

# Responses to Referee # 1

## General Comments

This manuscript uses the CALIOP satellite instrument to quantify the spatial, seasonal and interannual variability in aerosol burdens throughout the Arctic troposphere. I commend the authors on their excellent work – this paper provides an extremely thorough analysis and the first truly comprehensive picture of the seasonality and variability of Arctic pollution above the surface. It also provides context for the representativeness of long-term ground-based sites and short-term aircraft campaigns that have previously been the basis of most of our understanding of Arctic aerosol pollution. In addition, this work will serve as an important benchmark for model simulations of Arctic pollution. The manuscript is generally well written and well organized, and I recommend it for publication in ACP following a few minor revisions.

## Specific comments

Title – perhaps satellite instrument after CALIOP for those who are not familiar?

*Ok*

4866 line 14: “Arctic aerosols” – is this specific to sulfate? Inorganics? Would be better to specify here since lots of work has been done on BC.

*“Arctic aerosols” changed to: “sulfate and black carbon aerosols”*

4866 line 16 and line 23: “In addition to anthropogenic” – much of the biomass burning is anthropogenic in origin, especially the agricultural burning mentioned in line 23. Perhaps change “anthropogenic” to “fossil fuel burning”.

*We changed that according to the suggestion. We also added a sentence discussing early observations of lofted layers of mineral dust and added Rahn et al.(1977) reference.*

4867-4868: This whole section seems like it would fit better towards the beginning of the introduction, when the authors are discussing the aerosol sources. When I first read that section, I thought it was missing all the IPY results. Turns out they are all here, but seem to duplicate some of the other information.

*We moved the paragraph discussing the IPY results earlier in the introduction.*

4868 lines 11-12: It’s not clear to me why CO is being discussed since the manuscript is about aerosols.

*We think references that discuss Arctic CO are relevant to our work as CO is a meaningful proxy for long-range transport of anthropogenic pollution. CO often correlates with aerosols, and informs on the degree of aerosol scavenging pollution plumes undergo and provides information on the prevalent transport pathways during the spring of 2008.*

4868 lines 13-15: a reference is needed for these numbers.

*The missing reference was Wang et al. (2011) which we included.*

4868: It would be good to refer to Wang et al. 2011 somewhere in this section as there is very little discussion here on the black and organic carbon aerosols which are important to the spring aerosol budget. Also Bourgeois and Bey 2011 (which is cited elsewhere but is relevant to source descriptions). Also seemingly missing is a discussion of results from the summer IPY campaigns (e.g. Schmale et al., 2011).

*We included references to Wang et al. (2011), Schmale et al. (2011) and Bourgeois and Bey (2011), checked references and re-organized the paragraph.*

4870 lines 25-27: Where was this validation for? The Arctic or elsewhere?

*We clarified this point and revised the sentence to: "The CALIOP 532 nm calibration has been validated against aircraft measurements by the NASA Langley airborne High Spectral Resolution Lidar (AHSRL) instrument for a wide seasonal and latitude range covering diverse aerosol and cloud conditions."*

4874 lines 20-25: What were the results of the comparison? Should be stated at the end of this paragraph.

*A whole sentence was missing from the typeset version (highlighted in green below). We apologize for the oversight and we will make sure it will be included in the final version:*

*"The two populations differ in the mean by RH=3% (daytime higher). We find that the random variable representing the standardized difference of the two means falls within one standard deviation from zero, indicating that the two populations can be considered statistically indistinguishable."*

4875: The terminology is confusing here. On page 4871, the authors define "mean backscatter" (beta with bar) as  $f \times \beta$ . But here it just seems to be the average of individual beta values. I was confused as to when "mean backscatter" means  $\bar{\beta}$  and when simply an average of beta. Could  $\bar{\beta}$  be given a different name? E.g. "gridded backscatter"? Or something else? Or at least not defined until after this discussion?

*We changed the wording to avoid confusion:*

*"We also extract the mean backscatter ( $\beta$ ) and extinction ( $b_{ext}$ ) of the detected layers, along with their standard deviation. The "gridded extinction"  $\overline{b_{ext}}$  is then defined as the product between the aerosol detection frequency and the mean extinction of the detected layers,  $f \times b_{ext}$ . The "gridded backscatter",  $\bar{\beta}$ , is defined similarly as  $f \times \beta$ ."*

4875 line 15: This was defined for points south of 71N – any test of whether it is consistent north of 71N? I am not suggesting it to be changed, just curious whether the consistency for the whole region considered can be quantified.

