Authors' response to referee #3's comments

Specific comments, and authors' responses:

1) In the paper it is still worryingly unclear the SP2 calibration procedure, which is essential in order to have realistic absolute values of rBC concentration. In your response it is written that the SP2 calibration was performed by DMT and that "We used a custom code written in R and CRAN R open-source software to filter broadband incandescence peaks and convert incandescence peaks to mass using the calibration relationships (supplied by Droplet Measurement Technologies)". So the question is:

1a) The DMT supplied the calibration coefficients (as it happens usually)? If yes which calibration material was used by the DMT during the SP2 calibration? If DMT supplied the calibration curve, how the calibration results reported in Figure S4 were used? Only for calculate the nebulization efficiency of the CETAC system?

There are two calibrations. The first calibration was performed by DMT used DMA mobility size selected Aquadag particles to estimate the single rBC particle mass in relation to broad-band incandescent light intensity. There has been some discussion regarding the variability of incandescent emissivity between different rBC materials – in particular Aquadag versus "as produced" fullerene soot (from Sigma-Aldrich). For the ice core research we have continued to use Aquadag for the broad-band detector relationship to rBC mass in order to be consistent with past ice core analyses. We acknowledge the differences between rBC mass as determined using Aquadag and fullerene soot as discussed by Moteki and Kondo (2010; *Aerosol Sci. Tech.* 44: 663–675). However in this case fullerene soot was determined to be a good standard with regards to the analysis of ambient rBC in urban Tokyo. We do not know whether it would be a good standard for rBC incorporated into the remote the polar ice caps. In many respects these regions are the antithesis of urban Tokyo or any urban atmosphere.

The second calibration, (performed, before, during and after the analysis) was used to determined the relationship between liquid rBC concentration and the aggregate response of the entire system over a constant time step. This is not merely a calibration of the nebulizer, but rather the entire system, including mass transport through the nebulizer and all the sp2 parameters including detector efficiency, gas flows etc. As with many other instrumental platforms which are calibrated against liquid concentration the assumption is that the standard and its matrix is a good approximation of the sample. This is one of the reasons that we tried to choose a standard with a similar mass distribution to the sample. The nebulizer efficiency of the CETAC nebulizer was determined as a day to day check of the nebulizer system stability and is not necessary for the calibration.

We have changed lines on page 3 of the supplemental to read:

"The nebulizer efficiency (~25%, defined as the percent mass of water aerosol exiting the nebulizer chamber into the heated section) was monitored gravimetrically to check the nebulizer stability."

1b) Moreover, how did you make the standard? Did you assume a 100% content of BC particles in the original standard? This is a key step in order to know the actual BC concentration in a solution. No EC (Elemental carbon) measurement was performed to ensure this (as done for the Aquadag and Fullerene in other studies)? Even for Aquadag the total content of refractory BC particle is not 100%. If you have used the "new" standard for making the calibration of the SP2 then it will be important to make and estimate of the possible degree of underestimation/overestimation of the absolute concentration values caused by the use of a different standard.

To prepare the standard we first determined the dry mass and refractory rBC by thermogravimetry. Thermogravimetry was chosen over the NIOSH thermal/optical EC analysis since it is typically referenced to sucrose and is not considered a primary analysis. With regards to the thermogravimetric analysis we determined that the liquid standard contains 7% refractory mass, here defined as the stable mass at 400 °C in air. We determined the rBC mass contribution to be 92.7% by subtracting the remaining mass after autoignition at temperatures above 450 °C. After determining the mass at 400 °C the temperature was increased to 480 °C for 40 minutes and then 580 °C for 40 minutes. As the temperature increased from 400 to 480 °C the bulk of the material passed through its autoignition and became incandescent. No visible emissions could be seen. We assume that the mass lost during autoignition was rBC. The mass remaining after 40 min at 580 °C was 7.3% of the refractory mass. Based on the results of the thermogravimetry we prepared the concentration standards by serial dilution in ultrapure water.

We have added further details on page 2 of the Supplement:

"Thermogravimetric analysis of the standard revealed the dry mass to be 7.06% of the liquid mass. The dry mass was stable in air from 200 to 400°C. We have recently analysed a fresh batch of the same pigment and found a comparable dry mass of 7.23% at 400 °C (for 40 min in air). After increasing the temperature to 480 °C (for 40 min) the material glowed red with no visible emissions suggesting an autoignition temperature between 400 and 480 °C. The temperature was increased to a final temperature of 580 °C for 40 min. We found no mass change between the 480 and 580°C stages. The remaining mass after the 580 °C stage was 7.3% of the dry mass. Based on the thermogravimetric analysis we concluded that 92.7% of the dry mass at 400 °C was rBC. Concentration standards were prepared by serial dilution of the pigment with ultrapure water based on the results of the thermogravimetric analysis".

1c) In the paper by Moteki and Kondo (2010), a comparison of SP2's responses to different calibration material is given. Please use their results to establish a plausible degree of variability of your absolute concentration values if you were to use other standards (just use their slopes variability and change your calibration curve accordingly).

We do not think that we can simply changing the slopes to match that of Moteki and Kondo (2010) as described by the reviewer. The correct approach would be to determine the optical and morphological properties of rBC in Greenland snow after it has been melted and then make a direct comparison with the Aquadag standard used by DMT. However, our guess is that if the Eboni standard was at the extreme end of the values from the single particle masses our results using the

standard could vary from fullerene soot by a factor of two. We do not believe this is the case however. Even if this were the case the trend in the time series would not change.

2) Please show the plot related to the change in sensitivity also using the aging curve, so that it will be much easier to understand for the reader where there could possibly be a higher degree of uncertainty in the results.

Response: We have exchanged the depth scale for the age scale in Fig. S6.

3) Why are you using the units "rBC (10...)" in the y-axis of the Fig. S4? Are you sure it is not "Incandescence Signal"? Please add the unit also in the plot of the calibration curve.

The units are 10^{-15} g rBC per sec. Or in this case 10^{-15} g rBC per 2 sccm (standard cubic cm per minute). The calibration *y*-axis units are all the same.

We have added the following to the caprion of Fig. S4: "The y axis units for all panels are femtogram rBC per sec which is equivalent to units of femtogram rBC per 2 sccm of air."

4) You should report the plot of the mass distribution in "dN/dlnDp", not in "dN/lnDp" (use the logarithm of the diameters in the ratios).

This was a typographical error. We have changed the label to"dN/dlnDp"