Author response to:

Review on "Assessing recent measurement techniques for quantifying black carbon concentration in snow" by J. P. Schwarz et al.

Anonymous Referee #1

Author response italicized.

This study evaluates the accuracy of two instruments that have been applied previously to measure BC content in snow: the Single Particle Soot Photometer (SP2) and the Integrating Sandwich Spectrophotometer (ISSW). This study provides a more thorough analysis of measurement uncertainty for these two instruments (as applied to BC in snow) than any previous study, and includes an assessment of how particle size distribution, co-presence of mineral dust and other scattering particles, and nebulizer efficiency can bias derived BC concentrations. This is a useful contribution to the literature, and the material appears suitable for this journal. This study, ideally published years ago, will provide helpful context for interpreting published measurements of BC in snow.

> *The authors appreciate the careful assessment and useful comments of Anonymous Reviewer #1.*

Addressing the following issues will help clarify and improve the study:
The uncertainty associated with nebulizer efficiency is slightly confusing to those un-familiar with the SP2. Presumably the mass of water that is aerosolized is known precisely. If particles within this water do not make it to the SP2, where do they go? Do they become trapped in the nebulizer? Or, is the uncertainty truly associated with the mass of water that becomes nebulized (3777,9)?

> *The complexity associated with determining nebulizer efficiency a priori as a function of particle size is significant, especially for the ultrasonic nebulizer mentioned in the text. This is why an empirical method is used to directly measure nebulizer efficiency. However, we recognize the confusion of the reviewer, and have expanded this section with the following sentences:*
>
> > Our work with the nebulizer revealed that different size particles were aerosolized with different efficiencies, with larger particles having a reduced likelihood to be passed to the SP2. We assume that these differences result from the internal geometry of the nebulizer, which requires sharp turns of the airflow that may act to strip the air of larger particles (see, for example Hinds, 1999). Based on separate tests of transport efficiency of different size particles in the SP2 (Schwarz et al., 2006), we believe that these losses occur predominately in the nebulizer. To address this issue, we adopt an empirical approach to the problem, simply by directly measuring the efficiency with which particles in liquid were nebulized and transported to the SP2, as follows:

and

> The instability in the rate of flow in the peristaltic pump is a negligible contributor to the total uncertainty.

The issue of "detecting" larger particles with the SP2 (3774,19) is also a bit unclear to me. Section 2.1.1 mentions that the SP2 was originally calibrated with particles over the range of 0.5-40 fg. The text seems to infer that larger particles still incandesce when they pass through the SP2. Assuming so, is the problem that incandescence events associated with larger particles were beyond measurement detection in previous studies, or were they measured but discarded? Section 2.1.1 goes on to describe the extended calibration technique for measuring large particles in this study. It would be helpful to state the calibration equation that was applied here, so the variable dependencies can be known precisely by readers.

Previously particles larger than the traditional SP2 limit were detected, but then discarded. We have clarified this with the following sentence:

> Note that normally, BC particles larger than the traditional detection limit of the SP2 are detected, but discarded due to their saturation of the SP2 detectors.

And we have included explanation of the gain settings we used on the SP2 detectors to ensure retrieval both of particle BC mass, and color-ratio for large BC particles.

Additionally, to address the reviewer's point about the calibration equation, we have added the calibration equations explicitly in the text.

It is mentioned in several places that BC particles in snow are larger than those in the atmosphere, but little explanation is given for why. I am not sure that this result has been previously reported, and it may be important, e.g., for the optical properties of BC in snow. It would be helpful if potential explanations can be given for this observation, though reasons may be unknown. Is the primary attribution of this that: "individual freeze/thaw cycles cause the agglomeration of a small fraction of BC mass into larger sizes, without dramatically shifting the underlying BC size distribution." (3780,16)?

We also recognized the scientific significance of this observation, and have prepared a second manuscript describing the implications of the size shift and discussing some of the possible mechanisms. We feel that this topic is better treated in a separate manuscript that has less of the technical information relevant to the ISSW/SP2 comparison that forms a large component of this manuscript.

3784,5: "Undercatch was 38% based on refiltering postfilter liquid using finer-mesh (0.2um) nuclepore filters." Does this assessment assume that the BC collected on the 0.2um filter represents all BC missed by the 0.4um filter? Since a substantial portion of

BC particles in typical size distributions are smaller than 0.2um, I would think some C1704BC particles may also pass through the 0.2um filter.

We have added the following sentence to address this point:

> We assume that the 0.2 μm filters collects all the particles missed by the 0.4 μm filters, because limited tests with 0.1 μm filters show no difference to the 0.2 μm filters.

3786,8: "The monodisperse nature of the PSL-containing test standards produced strong wavelength dependence in the inferred ISSW absorption, hence these results are not quantitatively presented". - I don't see why this would preclude a quantitative assessment of the ISSW absorption. Please elaborate on this, or include some assessment of the magnitude of bias in ISSW-derived BC concentration that resulted with inclusion of the PSL standard (e.g., in the text and also potentially in Table 1 and Figure 3). Two sentences after this statement includes references to "size of excursions" and "significantly bias", implying that some sort of quantitative assessment was already performed.

We have modified the following sentences to clarify why the PSL results do not immediately relate to the ISSW uncertainties from more typical scattering particulate on the filters, and to add some quantification to support our statements about significant bias:

> The monodisperse nature of the PSL-containing test standards produced strong wavelength dependencies in the inferred ISSW absorption, at wavelengths used to separate BC and non-BC absorbance contributions. As this type of behavior is not seen in ambient samples, these results are not quantitatively presented in the context of absolute ISSW uncertainty. However, as the PSL scattering artifacts caused an ``absorption'' signal in the ISSW that was roughly equivalent to a factor 2-5 overestimate in BC loading for a range of PSL:BC mass mixing ratio of 10-450, this indicates that the ISSW does have a~positive artifact associated with non-absorbing particulate matter. The size of the excursions suggest that purely scattering particles could significantly bias ISSW absorption measurements.

