC variability is relatively small compared to other profiles such as in Fig. 6C. We assume the 20 ng kg^{-1} BC difference from the profile in Fig. 6B extends 300 m over the full depth of the ABL to calculate a deposition flux of BC to the snow in Sect. 4."

Figure 6: You should add a legend to the plot, in the legend you can include example error bars for the data points.

The slopes and R squared values of the two correlations shown in Figure 7 (formerly 6) are now stated explicitly in the legend. The figure caption has been updated:

"**Figure 7**. Correlation between BC mass and CO mixing ratios for the flight of 21 April. The 30 s data points are discriminated by altitude with the red points highlighting the correlation below 750 m altitude and the black points denoting the rest of the data. The slope with one standard deviation and the R^2 value are shown for each linear regression."

*p.15177, line 9ff: "Although open leads *also* inject water vapor into the ABL, clouds were generally not observed over the open leads during ARCPAC *because* many of the leads were observed to be at least partially covered with thin ice (Brock et al., 2010)." This explanation should be elaborated. It is not clear why there should be no clouds above "partially covered open leads" if water vapor injection from open leads is only a contribution to water vapor abundance in the atmosphere. So clouds might still form.*

This whole paragraph has been restructured to emphasize the potential role of moisture from the open leads in influencing the removal of BC particles from the ABL:

"Although many of the open leads during ARCPAC were observed to be at least partially covered with thin ice, the leads were still a major moisture source to the ABL and resulting diamond dust may have played a role in the removal of BC particles through impaction scavenging by these ice crystals (Feng, 2009). A wet removal process whereby BC particles act as ice nuclei is less likely because clouds were generally not observed over the open leads. In general, very limited precipitation scavenging of total aerosol was observed during ARCPAC (Brock et al., 2010). This is in contrast to previous observations of preferential scavenging of BC as ice nuclei in mixed-phase clouds at a high-altitude site in midlatitudes (Cozic et al., 2008). Because it was difficult to unambiguously identify open-lead influenced air in the aircraft data, we cannot definitively comment on the potential role of open leads in the removal of BC with this dataset."

p. 15177, line 22: Why did you choose 750 m in this case when using 700 m before? Do 700 and 750 m make a significant difference?

We have changed 700 to 750 m throughout the manuscript for consistency. There is no significant difference between 700 and 750 m.

p.15178, line 15ff: This sentence does not state the logical connection between Br2 release, i.e. sea-ice formation, and BC removal by contact with snow. A reference for this general theory should also be given.

As far as we are aware, the correlation between O_3 and BC in the ABL has not been published in the literature. We postulate the theory for this phenomenon in this manuscript. The proposed mechanisms for ODEs are given by Simpson et al. (2007) and bromine observations in ODEs are shown during ARCPAC by Neuman et al. (2010). The paragraph that describes the link between ODEs and dry deposition of BC has been rewritten in response to this and other reviewer comments:

"The O_3–BC correlation in ODEs is robust over the course of 5 flights spanning 10 days with a total of 7 hours sampling in the ABL. In the ODEs, O_3 is removed through catalytic destruction by active bromine [*Simpson et al.*, 2007]. Enhancements of molecular bromine were observed in the ODEs during ARCPAC [*Neuman et al.*, 2010]. Molecular bromine is believed to be released to the atmosphere from brine during sea-ice formation and then rapidly photolyzed to active bromine. As shown in Fig. 8B, lower O_3 was generally found at lower altitude in the ABL and higher O_3 near the top of the ABL. This correlation between O_3 and BC mass in ODEs suggests BC particles have been preferentially removed by a surface process such as dry deposition. The competing hypothesis that precipitation scavenging removes BC mass in the vicinity of open leads is less likely because this correlation would not be expected if ice crystals were scavenging BC particles through the depth of the ABL or even preferentially at the top of the ABL. Another possible hypothesis is that sedimentation of BC-containing particles, enhanced in size by hygroscopic materials in the ABL, could contribute to the deposition of the BC mass to the snow. However, the SP2 sizing information for the internally mixed BC particles sampled in the ABL suggests there is insignificant mass at the larger sizes required (approaching 1 μm) for sedimentation to make a significant contribution to BC removal."

