Manuscript Review

Airborne investigation of black carbon interaction with low-level, persistent, mixed-phase clouds in the Arctic summer

M Zanatta et al.

General Comments

The primary objective of this study is to evaluate rBC properties in cloud hydrometeor residuals and document how these compare under varying synoptic patterns and cloud conditions. A lot of text is created documenting these patterns and conditions, and the authors are quite thorough in the detail that they include describing these patterns and conditions; however, by the end of the manuscript, it becomes quite clear that they are unable to link differences in the observed rBC mass concentrations, size distributions or mixing state to any atmospherically relevant parameter. Put a different way, neither the synoptic patterns or microphysical properties of the clouds are obviously correlated with the rBC properties. Hence, a major portion of the manuscript that describes these patterns and cloud could either be greatly abbreviated or be relegated to the supplement. This includes much of the effort that seems to have gone into describing the SID, SIP, Nevzorov and PHIPS and how they are processed to derive size, shape and concentration since neither LWC, IWC or cloud hydrometeor size distributions are convincingly link to cloud processing of the rBC. In fact, the only features of importance are the regions below, within and above clouds, regardless of their meteorological states. I strongly recommend that the paper be greatly shortened and focus primarily on the possible mechanisms by which the rBC became part of a cloud hydrometeor and only keep those meteorological (T, RH, winds) or cloud (size distributions) parameters in the manuscript that can make the case for or against how these parameters influenced how ambient rBC is processed by the cloud.

Instrumentation:

1. Why isn't ice being distinguished in the Small Ice Detector? The high resolution CCD array is specifically designed to allow distinguishing liquid droplets from ice crystals. Why isn't this done? Why make the assumption that the SID only is measuring liquid droplets?
2. Why aren't you looking at both Nevzorov sensing elements to get ice fraction? The Nevzorov consists of a cylindrical LWC hot wire and a heated cup for TWC.
3. Throwing out the first two channels of the CIP is not a generally agreed upon action and doing so is ignoring an important size interval for drizzle.
4. How were ice crystals distinguished in the CIP? It is difficult to determine circularity until you have at least 6 – 8 pixels, i.e. > 200 µm.

5. Why isn't Phips being used to fill in 45-75 fraction?
6. If the cloud concentration/thresholding is from SID, and smallest size is 10 um, you are likely rejecting a lot of cloudy air in you analysis.
7. The SP2 also provides a way to derive the temperature of incandescence to separate light absorbing rBC from other types of material that can absorb at 1024 nm and incandesce. Have the data been properly screened to be sure the incandescing particles are only rBC?
8. The references to Moteki and Laborde for calibration standards and procedures are not the best since Schwarz et al. (2006) was the original publication to which all other subsequent paper refer. Please update.
9. Line 204 "Hence, cloud particle residuals were representative of cloud condensation nuclei and/or ice nucleating particles (Mertes et al., 2005, 2007)." No, this ignores inertial scavenging (see below).

Data evaluation and presentation:

1. Show distribution by mass not by derived size. The process of deriving a mass equivalent diameter is highly uncertain because it assumes that all particles with rBC are homogeneous, spherical and constant density. Clearly this is not what in situ samples, captured on filters, reveal. Every where that the rBC distribution by equivalent diameter are shown should be number concentrations or mass concentrations by the measured rBC mass. The same trends and patterns should be seen as are seen with diameter, they will just have more of a physical meaning.
2. The current study looks at ratios of the rBC residual concentrations to the droplet concentrations and rBC residual concentrations to out of cloud rBC concentrations; however, a very important comparison here that is missing should be the comparison of the rBC residual concentrations ratios to the total number of residuals compared with the rBC concentrations out of cloud to the total aerosol concentrations below cloud and above cloud. Baumgardner et al. (2008) used this approach to argue that inertial scavenging was the most likely mechanism for putting rBC in cloud hydrometeor residuals (See below), a mechanism that is almost completely ignored in this manuscript.
3. Where are the representative size distributions from the SID and CIP?
4. These clouds, based on how they are being defined, were between 300 and 400 m in depth, so why did the analysis divide them into 5 layers? I stably stratified clouds like these, is it physically reasonable to think that the properties will have such fine structure?
5. Where are the wind data, vertical and horizontal? Was there vertical shear that would lead to entrainment and mixing? The authors elude to entrainment as a possible mechanism introducing the rBC to the cloud but what drives this entrainment?