*We chose those latitudinal boundaries because both daytime and nighttime conditions exist at those latitudes between September and March. At latitudes poleward of 71N it is not possible to test its consistency as illumination conditions are persistently either daytime or nighttime.*

4877 line 1: Why the different distances for Barrow and Alert? Does this reflect a difference in latitudes affecting number of satellite overpasses? Again, not suggesting a change but a justification would be nice.

*That is correct. The box around Alert (82.5N) can be made smaller because many more satellite overpasses occur in its vicinity whereas the lower latitude of Barrow requires the box to be slightly enlarged to assure a large enough number of satellite overpasses. This has been clarified in the revised text.*

4877 lines 24-25: I don't understand what is meant by "the annual-mean value chosen by the CALIOP algorithm"

*The CALIOP algorithm chooses the lidar ratio based on which aerosol type a layer corresponds to, and use it in the inversion of extinction from the measured attenuated backscatter. We calculate the mean lidar ratio chosen by CALIOP of all aerosol layers detected as a function of month in the Alert and Barrow boxes. It is fairly constant around 40 sr.*

4878 lines 17-24 and Fig. 5d: On average, CALIOP at Alert overestimates the in-situ measurements in SONDJ and underestimates in FMA. I'm wondering whether there is any influence from the choice of gamma, which was chosen based on values from Barrow. I would expect there is more clean aerosol at Alert than at Barrow. Alternatively, perhaps the high altitude at Alert has an impact on the comparison? Again, not suggesting a change, but it would be nice to see some discussion of the causes, and perhaps a quick order of magnitude discussion of how a different choice of gamma would impact the Alert comparison.

*It is likely that gamma varies seasonally. For simplicity, we chose a constant values of gamma, so as not to make our results depending on largely unconstrained and arbitrary assumptions. In the revised manuscript we address the effect that a different choice of gamma makes on the MAM mean extinctions.*

4879 lines 22-23: If CALIOP can only detect the strongest haze events, why are the thickest plume excluded (lines 7-8)?

*In thick plumes, very high extinction values may occur at spatial scales that can be even smaller than the distance between consecutive CALIOP lidar shots (~330 m). These values can greatly affect the mean values in the observations. This exclusion wouldn't be necessary if CALIOP were sampled exactly along the flight track, but that would result in many fewer usable points.*

4881 lines 5-6: "Above 2 km" – it looks from Fig. 8 like there is also a winter to spring enhancement at around 1 km.

*That is correct. We have modified the text accordingly.*

4881 lines 11-12: It would be better for the units of backscatter to match between the text and the figure.

*Done*

4882 lines 19-21 and Fig. 4: I'm confused about the choice of boundaries for the European and Asian sectors. Why is Alaska included in Asia and not North America? And why does the European section extend so far east (typically would expect it to cut-off at the Urals, 60E). These counterintuitive choices should be justified.

*These sectors are slightly offset towards the east so as to capture the typical outflow of the source regions from which they take the name. This is described at the end of that first paragraph.*

4884 line 25- 4885 line 1: I'm a bit confused here. The authors seem to be discussing a minimum at 70-80N and a maximum at 60-70N. Can this be clarified?

*The opposite seasonality can be explained with a strong scavenging efficiency in the low arctic and the strong transport barrier that the polar front constitutes. Little polluted air from the south crosses the 70N boundary. The air that makes it across is cleaner (depleted of aerosols) because of the strong drizzle scavenging occurring between 60-70 N. We added a clarification by changing the sentence to:*

*"These very low summertime extinctions over the High Arctic could be associated with efficient wet scavenging that takes place during transport from lower to higher Arctic latitudes"*

Section 4.3: I think this section could precede Section 4.2. As I read 4.3 I found myself wondering why the high emissions in the Low Arctic aren't observed in the High Arctic, but that is generally explained in Section 4.2

*The two sections have been switched.*

4885 lines 23-24: This wording is confusing since Fig. 9c shows higher concentrations in the European sector than the North American sector. It could be rephrased to emphasize the change from the lower to mid troposphere (e.g. percentage enhancement from low to mid or something similar).

