18 September 2010

Reply to Reviewer #2 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 #2 (31 July 2010):

This study reports vertical measurements of black carbon, carbon monoxide, and ozone conducted during 5 flights in April 2008 over the Alaskan Arctic. The BC measurements were conducted with a sophisticated Single-Particle Soot Photometer (SP2) instrument. The authors conduct various analyses on the data to explore and characterize the nature of Arctic BC deposition, for which much research is needed.

Overall, this is a useful contribution to the literature and the manuscript is well-written. The authors go beyond simply reporting the measurements and present informative interpretations of the data. I recommend publication in ACP after these minor issues are addressed:

General comments:
Please include the duration of the flights and times of day that they took place. Are there any diurnal cycles in Arctic boundary layer processes that could have biased your interpretation of the measurements?

> All of the flights, between 7.5 to 9 hours in duration, occurred during the day with the time of flight midpoint generally occurring a couple hours past solar noon at Fairbanks. If the diurnal cycle of solar insolation affects the boundary layer turnover timescales this time of year, we do not anticipate any significant biases since the sampling over the sea-ice was generally the same time of day and latitude range. A sentence has been added at p. 15171, line 10 to include the flight information:
>
> "All of the flights were 7.5 to 9 hours in duration and occurred during daylight hours."

The explanation for why O3-BC correlation during O3 depletion events is evidence for BC removal by dry deposition needs more explanation/development. The following sentence is one example (p15178,15): "The general theory is that Br2 is released into the atmosphere from the brine during sea-ice formation so the correlation between O3 and BC mass suggests BC particles have been preferentially removed by contact with the snow." This suggests a link between sea-ice formation and BC removal, but what is the

physical mechanism? Does increased surface roughness from snow or sea-ice enhance near-surface turbulence, which increases the dry deposition rate? Please elaborate. Related to this comment, what value to you assume or calculate for the "removal efficiency factor" (p15180,2), and how do you justify it?

In this section of the paper, we propose a general removal hypothesis based on the BC–O_3 correlation in ozone depletion events (ODEs). The positive correlation does not suggest there is a common removal mechanism for ozone and BC but does suggest there is a common denominator in the removal processes for these species. We infer from this correlation that the common denominator is the surface. Bromine is released in regions of new sea-ice formation and catalytically destroys ozone. When a BC particle actually comes into contact with the surface, we expect it to be physically removed. The observations were generally made in the region of open leads and the surface roughness likely does influence the probability that a BC particle contact the surface but we cannot comment on the role of open leads because we cannot conclusively compare lead versus non-lead cases with this dataset as mentioned at p. 15177, lines 16-18.

The removal efficiency is not a true removal efficiency. Rather, it is an effective removal efficiency and represents the fraction of particles that actually come into direct contact with the surface within the assumed boundary layer e-folding turnover timescale. The removal efficiency is constrained by the observations. Section 4.2 has been rewritten to reflect these and other details.

Figure 8 (formerly 7) has been partitioned to explicitly show the O_3-BC correlations mentioned in the text and facilitate the discussion of the link between ODEs and dry deposition of BC mass. The paragraph that describes Figure 8 (formerly 7) has been separated 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."

Section 4.1 and Figure 6: Correlations are discussed qualitatively. It would be helpful to include statistics of these correlations (e.g., R^2 values) to make the analysis more rigorous.

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

"**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."

We have also added text to the body of the paper to discuss the linear regressions:

"As shown in Fig. 7, BC mass loadings and CO values were generally well correlated in the free troposphere for the 21 April flight. BC and CO were also correlated on 12 and 18 April but we only show the data from 21 April because the dynamic range in CO is largest on this flight. The slope of the line through the main body of points (black) is 4.58 ± 0.06 ng BC (kg dry air)$^{-1}$ (ppb CO)$^{-1}$. The data have been segregated by altitude to highlight the data over the sea-ice below 750 m altitude, the general altitude range for the observed BC vertical gradients as shown in Fig. 3 and Fig. 6. The lower altitude data fall off the free troposphere BC–CO line consistent with a process of BC removal in the ABL. The slope of this line (red), 2.59 ± 0.10 ng kg^{-1} ppb^{-1}, is less than in the free troposphere but an interpretation of the difference is not possible without detailed knowledge of the air parcel history."

p15172,14: Can you provide a brief description of how your measurements compare with those conducted for ARCTAS (Jacob et al, 2010)? I see that this paper is also in ACPD, and would be interesting to know the level of agreement (if described qualitatively) between measurements conducted during the same period with an identical instrument.