p.15178, line 25: how big is the uncertainty of the size distributions? Is there a significant difference between the two modes? What is your explanation for the more aged aerosol mode being smaller than the fresher one advected by the BB plume? Do you assume that more thickly coated particles have been removed from the aged air while they still survive within the fresher air mass?

The difference in the modes of the size distributions in the aged Arctic air (~160 nm) versus those in biomass burning plumes (~200 nm) is significant. In general, we have observed larger modes in biomass burning plumes compared to the remote

atmosphere (Schwarz et al., GRL, 2008). The difference in the modes is likely due to source type and the combustion process but also may be related to coagulation and aging during transport. We observed thickly coated particles in both the biomass-burning plumes and the aged air masses. We have now restructured the part of the paper on size distributions and added a new figure (Fig. 4) to the paper showing size distributions from the free troposphere and ABL on 12 April in the aged Arctic air. We have also moved the discussion of the size distributions to Section 3.2 as supporting text for the argument for BC removal in the ABL. In this context, we do not explicitly state the mode for the size distribution in biomass-burning plumes because it turns out there is a lot of variability in the size distributions in the plumes and this is not directly relevant to the discussion of size distributions on 12 April in the aged Arctic air. Here is the updated text from paragraph 2 of Section 3.2:

"Rather, the simultaneous measurements are consistent with the physical removal of BC aerosol from this air mass through deposition. Size distributions of BC mass in the ABL and free troposphere on 12 April support this conclusion (Fig. 4). The mode for both these mass distributions is ~160 nm, suggesting the air masses in the ABL and the free troposphere have similar sources. The data from two other aerosol instruments (for details of the NMASS and UHSAS instruments, see Brock et al., 2004) aboard the WP-3D aircraft that measure particle number and size distributions also showed evidence for particle removal in the ultrafine and fine modes (i.e., 4–1000 nm). On the whole, the tracer and aerosol data across the boundary layer transition strongly suggest the air masses in the ABL and free troposphere are from the same sources."

p.15178, line 26: Is there a specific reason you only refer to 18 April and not the other flights?

We referred to the 18 April flight because we were comparing size distributions from 12 April in the aged Arctic air and 18 April in biomass burning plumes. The WP-3D sampled some of the most intense biomass-burning plumes on the 18 April flight. As mentioned in the response to the above comment, we have now restructured this part of the paper on size distributions and do not explicitly state the mode for the size distribution in biomass-burning plumes.

p. 15180, line 4: What is the basis for this assumption?

One to 8 hours are rough estimates for diffusion timescales for a stable boundary layer based on the observed vertical wind shear. Time scales as long as 8 hours have been found at the South Pole for very stable, shallow boundary layers (Neff et al., Atmos. Environ., 2008). It would be expected that under higher wind conditions with the potential for solar heating at the surface, the time scale could be much shorter. As a result, we have rewritten Sect. 4.2 (box model discussion) and now base the dry deposition flux on a boundary layer mixing timescale of 1 h and then extrapolate the results to different possible timescales in the discussion. We have also added clarifying remarks about this assumption in this section:

"Diffusion timescales as long as 8 hours have been found at the South Pole for very stable, shallow boundary layers (Neff et al., 2008). However, the timescale could be much short under higher wind conditions with the potential for solar heating at the surface in the springtime Arctic."

p. 15181, line 3: A reference should be given.

