Response to the interactive comments of referee #1 (RC C1199):

First, we would like to thank the anonymous referee very much for the comments and suggestions. Below the original comments -shown in italics- answered point-by-point, which will be taken into account in the revised manuscript accordingly.

1. Ideally the nebulizer / desolvation system will not change the mass or size distribution of the particles in solution. The study found that the APEX nebulizer alters the particle size distribution the least. However, a significant problem with the APEX system is that it amplifies pulsations from peristaltic pumps. Self-aspiration can be extremely fickle and the uptake rate may vary from sample to sample. Other analytical systems that use the APEX include an internal standard to correct for uptake variations. Without an internal standard, how can self-aspiration be reliable for the coupled SP2 system? Self-aspiration may be stable for one sample, but not for another. Is there a way to ensure the sample introduction conditions match that of the external calibration?

The uptake rate of the APEX by self-aspiration is tested by measuring the sample uptake. The best way to ensure the sample introduction conditions to match that of the external calibration is to repeatedly compare the measurements of the external BC standards before and after analysis of the samples. The repeated measurements of the dilution series of BC standards for external calibration indicated that the “averaged” external calibration factors varied by ~22% for the CETAC and by ~8% for the APEX-Q within two months (see chapter 3.3 in manuscript). Since the weekly external calibration is applied to the samples analyzed within the very same week the variation in sample uptake between standard and sample are thought to be within the error. The main problem with the APEX system remains the clogging of the nebulizer capillary which results in no uptake but can be checked visually.

2. The study found that BC mass transport through the CETAC ultrasonic nebuliser was inefficient for particles bigger than 500 micron (spherical equivalent diameter). Are BC particles > 500 micron typically found in Arctic and Antarctic snow?

We do see BC particles in the size range >500 nm in the samples from Lomonosovfonna, a low-altitude Arctic site in Svalbard, similar to BC in snow samples from Colorado (Schwarz et al., 2012, 2013).
We cannot solve this question for Antarctica, but the transport pathways for BC reaching Greenland and low-altitude Arctic sites, such as Svalbard, are different. Greenland does not receive low-level transport and is instead more directly connected to transport from lower latitudes (AMAP, 2011), whereas low-altitude Arctic sites are more influenced by low-level transport of pollutants from high-latitude Eurasia (AMAP, 2011; Hirdman et al., 2010). This might result in a difference in BC particle sizes for Greenland, so that particle sizes larger than 500 nm do not occur in Greenland.

It is important to note that the BC particle sizes observed in snow is not thought to be only related to BC particle size in the atmosphere-freeze cycles and the post deposition thermal history are also thought to play a role. Further it needs to be mentioned that the mass absorption cross section (MAC) of BC particles smaller than 500 nm is much larger than for larger BC particles (see Figure 1 in Schwarz et al. (2013)). This means that even though some of the larger BC particles are not measured with the CETAC ultrasonic nebulizer, these particles have a much smaller MAC and thus have lower albedo relevancy.

References

AMAP: The Impact of Black Carbon on Arctic Climate, Arctic Monitoring and Assessment Programme (AMAP), Oslo, 2011.


Additional correction:

Page 3082, line 19: “...commonly done with a mass-selected Fullerene Soot...”

We changed this in the manuscript as follows:

“...commonly done with mass-selected Fullerene Soot...”