Jacob et al. (2010) provide an overview of the NASA ARCTAS campaign but do not show SP2 BC measurement comparisons. McNaughton et al. (2010) show comparisons between the SP2 measurements on the NASA DC-8 (Kondo), NASA P3-B (Clarke), and NOAA WP-3D (Fahey) but this work is still in preparation. The SP2 data from ARCTAS/ARCPAC are also shown in Figure 10 of Koch et al. (2009) as composite profiles. Wingtip-to-wingtip intercomparisons between aircraft were performed during the campaigns and the results suggest the SP2 data agreed to within stated uncertainties. These results have not yet been published so we include here a private communication to Gao Chen who prepared the intercomparison results. The paragraph mentioning the NASA measurements has been expanded to include Koch et al. (2009) and summarize the intercomparisons:

" . . . In addition to the measurements presented in this work, two other SP2 instruments aboard the NASA DC-8 and P3-B research aircraft collected single-particle BC data in spring 2008 during the NASA-sponsored Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS) campaign (Jacob et al., 2010). The NASA aircraft sampled biomass-burning plumes less frequently than the NOAA WP-3D. Also, the NOAA WP-3D probed the ABL around open leads north of Alaska for a longer time than either NASA aircraft that sampled a larger geographical area of the Arctic. The SP2 measurements of BC mass loadings generally agreed to within stated uncertainties during wingtip-to-wingtip intercomparisons between the NOAA and NASA aircraft (G. Chen, private communication). The Arctic vertical profiles from all the SP2 measurements are shown in the context of a measurement-model intercomparison in Fig. 10 of Koch et al. (2009)."

Specific comments:

abstract, sentence spanning lines 18-22 is awkward.

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."

p15169,20: Explain why colder temperatures and weaker insolation imply longer residence times for aerosols (specifically BC, for which photochemistry plays less important of a role).

In the Arctic, aerosol and chemical tracer abundances are governed by temperature and solar insolation. Chemical tracers are affected by colder temperatures (reaction rate dependence) and weaker insolation (photochemistry). Aerosols are also affected by colder temperatures and weaker insolation that lead to stratification and inhibit wet

removal. The paragraph describing the BC buildup in the Arctic has been rewritten per the suggestions of this and another reviewer. Please see the response to the comment below for the revisions.

p15169,20: The statement "high-latitudes are isolated from lower latitudes in winter...induces a weak meridional barrier to transport" seems contradictory to me. A "weak" barrier would seem to imply _less_ isolation. Please clarify this description.

The polar jet can be considered a barrier to meridional transport and is one reason why midlatitude and polar air masses generally have different tracer and aerosol characteristics. The isolation is relatively weak, however. Since the aerosol and tracer abundances are enhanced in winter and early spring compared to summer more due to the colder temperatures and weaker insolation, to avoid confusion we have decided to remove this sentence about the meridional barrier from the text. The paragraph as a whole has been restructured at the suggestion of another reviewer and is reproduced here:

"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."

p15170,4: "blend" -> perhaps "partitioning" ?

We have adopted "partitioning" because we agree this word choice better describes the relative contributions from anthropogenic and biomass-burning influenced air masses.

p15171,19: Does the range of particle sizes measured by the SP2 refer to the particles containing BC, or just the BC component?

The SP2 measures the refractory BC mass of BC containing particles in the 90 to 600 nm BC core size range. Another custom detector measures the size of internally mixed BC particles but that data is not analyzed in this work. The text has been edited to clarify the size range for data used in this paper:

"For the data presented here, the SP2 detected BC core sizes in the range 90 to 600 nm volume-equivalent diameter . . . "

p15173,2: "This important observation constrains global aerosol models": This

statement needs refining. Although very important, these measurements (spanning a limited spatial domain over 5 days) must be augmented with other data to truly constrain "global" aerosol models.

This sentence has been edited to note these observations can be used to constrain models when placed in representative context:

"It is worth noting here that global aerosol models generally underestimate advection of BC or overestimate removal of BC during transport to the Arctic (Koch et al., 2009). When placed in a representative context, these important observations can be used to apply bounds to physical processes in models."

