We would like to thank the two anonymous referees for their careful reading of the revised manuscript, and also the time they dedicated to evaluating this paper. All comments were highly insightful. Please find below our point-by-point response to the critiques and a highlight to the changes made to the manuscript to address these. For ease of discussion, we have continuously numbered the reviewer's comments. Our responses are shown using blue color, and we strongly feel that we were able to address all the points raised.

RESPONSES TO COMMENTS OF ANONYMOUS REFEREE #1

1. 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.

   We have strengthened our conclusion section by adding a table which summarizes advantages and disadvantages of both techniques. We are not able to state that one of the techniques investigated is obviously more reliable, as such conclusions should be based on multiple studies published by independent research groups. However, we point out numerous potential artifacts (e.g., the dependence of filtration efficiency on quartz filters with rBC particle sizes), and we believe that these results can help users to diagnostic potential issues in their own analytical work.

2. 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.

   To address this comment, we have included new data to our revised manuscript, i.e. PSL calibration of the APEX-Q aerosolization efficiency. See response to comment (11) for more details.

3. 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.

We have deeply modified the abstract to better report above the relative value of both SP2 and OC/EC methods. We agree with the reviewer that the EC/rBC ratios are not a strong output of our study, and not a result which can be directly interpreted in the abstract. EC/rBC ratios are influenced by numerous parameters and artifacts, and we have consequently removed from the abstract all discussion related EC/rBC ratios.

4. 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.
   We agree with the reviewer. The second part of the sentence was removed and the sentence was revised as follow: "It is now generally agreed that, when determined by thermal methods using its chemical properties, BC should be called "elemental carbon (EC)".

5. 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.
   The sentence has been corrected as follow : "This technique is nearly independent of the morphology of BC and of the presence of other materials such as light-absorbing organics (Bisiaux et al., 2012a, 2012b; Kaspari et al., 2011; Jenkins et al., 2013; McConnell et al., 2007; Sterle et al., 2012).

6. 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.
   The reference (Kondo et al. 2011) was added with relevant discussion in the page 3553. In page 3553 line 21, the following sentence is added. "An inter-comparison study between a SP2 and a filter-based absorption photometer continuous soot monitoring system (COSMOS, using a heated inlet) revealed the good agreement within 10 % on average from measured BC concentrations in Tokyo (Kondo et al., 2011)."

7. 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).
   The sentence was simplified as follow: "The mass of individual rBC particle is proportional to its incandescence signal, as detected by two PMT photodetectors with broad band and narrow band detection capabilities."

8. P3556, Line 8: 0.2 fg corresponds to ~60 nm

   The manuscript was corrected.

9. 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.

   In this study, the SP2 settings allow for detection of rBC sizes ranging from 60 to 620 nm, which correspond to MED ranging from 0.2 to 220 fg. This range is too limited, and does not allow to capture the entire mass distribution of rBC in snow or ice samples. Recent studies (Schwarz et al., 2012, Wendl et al., 2014) have reported extended size detection ranges. The revised manuscript highlights the limitation of the [60-620] nm detection range, and we now recommend to follow the approach of Schwarz et al. (2012) and Wendl et al. (2014), i.e. to modify the SP2 setting to enlarge the size detection range of the analyzer.

   Thus, as pointed out by the reviewer, 0.1% by number observed at saturation does not allow for an accurate estimate of on the fraction of mass not correctly detected with our SP2. However, we believe that our results are still relevant. We can apply a lognormal fit to extend the mass distributions observed in this study for sizes larger than 620 nm (such fits are reported on Fig 2). When using these fit curves to evaluate the mass contribution at sizes above 620 nm for the SUM, CDD, and ELB samples, we find that our concentrations are underestimated of 4-12 %.

   We understand that this calculation would not apply if large particles (e.g., ~1µm diameter) exist in the sample: then the masses as reported in this manuscript would be largely underestimated. Such large particles have been reported by Schwarz et al. (2012) for seasonal snow samples collected near Boulder and in the Arctic. The former samples are thus likely impacted by local burning sources, and the latter samples were melted and refrozen before analysis. We assume that both the vicinity of BC sources and the coagulation of small rBC particles driven by thaw/freezing cycles within low-altitudes snowpacks can explain the occurrence of larges rBC particles. Consequently, the SUM, ELB, and CDD samples should not contain large particles, as they have been collected at high altitude sites where there is no seasonal melting. However, the HIM samples may have been affected by coagulation of rBC particles during the long liquid-phase storage prior to analysis.

