18 September 2010

Reply to Reviewer #3 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

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Reviewer #3 (12 August 2010):

General:
BC is an important contributor to warming in the Arctic. Most BC measurements in the Arctic have been done at surface stations even though for estimating the atmospheric heating information on the concentrations aloft are of utmost importance. This paper presents vertical profiles of BC measured with a new method, the SP2, in Alaska and north of it. The paper is well written, and figures are mainly clear. The authors have also used a simple model for estimating mixing between BL and FT and dry deposition. This certainly adds value to the paper and I can recommend it to be published in ACP after some additions and modifications.

Detailed comments:
P15168, Abstract, L18 – 22. Too long a sentence and cannot actually be understood without reading the section in the actual article. I suggest splitting it and adding some explanation, one sentence?

    We have restructured this part of the abstract describing the BC–O_3 correlation to convey the ideas 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."

P15170, L11- "While BC is observed in snow at sites throughout northern high latitudes (e.g., Hegg et al., 2009), scattering aerosol such as sulfate may be preferentially scavenged leaving behind BC aerosol suspended in the atmosphere." References? This is something that requires some explanation. Fresh soot aerosol gets coated with both inorganics and organics very quickly – typically within minutes or hours, externally mixed soot is there practically only in the immediate vicinity of traffic or biomass burning – when the BC particles have gone to the Arctic they most probably are internally mixed with scattering material and the statement that "leaving BC aerosol suspended" is

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somewhat hard to believe. Or does the SP2 show that there were externally mixed BC particles?

A brief mixing state discussion with references has been added to the introduction in response to this comment and another reviewer's suggestions. The relevant paragraph has been expanded 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)."

P15173 L8 – " the profiles obtained with the SP2 generally show lower BC mass loadings compared to those from an aethalometer during the Arctic Gas and Aerosol Sampling Programs (AGASP) campaigns conducted in 1983 and 1986." Will you please add some numbers, i.e., actual concentrations from the other campaigns and yours, rather than telling that "generally show lower BC" and let the reader to start digging the papers and numbers. Here it would be good to give some comparison of the concentrations.

We have rewritten this paragraph to include the quantitative results from the aethalometer measurements during AGASP-I and –II:

"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)."

P15178, L10 – " In (ii), the black points embedded in the main body of data points for 40<O3<55 ppb and 30<BC<200 ng kg−1, the correlation is a mixing line between the air masses in the ABL and the free troposphere." I cannot really see these points in Fig 7. There is so much data in the plot, it is messy, even though you have used the color coding. I would suggest separate plots, for example 7A, 7B, 7C where there could be one with all data and then separated to different altitudes.

This is a good suggestion. Figure 8 (formerly 7) has been modified to add a panel to explicitly show the O_3-BC correlations mentioned in the text. The paragraph that describes Figure 8 and the link between ODEs and dry deposition of BC mass has been partitioned into 2 paragraphs and rewritten accordingly:

"The O_3–BC correlation for all the flights and the entire sample altitude range is shown in Fig. 8A. Ozone is positively correlated with BC for the main body of points (i.e., O_3 > 40 ppb, BC > 30 ng kg^{-1}), associated with biomass-burning plumes and anthropogenic pollution. Two other positive correlations are shown in Fig. 8B for the data in the ABL (red points) and across the boundary layer transition (blue points). In the ABL, O_3 and BC mass loadings are well correlated in ozone-depletion events (ODEs). Also, note most of the data in the ABL were acquired during ODEs. A different correlation (blue points) is observed along a shallow mixing layer at the top of the ABL for 40 < O_3 < 55 ppb and 30 < BC < 200 ng kg^{-1}, corresponding to a mixing line between the ozone-depleted air masses in the ABL and the pollution in the free troposphere. This mixing line corresponds to the largest BC vertical gradient which was usually observed between 300 and 750 m altitude. The maximum altitude shown in Fig. 8B is 600 m because there is a lot of variability between 600 and 750 m altitude that obscures the correlations.

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."

The figure caption has also been updated to reflect the panel addition:

"**Figure 8**. Scatter plot of ozone and BC mass for 5 flights (12–21 April 2008) during ARCPAC for (A) the entire sample altitude range and (B) only up to 600 m altitude to explicitly show the different O_3–BC mixing lines in the ABL (red) and across the boundary layer transition (blue). The data points are 30 s averages and color-coded by altitude."

P15178, L17 – Here is the hypothesis of BC dry deposition. I have no objections but I suggest you also consider this: how about if the soot particles are actually internally mixed with some more hygroscopic material, such as sulfates, nitrates etc. At low altitudes RH is probably larger, especially above the leads. Then hygroscopic particles would grow and that would increase deposition velocity. Later, on lines 22 – you discuss the size distributions. I really would like to see them here, including the modal parameters.