We assume the snow exposed to the air is still fresh and uncompacted with a density of 100 kg m^{-3} when deposition occurs. The value of 100 kg m^{-3} is based on current operational meteorology practice of assuming a 10-to-1 ratio for snow-to-liquid density ratios (Roebber et al., 2003). Baxter et al. (2005) determined average snow-to-liquid ratios over the contiguous US are 13:1. The average temperature range in Barrow in April is -14 to -22°C may justify increasing this ratio. However, for the purposes of this rough calculation, we prefer to keep to the simple 10:1 rule especially since the snow may get compacted upon initial impact with the surface resulting in a slightly higher snow density. The text has been revised to clarify this choice and add a citation to Roebber et al. (2003) at p. 15181, line 1:

"When deposition occurs, we assume the snow exposed to the air is relatively uncompacted with a density of 100 kg m^{-3} based on a 10:1 snow-to-liquid density ratio (Roebber et al., 2003). The climate data (1971-2000) at Barrow, Alaska, are probably representative of weather conditions over the Arctic Ocean in the general region where the aircraft measurements took place. The April mean temperature in Barrow of –18°C may justify selecting a higher snow-to-liquid ratio but since the snow may be compacted somewhat upon initial impact with the surface, the 10:1 ratio is still used for this calculation. We then use the climatological April mean snowfall of 5.3 cm at Barrow to calculate the average BC mass in the snow based on the dry deposition fluxes from the box model. This equates to 0.5 to 28 ng BC (g snow)$^{-1}$ for a 1 h boundary layer turnover timescale."

p. 15181, line 12ff: ". . . amount of snowfall at a given site and the mass of BC advected to the Arctic." If, for example, there are only very little biomass burnings during springtime season the amount of BC will be less, so less can be deposited.

This important comment alludes to one of the major motivations for this work (p. 15170, lines 14-17). Interannual variability in BC mass advected to the Arctic may affect the amount of BC mass deposited to the snow. But this fate ultimately depends on whether BC aerosol is mixed from the free troposphere into the boundary layer before it is transported out of the Arctic. The sentence at p. 15180, lines 12-16 states that we did not observe any (with one exception) evidence of biomass-burning plumes in the ABL during the observation period. We have added a sentence at p. 15181, line 14 to incorporate this point:

"The variability may also be affected by the interannual variability in BC mass advected to the Arctic and to what extent it is then mixed into the ABL and deposited to the snow surface."

We have also expanded the end of Section 5 (Summary) into a new paragraph to include these important points:

"Interannual variability in BC mass advected to the Arctic may affect the amount of BC deposited to the snow. However, the fate of BC aerosol ultimately depends on whether it is mixed from the free troposphere into the boundary layer before it is transported out of the Arctic. The analysis in this work indicates dry deposition of BC aerosol occurred in the aged Arctic air mass. There is no evidence during ARCPAC that the biomass-burning layers in the free troposphere mixed into the ABL and reached the snow surface. However, the work of Hegg et al. (2009) presents a conundrum because it suggests that the bulk of the BC in Arctic snow is attributed to fire sources. The spatially and temporally limited representativeness of the observations during ARCPAC may explain some of this disparity. On the other hand, the possible role of open leads in the sea-ice in facilitating the entrainment of biomass-burning layers aloft may also help to explain this discrepancy. Enhanced deposition may be occurring in regions influenced by open leads in the ice because the open leads facilitate mixing between the free troposphere and ABL under certain meteorological conditions. Additional observations will be important to understanding the processes controlling BC aerosol in the ABL that have important implications for Arctic climate."

p.15182, line 3ff: From this sentence and the following I understand that enhanced deposition of BC occurring in the region of open leads is an observation made during your measurements. From the paper I understand that this is an interpretation of the data.

The data do not provide hard evidence for a causal link between the leads and dry deposition of BC particles. At p. 15177, lines 16-18, a caveat was stated that the analysis could not evaluate lead vs. non-lead cases. However, the data certainly give the impression that the open leads may have enhanced deposition of BC to snow, keeping in mind the ARCPAC objectives consisted of sampling air in the vicinity of the open leads. We feel the statements in the text currently reflect this weight of evidence from the data so no changes have been made.