The two paragraphs beginning at p15175,17 lack coherence and context. Please polish them before publication.

These paragraphs have been edited extensively through specific comments by this and the other reviewers and are reproduced here:

"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. Enhanced CO values of 180 to 220 ppb at the top of the ABL during this period indicate the prevalence of biomass-burning plumes in the free troposphere and suggest the BC gradient is attributable to both advection and depletion. To avoid the complication from the advection of biomass-burning layers aloft, we quantify the BC removal based only on data from 12 April, in the aged Arctic air mass before the biomass-burning plumes reached the Arctic. The time series of altitude, latitude, O_3, CO, BC mass, and sea-surface temperature (SST) for the 12 April flight are shown in Fig. 5. Sea-surface temperature is a remote sensing measurement of the temperature at the sea-ice or open water surface when the aircraft is a couple kilometers or less above the surface. The spikes in the SST data in the shaded regions of Fig. 5 indicate the aircraft is flying repeatedly over the sea-ice and open leads. We include O_3 data here because O_3 is typically depleted in the ABL over the snow and ice due to bromine-catalyzed O_3 destruction (Simpson et al., 2007) coincident with the observations of BC removal. Note the nearly complete removal of O_3 in the second and third segments. This correlation will be discussed in more detail in the next section in the context of the BC mass depletions.

Low-altitude profile segments for the shaded regions in Fig. 5 are shown in Fig. 6 for BC, CO, O_3, and potential temperature. The shaded region in Fig. 6 marks the ABL and the top of this layer is defined as the lowest altitude point above the surface where the vertical derivative of the potential temperature begins to increase which is often coincident with a weak low-level jet, generally at ~300 m altitude. In both Fig. 6B and C, there is a positive vertical gradient in BC mass loading in the ABL which is well correlated with ozone depletion and generally occurs in the vicinity of open leads in the sea-ice. As mentioned earlier, this gradient occurs while CO is unvarying. In Fig. 6B, the spiral ascent (red) and slantwise descent data (blue) are shown separately because of the variability in the free troposphere over the

geographical area of the profiles. In Fig. 6A, the limited BC data in the ABL prevent any conclusions from these particular profiles. 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."

p15174,5: "minimum observed altitude" -> "altitude of minimum concentration"(?)

We refer to the minimum altitude of the measurements here. This has been clarified:

"BC mass mixing ratios increase by up to a factor of 6 from the minimum altitude of the WP-3D in the ABL up to 750 m in the free troposphere, . . .

p15174,13-20: This passage is a bit awkward.

This portion of the text has been edited for clarity:

"Since BC aerosol and CO are both byproducts of incomplete combustion, they are often well correlated in the troposphere (e.g., Spackman et al., 2008). In the springtime Arctic, background values of CO are ~160 ppb (Brock et al., 2010). As shown in Fig. 3 on the 12 April flight in the aged Arctic air mass, background values of CO persisted throughout the lowest 2 km while BC mass increased by a factor of 2 to 3 from 100 to 500 m altitude. The observed gradient in BC with respect to constant values of CO indicates that the lower BC mass loadings in the ABL cannot be explained by air mass advection in which a cleaner air mass with less BC and CO was mixed into the ABL from the free troposphere at an earlier time."

p15174,26: "evidence for particle removal in the ultrafine and fine modes"... but no evidence for removal in coarser modes?

The coarse mode was measured during ARCPAC. This mode was dominated by sea salt in the ABL making it difficult to evaluate the removal of other aerosol types, such as dust. No change has been made to the text.

p15174,29: "pushing the Arctic air further north": What is the evidence for this? Could the air have been displaced vertically and/or southward instead?

Brock et al. 2010 show the evidence for the meteorological shift and Fuelberg et al. (2010) provide the detailed meteorological context. The sentence has been simplified and the 2 references added to the text:

"Polluted midlatitude air was advected into the Alaskan Arctic and was sampled from 15 April through the remainder of the campaign (Brock et al., 2010; Fuelberg et al.,

2010)."

p15175,9-10: 200ppb CO criterion for defining anthropogenic or biomass-burning plumes: This seems somewhat arbitrary, especially in situation where CO and BC become decoupled. Is this threshold used elsewhere? Why exclude BC from the defining criteria of pollution/burning-affected plumes? (It is not even clear to me where this threshold is applied in the study, so perhaps it is not relevant).