We explored this interesting hypothesis by calculating the sedimentation velocities for internally mixed BC particles. We assume T = 250 K, P = 1000 hPa in the ABL. We also assume internally mixed BC particles have a density of 2 g/cm^3 which may be an overestimate especially if the particles are hygroscopic. Then, we calculate that spherical 0.2, 0.5, and 1 μm internally mixed BC particles fall at 2, 7, and 16 m/day. The mixing timescales in the Arctic boundary layer are probably on the order of 1 h which means that for a 300 m deep boundary layer, the mixing timescales dwarf the sedimentation rates of submicron BC-containing particles. The boundary layer turnover timescales would have to be much longer and the BC-containing particles large enough (up to 1 μm diameter since all the SP2 data presented in this paper were sampled off an inlet with a 1 μm impactor) for sedimentation to play a significant role. However, there is no evidence for a significant mode of mass at these larger sizes in the accumulation mode.

We have added a sentence at pp. 15178, line 21 to explain that we do not believe sedimentation contributes significantly to the deposition of BC to the snow:

"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."

P15179, L19 – I would like to see the differential equations in the final paper. Is it just four differential equations? Please present them, other people may use the same and test them.

The system of coupled linear differential equations is now included in the text when the box model is introduced. The variables and constraints are discussed in the context of Figures 9 (formerly 8) and 10 (formerly 9). We have also restructured the text of this section in accordance with the other reviewers' suggestions:

"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:

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

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

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

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

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."

P15180, L28 – "The corresponding deposition fluxes are 170 to 1700 ng (m2 day)−1"
How did you get these numbers. Add a few lines of explanation, maybe as an equation, so that the reader may follow and possibly recalculate these numbers.

The deposition flux is the quotient of the BC removal and the time estimated for mixing between the free troposphere and the ABL on e-folding time basis. These

flux estimates have now changed with the revisions of Fig. 11 (formerly 10). This affects the extrapolation to the estimate of BC in the snow which has also been revised in the text. To make the calculations of the flux more transparent to the reader, we now include the derivation as an equation. We have also rewritten this paragraph to incorporate the new information from Fig. 11:

"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:

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

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, $\Delta BC \cdot f\left[1 - \left(1/e\right)\right]$. 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.

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. If we assume the boundary layer mixing timescale, given by $3k_{BL}^{-1}$, is ~1 h, then the model solutions indicate the free troposphere-to-boundary layer mixing timescales, denoted by k_{FT}^{-1}, are 2.2 to 16.7 h for the cases $k_{FT}/k_{BL} = 0.15$ and 0.02, respectively. The corresponding dry deposition fluxes then span a range between 5000 and 100 ng (m^2 day)$^{-1}$, respectively. If there had been more sampling at the boundary layer transition where the BC vertical gradient was the largest, it might have been possible to more tightly constrain the profiles in the box model and reduce this large range in deposition fluxes.

These flux estimates decrease for longer boundary layer turnover timescales in accordance with Eq. (5). A tracer-tracer correlation of O$_3$ and CO (not shown) indicates the biomass-burning plumes in the free troposphere 15–21 April generally did not mix into the ABL, except the synoptic-scale event on 19 April mentioned earlier, suggesting even longer free troposphere-to-boundary layer timescales than given by the box model results for a boundary layer turnover timescale of 1 h. 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. During the ARCPAC sampling period, the WP-3D aircraft may not have sampled these episodic exchange processes."

Several lines: you write about gradient when you mean a difference. If you write about

gradient, then you should present it as "concentration difference /altitude difference"
with the respective units.

Where applicable, we have changed the wording from "gradient" to "difference" to be more accurate. Otherwise, we have used the phrase "BC vertical gradient" in the text to be more clear we are referring to d(BC)/dZ.

Why do you use ng/kg as the unit of BC concentration? In a vast majority of BC literature
it is expressed as mass/volume.

We consider it important to express BC mass concentration in units that do not depend on the ambient pressure and temperature at which the measurement was made, especially when measured from an airborne platform. We report BC mass concentration as a mass mixing ratio in standard units of ng BC $(kg\ dry\ air)^{-1}$. To convert from ng/kg to ng/m^3 at STP (0ºC, 1 atm), the mass mixing ratio simply needs to be multiplied by 1.29 kg/m^3, the density of air at STP. Often times in the literature, BC mass concentration is expressed as ng/m^3 but it is sometimes not clear whether the measurements were reported at standard (i.e., STP) or ambient conditions. For these reasons, we have not changed the units of BC concentration in the text.