Technical Comments

Figure 1: The aircraft base, degree latitude and longitude and a scale should be indicated.

Figure 1 has been modified to zoom in on the flights tracks and to include a label for the deployment site and latitude/longitude gridlines. The figure caption has also been updated.

Figure 5: "The gray-shaded regions. . ." in the online version the shaded areas appear red.

The text describing Figure 6 (formerly 5) and the caption have been edited to only make reference to "shaded" regions without mentioning color to avoid any confusion.

p.15168, line 8: POLARCAT should be spelled out

The acronym, POLARCAT, has been removed from the abstract to avoid distracting the reader from the main thesis of the abstract. POLARCAT is defined in the body of the paper.

p.15168, line 18: rephrase "remained constant, evidence for" to ". . .remained constant. This is evidence for . . ."

Done.

p.15168, line 20: rephrase "in the ABL suggesting that ..." to "... in the ABL. This suggests that. . ."

As described in the reply to the specific comments above, the sentences at lines 18-22 have been restructured:

"BC mass loadings were positively correlated with O_3 in ozone depletion events (ODEs) for all the observations in the ABL. Since bromine catalytically destroys ozone in the ABL after being released as molecular bromine in regions of new sea-ice formation at the surface, the BC–O_3 correlation suggests that BC particles were removed by a surface process such as dry deposition."

p.15170, line 23: ". . .in the rest of the paper." can be dropped.

Done.

p.15171, line 1ff: rephrase "The . . . research aircraft deployed to. . ." "The . . . research aircraft was deployed to. . ."

Done.

p.15171, line 19: there is a "-" between 600 and nm

The hyphen is removed here and in all other cases in the manuscript with the construction, numeral + unit of measure + noun.

p.15172, line19f: "The individual data points are 30-s averages of BC mass each representing a horizontal spatial resolution of 3 km." Rephrase to either: "The data points are 30-s averages with a horizontal spatial resolution of 3 km." or: "The data points are 30-s averages each representing the average mass concentration along 3 km of the flight track."

Thank you for the clarification. The sentence has been edited:

"The data points are 30 s averages with a horizontal spatial resolution of 3 km."

p.15175, line 17: for more clarity you can repeat the vertical interval you refer to

We have edited this sentence for clarity:

"BC mass mixing ratios increased by a factor of 6 with increasing altitude in the ABL and across the boundary layer transition up to 750 m in the free troposphere on 18 and 21 April."

p.15175, line 22: Is this meant to be a new paragraph which is not visible due to current formatting? If not, you should start a new one here.

A new paragraph is not necessary here because the statement at line 22 is a direct consequence of the remarks in the first three sentences of the paragraph.

p. 15176, line 22: change "... limited anthropogenic..." to "... limited local anthropogenic . . ."

This sentence has been edited:

"Since there are limited local anthropogenic sources . . . "

p. 15177, line 22: there is a "-" between 750 and m

The hyphen has been removed.

p.15180, line 11: change "... to be a 10 times..." to "... to be 10 times ..."

Done.

p. 15180, line 19: change ". . . less likely continuous as modeled and more likely. . ." to ". . . less likely continuous as modeled but more likely . . ."

This sentence has been rewritten:

"A possible reason for this discrepancy may be that the physical mixing processes are less likely continuous as modeled but more likely episodic as the temperature difference between the sea-ice and open leads enhances the mixing."

*p. 15181, line 5ff: "The meteorological surface data at Barrow indicate an average *5.3-cm* snow accumulation with measurable snow *an* average of 18 days in April." Remove the "-". I don't understand the meaning of this sentence.*

The hyphen has been removed and the sentence has been rephrased for clarity:

"We then use the climatological April mean snowfall of 5.3 cm at Barrow to calculate the average BC mass in the snow based on the dry deposition fluxes from the box model."