*The text has been modified to clarify this point.*

Table 1: Footnote "a" is only about the in situ extinction and doesn't need to be listed for the CALIOP column

*We apologize, this was a typesetting error and we will make sure it is corrected in the final version.*

Fig. 2: See earlier comments – is "Mean backscatter" on the x axis mean(beta) or beta-bar? If the latter, why isn't it affected by the day-night variations in f?

*It is the mean backscatter of the detected layers (beta). We corrected "mean backscatter" with "gridded backscatter" consistently with the text, which also helps distinguish beta from beta bar ("gridded backscatter")*

Fig.3 Colorbar needs unit label.

*Done*

Fig. 6: I'm not convinced this figure is necessary. The most important part – the observed profiles - are also shown in Fig. 7. The thresholds are described in the text. If they are really necessary, they could be added to Fig. 7, but I think that figure is fine as is.

*True, it could be merged with Figure 8 but it would make it too busy. We believe Figure 7 should be kept because it shows the observed distribution of observations with respect to the satellite thresholds, and helps the reader with its strong visual impact.*

Fig. 9: It would be a lot easier to read through the text and look at the figure simultaneously if the panels of this figure were rearranged to reflect the order in the text (first f, then e). This would mean the left column would be the High Arctic and the right would be the Low Arctic, with 0-2 km at the top and 5-8 km at the bottom. This would also match the altitude structure of Fig. 10. Note that this would also mean rearranging of Fig. 13.

*Both figures have been rearranged, captions and references updated.*

Fig. 10: Can the continent lines be made thicker (and/or the circles bigger)? It's very hard to distinguish the sectors discussed in the text, especially for the 0-2 km sectors. Also, is the dashed line the division between the low and high Arctic? If so, please add the caption.

*Continent boundaries were made thicker. We made a note in the caption that the 70 and 80 N boundaries are marked by dashed circles.*

Fig. 12: Is it possible to demarcate the boundaries between the years on the axis as well as in the figure? It would make it easier to figure out e.g. which Jan goes with which year.

*Next to each January on the x axis we added '07, '08 to facilitate reading.*

#### **Technical corrections**

4864 line 19: "observations factor of" should be "observations by a factor of"

*Corrected*

4864 line 27: "it remains" should be "they remain"

*Corrected*

4866 line 22: "in presence of" should be "in the presence of"

*Corrected*

4866 line 29: delete "have"

*Corrected*

4871 line 9: "value is" should be "values are"

*Corrected*

4872 line 24: "method cloud screening method" – delete first "method"

*Corrected*

4873 line 25: "7 months period" should be "7-month period"

*Corrected*

4879 line 16: "displayed" should be "displays"

*Corrected*

4881 line 9: "HRSL" should be "HSRL"

*Corrected*

4882 line 18: "low" should be "lower" (especially to help distinguish between Low Arctic and lower troposphere on quick reading)

*Corrected*

4884 line 8: "produces" should be "produced"

*Corrected*

4885 line 2: "relative CO" should be "relative to CO". In general I think the sentence would read more clearly as "... examined the efficiency of transport to the Arctic of BC relative to CO".

*Agree. We fixed that.*

### **References**

Bourgeois, Q., and I. Bey (2011), Pollution transport efficiency toward the Arctic: Sensitivity to aerosol scavenging and source regions, *J. Geophys. Res.*, 116, D08213, doi:10.1029/2010JD015096.

Schmale, J., et al. "Source identification and airborne chemical characterisation of aerosol pollution from long-range transport over Greenland during POLARCAT summer campaign 2008." *Atmospheric Chemistry and Physics* 11.19 (2011): 10097-10123.

Wang, Q., D.J. Jacob, J.A. Fisher, J. Mao, E.M. Leibensperger, C.C. Carouge, P. Le Sager, Y. Kondo, J.L. Jimenez, M.J. Cubison, and S.J. Doherty. Sources of carbonaceous aerosols and deposited black carbon in the Arctic in winter- spring: implications for radiative forcing. *Atmospheric Chemistry and Physics*, 11: 12453-12473 (2011).