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Interactive comment on “Refractory black carbon mass concentrations in snow and ice: method evaluation and inter-comparison with elemental carbon measurement” by S. Lim et al.

Anonymous Referee #1

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Refractory black carbon mass concentrations in snow and ice: method evaluation and intercomparison with elemental carbon measurement

S. Lim et al., 2014 AMTD

The paper is a useful contribution exploring various sources of uncertainty in measurements of rBC or EC in snow and ice. I was pleased to see careful analysis of many different possible sources of systematic error in sample storage and preparation, and think the paper will be a useful contribution to the literature. The comparison of SP2 to OC/EC analysis is also a needed effort. However, the manuscript has some weak-

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nesses. First, there is no real conclusion about the appropriateness of either the SP2 or OC/EC analysis to measuring BC in snow and ice, so the value of the comparison lies more in identifying possible sources of uncertainty than evaluating the overall value of the techniques. I recommend that the authors attempt to consolidate their evaluations of the two techniques in a form that users will find easy to interpret and apply to uncertainty analysis in their own work. Second, the evaluation of the SP2 technique does not adequately explore the question of the size dependence of the nebulization efficiency. As other works deal with this issue, I suggest the authors explicitly recognize the narrower range of their results that do not speak to this issue, and focus on the variability in their results using the SP2 on laboratory and real samples with a system that has not been fully calibrated.

Specific comments:

Abstract: The abstract needs a “bottom line” indicating the overall assessment of the relative value of the SP2 and OC/EC approaches. If the conclusion is merely that neither can be trusted at the level of a factor X, this should be clearly stated. The EC:rBC ratio in different regions is not useful without a synthesis of its meaning. I was surprised that these results did not lead to a presentation/estimation of total uncertainty for each technique in “typical” conditions.

Introduction: 1) The usage suggested here is not consistent with Pezold et al: line 27 of page 3551: “BC” should not be used to refer to optical measures of equivalent BC. This is corrected in line 6 of the follow page. Please make consistent. 2) Line 12 of pg 3552: “refractory BC” was adopted because the method only senses thermal radiation from refractory materials. The technique happens to have the features mentioned. Please correct. 3) A citation should be added to the discussion on pages 3553 of consistency of different measures of rBC/EC/equivalent BC: Kondo, Y. et al., Consistency and traceability of black carbon measurements made by laser-induced incandescence, thermal-optical transmittance, and filter-based photo-absorption techniques, *Aerosol Science and Technology*, 45, 295–312, 2011.

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Experimental section:

1: Page 3555, line 13,14: the PMTs have very little sensitivity at the longer wavelengths. I suggest the authors simply remove the wavelength ranges (which are inaccurate). 2: P3556, Line 8: 0.2 fg corresponds to ~60 nm 3: P3556, line 20: 0.1% by number does not put a strong limit on the fraction of mass not correctly detected. If the average rBC mass in a particle was ~2 fg (typical), then just 0.1% of particles of 1000 fg could significantly (50%) shift the observed mass. On page 3560, the CMD is only 67 nm, so even just 0.1% of particles at 620nm still represents a substantial shift in observed mass.

4: p 3558: more information is needed about the aquadag gravimetric standard. How was this made? Aquadag is a colloid incorporating substantial non-rBC material. How do the authors know the rBC mass in a given mass of Aquadag? 5: p3558: Size dependent nebulization is neglected here. This has been shown to be a significant issue in the U5000. A different AMTD paper also discussing the APEX-Q and U5000 nebulizers, Wendl et al., Atmos. Meas. Tech. Discuss., 7, 3075-3111, 2014 www.atmos-meas-tech-discuss.net/7/3075/2014/ doi:10.5194/amtd-7-3075-2014 has more detailed evaluations of these issues. That paper can be cited here. 6: p 3559, line 25 – 30. This manuscript does not show any data supporting the statement that the size distributions observed with the sp2 represent the size distributions in the liquid. To explore this, one would either need to follow the procedure of Schwarz et al 2012 (cited) over a wide range PSL size (as in Wendl et al, 2014), or start with a known in-liquid size distribution. Therefore the final sentence of the page should be removed. 7:P3560, line 3-5: The authors performed calibrations every two weeks to get information about stability, but it is not clear what frequency they recommend. 8:p 3561, line 13: “be negligible” 9: p3564, line 10: How long were the samples stored after melting and before sampling? 10:Section 3.1.4: these results do not appear to be statistically significant; i.e. there is not enough statistical confidence to state that the surface-to-volume ratio is driving the loss rates of rBC. 11: Section 3.1.5: It seems that there is more variability towards

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over estimating rBC concentration than one would expect; i.e. when repeating measurements, one would expect only statistical uncertainty would contribute to the upper limit on the amount of rBC in a sample, yet here it appears larger. Perhaps the authors can explicitly represent the “measurement-to-measurement” uncertainty in the method.

12: Section 3.2.1 – Here, too, the point about the number frequency of saturating rBC particles is only a weak constraint on the impact on mass concentrations of those large particles. It appears that there is also an error here: if 0.1% of the particles were saturated, and saturation occurs at a mass of 220 fg or higher, and the average mass (i.e. count median mass) is closer to 0.3 fg (for ~70 nm MED), then it is not possible that the saturating particles only contribute less than 0.05% to the total mass. The statement that large particles are unimportant is also not consistent with the sentences attributing the high EC:rBC ratio in HIM to its larger sizes.

13: Schwarz et al., - Black carbon aerosol size in snow, Sci. Rep. 2013, UR - <http://dx.doi.org/10.1038/srep01356> M3 - 10.1038/srep01356 showed that large BC in snow could shift BC’s climate impact. How does the HIM snow size distribution compare to that which they found significant?

14: p 3573: lines 3-5: was BC adsorption to larger dust particles supported in micrographs?

15: Why don’t the authors recommend best practices for the OC/EC measurement?

16: It appears from Table 2 that <1ml volume was sampled for all the samples for the OC/EC measurements, but all the testing of OC/EC was based on much larger samples. How does this affect the uncertainties in the OC/EC measurement. For example, could instrument artifacts become much more significant in analysis of small samples?

Conclusions: The authors should make a stronger effort to synthesize their findings so that users of either/both SP2 and OC/EC can readily estimate their uncertainties. The EC:rBC ratios should not be presented without accompanying explanations/assessment.

Interactive comment on *Atmos. Meas. Tech. Discuss.*, 7, 3549, 2014.

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