Cloud definition:

1. This study defines a cloud as N_{Dro} > 10 cm^{-3} and LWC > 0.01 g m^{-3}. How was this this threshold arrived at and how sensitive are the results to this definition?
2. Since N_{Dro} is derived from the SID, and since the lower threshold of the SID is cutoff at 10 µm, in Arctic stratus clouds, this is likely eliminating a lot of cloud data and likely biases measurement depending if they are at the botton half or top half of cloud. This could be part of the reason that you seen differences in the rBC properties at the bottom or top of the cloud.
3. Where are the size distributions in cloud, compare cloud base to cloud top?

I strongly recommend that the authors read the paper by Baumgardner et al. (2008), who measured rBC in cirrus cystals over the Pacific Ocean, and concluded ***"Comparison of BC properties in the crystal residuals and cloud-free particles show that BC is scavenged by the crystals with an efficiency of as much as 44 ng of carbon per gram of ice water. The average number fraction of BC in crystal residuals was 40% greater than that of the particles in the nearby, cloud-free regions and, on average, the mass equivalent diameter of BC was 10% larger. An average of 24% of the ice crystal residuals contained BC compared to 17% in cloud-free air. The large, in-cloud variability of BC properties prevents a more rigorous proof of our hypothesis that inertial scavenging is a dominant mechanism for removal of BC by clouds. The lack of correlation between the inside and out of cloud concentrations, coupled with the differences between the number fraction and MED in the crystal residuals and the background aerosol, is consistent with the inertial scavenging process and the evidence is sufficiently compelling to warrant further study."***

This study, while sampling a very different type of cloud than the current one, makes the case that all the data points to inertial scavenging as the primary mechanism by which the rBC ends up in cloud hydrometeors. The same arguments can be made in this study. The very limited analysis of the "coating" suggests that these are uncoated or very lighty coated (read "lightly mixed") rBC particles. Laboratory studies, although admittedly sparse and with varying conclusions, generally conclude that fresh, or aged yet lightly coated, rBC are not hygroscopic, i.e. not good CCN; hence, their presence in cloud residual does not imply that they were the nuclei of the droplet. In addition, vertical motion by adiabatic lifting is unlikely to lead to supersaturations large enough to activate these particles, even if slight hygroscopic. The study by Baumgardner et al (2008) claims that that the higher concentrations of larger rBC, seen in their studies and in the present one, is most easily explained by inertial scavenging. If the authors of the current study followed the same methodology as Baumgardner et al., I think that they would like reach a similar conclusion.

To summarize, I think that this manuscript needs to be edited to decrease its size and focus on better explaining the results than are currently being done. I have raised a number questions that need to be addressed before I am ready to recommend this submission for publication.

References

Baumgardner, D., R. Subramanian, C. Twohy, J. Stith and G. Kok, **2008**: Scavenging of Black Carbon by Ice Crystals Over the Northern Pacific, *Geophys. Res. Letters*, 35, L22815, doi:10.1029/2008GL035764.

Schwarz, J.P., R. S. Gao, D. W. Fahey,D. S. Thomson,L. A. Watts,J. C. Wilson, J.M., Reeves, D. G. Baumgardner, G. L. Kok, S. Chung, M. Schulz, J. Hendricks, A. Lauer, B. Kärcher, J. G. Slowik, K. H. Rosenlof, T. L. Thompson, A. O. Langford, M. Lowenstein, K. C. Aikin, **2006:** Single-particle measurements of mid latitude black carbon and light-scattering aerosols from the boundary layer to the lower stratosphere, *J. Geophys. Res.* 111, D16207, doi:10.1029/2006JD007076.