   The section 3.2.1 entitled “Measurement sensitivity to SP2 mass detection range” has been fully rewritten. The revised manuscript now highlights the limitation of our size detection range.

However, the mass determined for the CDD, SUM, and ELB samples may be only underestimated by less than 12%. The HIM samples rBC concentrations are more uncertain.

10. 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?

    This is an excellent point: we agree that the rBC content of our Aquadag batch should be determined, and later considered when estimating aerosolization efficiencies of nebulizers. Consequently, we have included in the manuscript a new section 2.2.2 entitled “BC standard solutions”. This new section includes details about how Aquadag standard solutions were prepared. Notably, we now report the solid content of our Aquadag batch, 28 %. We also describe the determination of Aquadag rBC mass using the thermal-optical method. Interestingly, thermal-optical analyses indicate that EC makes up ~ 87 % of the total Aquadag mass. Previous studies using similar method reported ~71 and 76 % rBC content for Aquadag (Gysel et al., 2011; Wendl et al., 2014), but we expect this content to vary with Aquadag batches.

    We have revised through the manuscript our estimates of aerosolization efficiencies based or Aquadag standard solution nebulization to take into account this updated rBC content of our Aquadag batch. E.g., the APEX-Q aerosolization efficiency has been revised from 65±4 % to 75±7 % and final rBC concentrations of field samples have been revised in Table 1 and Table 2.

11. 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-meastech-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.

    We agree with the reviewer’s point since aerosolization efficiency is size-dependent and should be evaluated for any individual nebulizer. To address this concern, we have conducted new evaluations of the size dependence of the aerosolization for the APEX-Q nebulizer. These data are now included in a new section, 2.2.3, entitled “Size-dependence of the aerosolization efficiency”.

    Briefly, solutions containing PSL of specific sizes (150, 200, 240, 350 and 600 nm, covering our SP2 detection range) were nebulized with the APEX-Q. We found aerosolization efficiencies ranging from 67 to 80 %, with no significant trend with size. Overall, we established a mean efficiency of 72±5 %. These results are also reported with a new figure in SI.

We did not extend our PSL analyses above our SP2 detection range (i.e., > 620 nm). However, our results in the 150-600 nm range are really similar to the one recently reported by Wendl et al. (2014). Both our data and the Wendl data show no decrease in aerosolization efficiency for larger particles, as reported previously for the U5000AT nebulizer (Schwarz et al., 2012). Wendl et al. (2014) PSL analyses covered a 100-1000 nm range, and these authors did not find any decrease in aerosolization efficiency above 620 nm for the APEX-Q. We now clearly refer to this study in our manuscript.

Finally, our revised manuscript highlights that, interestingly, the aerosolization efficiencies we determined for the APEX-Q from both PSL (Sect. 2.2.3) and Aquadag (Sect 2.2.4) analyses are very comparable (75±7 % and 72±5 %, respectively).

12. 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.

    As described in the response to comment (11), we evaluated the size-dependency of particle aerosolization for PSL of known diameters. A new section (2.2.3) has been included to the revised manuscript.

    We also now discuss in more details in section 2.2.5 that coagulation of rBC particles is unlikely during nebulization with the APEX-Q. This results is based on both PSLs analysis, investigation of size distributions for AQ solutions of concentrations ranging 0.1 – 100 µg L^{-1}.

    Note that the sentence pointed out by the reviewer has been removed. This discussion related to coagulation is no longer included in the section discussing aerosolization.

13. 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.

    The manuscript has been clarified (section 2.2.4, last paragraph). We recommend a full, 8 standards, calibration every two weeks, and a more limited (3 standards) calibration daily. The daily calibration allows to control that aerosolization efficiency remains stable and to that no unlikely dysfunction may be affecting the nebulizer.

14. p 3561, line 13: “be negligible”

    The manuscript was corrected.

15. p3564, line 10: How long were the samples stored after melting and before sampling?

The reviewer discusses here the Himalayan (HIM) samples. These samples were collected in the field in 2010-2012 and analyzed using the SP2 and the sunset OC-EC analyzer in February 2013. Consequently, they were stored in the refrigerator for 1-2 years. Note that the HIM samples melted during their transport from the collection field to our laboratory in France which lasted for more than a week. This is now clearly stated in Sect 2.4.4 of the revised manuscript.

16. 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.
    We agree with the reviewer that the limited numbers of samples used to characterize the impact of surface-to-volume (S/V) ratio of containers on rBC analysis is too limited to bring solid conclusions. We have not removed this section of the manuscript as we expect this parameter to S/V ratio to influence rBC in frozen liquid samples; however, we have rewritten the last sentence of Sect. 3.1.4 so as to be much more careful with our interpretations.