In general it would he useful to mention specific methods applied in previous studies, if they are known, so that findings from the current study can be easily applied by readers to help interpret previous quantifications of BC in snow. Some specific examples where previous studies could be cited are listed below.

Unfortunately, it is not possible to quantitatively assess the findings from previous studies in light of our results. However, we have tried, in the case of the SP2, to point to previous studies that may have underestimated their uncertainties.

Finally, this study applies well-characterized laboratory standards of soot and other particles for evaluating the instruments. This is certainly a logical starting point for analyzing instrument accuracy. A nice addition to this study would be to compare side-by-side measurements of different natural snow samples with unknown BC concentrations and physical properties.

> *Again, we agree whole-heartedly with the reviewer. At present, we don't have snow samples available that have been stored in a manner consistent with accurate particulate loading determinations. We hope to address this comment in a future manuscript.*

Minors issues: 3772,11: ISSW was already defined.
Corrected.

3772,17: "the ratio of light scattering to light extinction" is not "snow albedo".
Corrected.

3773,27: "to quantification"
Corrected.

3775,27: I don't think "PSL" has been previously defined.
Definition added at this point in the paper.

3776,1: How does this laser intensity compare with that applied in previous studies (if known)? Were previous intensities sufficient to measure large particles, if a proper calibration had been implemented?
The laser intensity was slightly better than the best intensity used in the 2010 study referenced. We have added the following sentences to address the reviewer's comment:

> The SP2 was operated with laser intensity $\sim$700\,\unit{nW}/(220-nm PSL), slightly higher than the maximum intensity of that study. This intensity was sufficient to insure proper detection of BC over the range studied here including the large particles. Note that, with sufficient laser intensity, the SP2 calibration to BC mass is independent of laser intensity.

3776,14-16: This wording is slightly confusing.
Reworded for improved clarity.

3776,28: "used in previous SP2 determinations of BC in ice" - Please include refer- ences for the previous studies mentioned.
We have included these citations again at this point of the paper.

3778,7: Please define "particle stopping distance".
We have added an equation defining particle stopping distance.

3779,26: I don't think the Arctic samples are discussed anywhere else, so I see no need to even mention them here or in the previous paragraph.

The Arctic samples are the source of our conclusions about the impact of acidification on size distribution, and also contribute strongly to our confidence that BC size in snow can larger than in air even in the Arctic.

3779,26: "sonewhat"

Corrected

3780,4: Please quantify "slight reduction", so the sentence afterward can be placed in proper context.

The sentence has been changed to:

> There was reduction in the mass fraction of super-micron BC particles after the acidification step, likely due to the acid helping break apart coagulated BC fragments. This did not come close to reducing the BC size to that in the atmosphere, but did essentially eliminate BC larger than ~1500 nm.

3780,10: "showin"

Corrected

3781,23: Did previous BC studies store melted snow in polyethylene bottles before conducting measurements?

We understand that this was not an uncommon approach due to logistical issues, however, as we don't wish to point to specific studies in which perhaps entirely different timescales for sampling/refreezing/testing etc. where used, we hesitate to include citations.

3783,1: "The total uncertainty associated with the ISSW BC concentration determination for ambient snow is estimated as 40%..." - This uncertainty in 'concentration' assumes that the sample BC MAE is identical to the calibration MAE. As discussed later in the manuscript, differences in MAE may introduce another 40% or more under- tainty in derivation of BC concentration.

No, the 40% uncertainty value here includes the previous ISSW estimates of the contribution of MAE uncertainty. We have rephrased the text to make this clearer. The additional 40% uncertainty referred to between fullerene soot and the ISSW calibration material may not all come from absorption, but also potentially the scattering action of the materials. We make this clearer in the text, and add a sentence stating that this uncertainty is beyond those previously understood by the ISSW team.

3783,1-4: Were these uncertainty components published previously? If not, they should be discussed a bit more here, including the basis for each uncertainty component.

We have added the citations at this point in the manuscript.

3783,7: Are the optical properties of the test dust known? If so, please include them. In particular, it would be useful to know how absorptive the dust is. If the properties are unknown, perhaps the dust could be described qualitatively in terms of its absorptance, color, and mineralogy. This seems especially relevant for the ISSW interpretation of BC+dust samples.

We include a qualitative description of the test dust color, and state the main components of the test dust.

3783, 23: "range observed in ... China" - please provide a reference for these observations.

> *This is a statement of Sarah Doherty, the 2nd author. She was involved with a manuscript describing the Chinese work, but the manuscript did not include this piece of information.*

3786,3: "high dust:BC" -> "high dust:BC ratios"

> *Corrected*

3786,13: "significantly bias affect"

> *Corrected*

3787,13: Do you mean "accuracy" instead of "efficiency"?

No, efficiency is correct: if the nebulizer efficiency is too low, we won't get enough BC into the aerosol phase for measurement with the SP2.

Table 1: I suggest adding "ratio" to describe the SP2:grav and ISSW:grav headings. "Absorp. A" could instead be "AAE" for consistency with the text. If need be, the headings could be described in more detail in table footnotes.

> *We have made these changes in the file, and will work with proof-editing to improve clarity of the table.*

Figure 1: Although this depicts a normalized (unitless) efficiency, it would be helpful to list the units of the absolute efficiency in the caption, so the quantity can be understood more clearly.

> *We have added the approximate absolute efficiency to the caption with appropriate units, and added an explanation of the unit at the point of reference in the text.*

Figure 3: These figures should also include 1-to-1 lines.

> *Added.*