The 200 ppb CO criterion was inadvertently left over from an earlier version of the manuscript and is not used in the analysis. The sentence has been edited to support the claim that the ABL is decoupled from the free troposphere on the timescales of the measurement campaign:

"In fact, less than 1% of the data from air sampled in the ABL during ARCPAC are characterized by CO values greater than 200 ppb, a value typical of biomass-burning plumes sampled in the free troposphere."

p15176,17: "BC vertical gradient approaching a factor of 5": Define "gradient" and how the factor of 5 applies. Is this the spread between minimum and maximum values?

"BC vertical gradient" is simply the difference between the minimum BC mass loading at the bottom of the profile in the ABL and the maximum usually at 750 m above the boundary layer transition. We describe this more accurately:

"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 on 18 and 21 April."

p15175,21: "support this conclusion": Do the enhanced CO values support the conclusion that the observed gradients _can_ or _cannot_ be attributed to depletion?

The enhanced CO values suggest the BC gradient shown on 18 and 21 April in Figure 3, during the period with influence from biomass-burning plumes in the free troposphere, is attributable to both advection and depletion. This was not the case on 12 April in the aged Arctic air mass when the CO values support the conclusion that the gradient can be fully attributed to depletion. This and preceding sentence have been edited:

"Enhanced CO values of 180 to 220 ppb at the top of the ABL during this period indicate the prevalence of biomass-burning plumes streaming through the free troposphere and suggest the BC gradient is attributable to both advection and depletion."

p15176,15: "estimate BC removal at 15 ng/kg." Over what time, or at what rate, did this removal occur? (i.e., It may be more informative to report the removal with a time dimension).

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."

p15177,1: "Sampling...may play a role": It is not the "sampling" that plays a role.

This sentence introduces the role of open leads in BC removal in the ABL and has been restructured accordingly:

"Open leads emit heat and moisture to the Arctic atmosphere and both may influence BC removal in the ABL."

p15177,12: Is there a connection between the diamond dust and the moisture source from the leads? If so, you may want to clarify this with wording such as "resulting diamond dust".

Yes, the open leads are a moisture source and may lead to the formation of diamond dust. The text has been edited to clarify the connection between open leads, diamond dust, and possible impaction scavenging:

"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)."

p15178,25: For a mode of 160nm, would the fraction of BC mass missed by the SP2 be much different from the 10% cited earlier for a mode of 200nm? (presumably it would be less).

This is a good comment and is a major reason why it is important for the SP2 user community to be clear about measured size distributions and unmeasured mass. A small error (< 10%) in reported BC mass loadings may be caused by changes in the size distribution at the level observed here with the given configuration of the SP2. There is some variability in the mode (160–200 nm) of the size distributions in the Arctic free troposphere generally associated with biomass-burning plumes but we prefer to apply an overall 10% correction to the dataset here because, in practice, it is not possible to apply different correction factors to segments of the data where

particle statistics are poor. We have edited the instrument section at p. 15171, line 24 to add that variability in the size distributions may introduce a small error:

"However, this may introduce a small error (< 10%) in the reported BC mass mixing ratio due to variability in the mode of the size distributions."

p15178,29: "On the other hand, the size distributions support the CO data that indicate similar sources across the boundary layer transition": How, specifically, do the size distributions support inferences made from the CO data?

In response to the reviews, we have added a new figure (Fig. 4) showing the size distributions of BC in the free troposphere and ABL in the aged Arctic air (12 April flight). The text discussing the size distributions has been moved to paragraph 2 of Sect. 3.2 in support of the argument for removal of BC in the ABL. Specifically, the modes for both these size distributions is ~160 nm suggesting the air masses in the ABL and free troposphere have similar sources. The revised text is reproduced here:

"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."

Figure 5: Using log(altitude) or pressure as the vertical coordinate could improve clarity of the boundary layer data.

We experimented with the log(height) as a vertical coordinate but prefer a linear altitude scale for overall clarity in the figure.

Figure 6: Please include correlation statistics for these data.

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 loadings 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."

Figure 9: Are all k_BL values the same in this box model?

Yes, the values of k_{BL} are the same between all the boundary layer boxes. This is made clearer in the text with the addition of the system of coupled linear differential equations to the body of the text, as requested by another reviewer, and the statement:

"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."