17. Section 3.1.5: It seems that there is more variability towards 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.
    In section 3.1.5, we specifically investigate how storage and melting procedures may impact rBC concentrations of snow samples. We report quite large variability in rBC levels, e.g. when repeating analysis over 24-h. However, we attribute most of this variability to storage and melting procedures, as stated in section 3.1.5.
    New results have been included to the manuscript in section 2.2.5 to provide a clear estimate of the "measurement-to-measurement" uncertainty of our technique. Analyses conducted on alpine snow were repeated every 2-minutes during a 1-hour long period (n=30) just after melting. We assumed that storage-driven rBC losses should be negligible over such 1-hour long period. We observed a mean rBC concentration for this sample of 6.3±0.25 µg/l (1hour, n=30), resulting in a relative standard deviation of 3.9%. We believe that such test provides an adequate description of the "measurement-to-measurement" uncertainty of our method.

18. 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.

The point raised by the reviewer is perfectly true, see response to comment (9) for more details. The contribution of saturating particles to total samples mass ranges 4-12%, and not 0.05%. This estimate applies to CDD, ELB, and SUM samples, but could not be determined for the HIM samples. Section 3.2.1 of the revised manuscript has been completely rewritten.

19. 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?

    Schwarz et al. (2013) discuss that the mass absorption cross section (MAC) of BC particles smaller than 500 nm is much larger than for larger BC particles; consequently large particles have smaller MAC and thus lower albedo relevancy. The reviewer points out that size distributions reported in our study for HIM samples suggest the occurrence of larger rBC particles in the Himalayan snow cover may, and consequently might suggest a lower-than-expected climate impact of BC in this area.

    Although it is a nice argumentation, we prefer to be careful and not discuss it in the manuscript. The HIM samples experienced a week long transport from the field to the lab refrigerator, due to the really remote sampling locations (i.e., Climate Observatory-Pyramid and Changri Nup glacier, Nepal). We hope future studies will be able to tackle this question.

20. p3573: lines 3-5: was BC adsorption to larger dust particles supported in micrographs?

    Unfortunately, electron microscopy analysis was not directly perform on the quartz filters used for the thermal-optical analysis, but rather on 0.2 um-nuclepore filter were samples were filtered a second time. Consequently, it allowed to observe BC aggregates with diameters ranging between 100 and 200 nm particles not initially collected on quartz filters. Furthermore, electron microscopy was only applied to CDD samples characterized by low dust and organic carbon content compared to other samples (eg., ELB and HIM).

    Thus we have no direct observation of BC adsorption to larger dust particles. However, this hypothesis seems reasonable for the HIM samples considering there elevated dust concentrations (mean concentration of 1,318 ppb, yellowish or brownish filters). Furthermore, particles agglomeration in Himalayan snow samples has been observed previously using microscopy analysis (Wang et al., 2012).

21. Why don't the authors recommend best practices for the OC/EC measurement?

We demonstrated that thermal-optical measurements of EC in snow samples are challenging, and three important recommendations can be extracted from our study: (i) specific evaluation of the filtration efficiency is required for different samples, (ii) monitoring and correction of thermograms is needed for highly-dusty samples, and (iii) modification of temperature program can limit the effect of dust or pyrolized OC fraction on the OC/EC split point.

The conclusion has been reformulated to present these clear recommendations.

22. 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?

    This was a typo. The unit of filtrated sample volume for EC should be “l”, and has been corrected.

23. 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.

    The conclusion of the manuscript has been fully rewritten. It now includes a detailed interpretation of the EC/rBC ratios, as well a better statement of the limitations of each methods. The revised conclusion also includes a new table (Table3) summarizing advantages and disadvantages of both methods.

RESPONSES TO COMMENTS OF ANONYMOUS REFEREE #2

24. One major concern I have is that analysis of known lab standards of BC in water (Aquadag) were not conducted using both methods. The Aquadag standards used to evaluate aerosolization efficiency should also be measured with the sunset labs EC/OC instrument following the same protocol as used for samples. A plot showing this comparison would be good too. This would provide a nice baseline comparison between the two methods for ideal standards not subject to artifacts caused by OC and dust. The primary artifacts affecting analysis of the standards are size distribution (already characterized by the authors) and filtration efficiency, which the authors evaluated for the field samples. Analyzing lab standards in the same way would add insight into how much of a role the other artifacts play in over or under-estimating EC or rBC.
    This is an excellent suggestion, but unfortunately we were not able to address it due to analytical issues with our Sunset EC/OC analyzer. We understand that this is a weakness of our manuscript. Thus we modified our conclusion section to strongly advice that future studies conduct such Aquadag-based comparison between the two techniques.

25. Page 3551, line 3. The statement that BC is the second most important component of global warming is a bit strong and the uncertainties surrounding BC's role in climate change are pretty high. I would replace "is" with "may be".
    The sentence was modified as suggested by the reviewer.

26. Page 3552, line 22. Run on sentence beginning with "Appropriate Treatment…"
    This sentence has been rephrased.

27. Page 3553, line 6. Run on sentence beginning with "These differences appear despite…" Suggested re-write "These differences appear despite good precision of individual laboratory analyses and are mainly attributed to organics that pyrolize during thermal analysis."
    The sentence was modified as suggested by the reviewer.

28. Page 3555 line 11, "incandesces while emitting visible radiation". The definition of incandesce is to emit light. Did the authors intend to say the incandescence was in the visible spectrum? Is there a specific wavelength or range of the spectrum that should be specified here?
    We agree with the reviewer that "incandescence" relates to light emission, but is not wavelength specific. To clarify our manuscript, we have removed all information about wavelength ranges detected by SP2 PMT photodetectors. The sentence now reads: "The rBC particle absorbs light, reaches its vaporization temperature (~3700 to 4300 K) and incandesces."

29. Page 3555 line 16, where did the rBC density of 1.8 g cm-3 come from. Reference please.
    The relevant reference (Moteki and Kondo, 2010) was added to the manuscript.

30. Page 3555 line 17, Please provide a published reference for the SP2 toolkit, or indicate if you will be describing this toolkit further in the paper or in SI.
    To our knowledge, there is no published reference for the SP2 toolkit. However, this SP2 toolkit has been used to retrieve SP2 data in previous studies (e.g., Gysel et al., 2011; Laborde et al., 2012; Wendl et al., 2014). We added to the manuscript the website link where the SP2 toolkit can be downloaded (i.e., http://aerosolsoftware.web.psi.ch/)

31. Page 3556 line 7. Starting this paragraph with the same phrase as the previous paragraph is confusing. Are both paragraphs referring to the same calibration method? If the second calibration is specific to rBC, what was the first calibration for?
    The first paragraph of section 2.1.2 starting page 3556 line 20 is a general description of the SP2 internal calibration, which involve the other instruments such as atomizer, silica gel drier and differential mobility analyzer (DMA), and condensation particle counter (CPC). The second paragraph of section 2.1.1 starting in page 3556 line 7 reports specific information on calibration size ranges of BC standard including the lower and upper limits of BC detection. To clarify the manuscript, the first sentence of the second paragraph was rephrased as follow: "Fullerene soot with a rBC mass ranging from ~0.2 to 70 fg, corresponding to ~60 to 420 nm MED, was used for calibration of the SP2."

32. Was any comparison between SP2 and Sunset Lab EC/OC analysis done using the Aquadag standards? It would be interesting to see how comparisons between the two methods using an idealized standard as well as the field samples.
    See response to comment (24)

33. Page 3561 line 10 and 11, in discussing why HULIS would have a negligible impact on particle aerosolization, use of consistent units would be helpful; use either mg L-1 or ppbC.
    The sentence was modified to have consistent units as pointed out, and only the unit, mg L^{-1} is now reported.

34. Page 3561 line 13 space between be and negligible.
    The manuscript was modified.

35. Page 3562, line 1 "Since concentrations of EC and OC are low in snow and ice." Please give references for statement and a range of what the authors consider to be "low". Why is

decarbonatation necessary when EC/OC concentrations are low? Although the detailed description is in the SI, a brief statement explaining why this needs to be done would be helpful. When referencing SI, provide section number or page number.

We have completely rewritten and clarify the paragraph of section 2.3 describing the decarbonation treatment applies to filters before thermal-optical analysis. This artefact depends on the calcium carbonate load on filter, and not on EC or OC concentrations. Actually, the lowest is the EC levels, the more important will be the relative contamination from calcium carbonate. Most of the samples analyzed in this study (i.e., CDD, ELB, and HIM) originate from area potentially impacted by Saharan or Asian dust. This dust could be an important source of calcium carbonate, and thus the decarbonation procedure was applied to all samples.

36. Page 3572, line 10-13, more recent studies have shown that filtration efficiency through quartz fiber filters can be 30% or less (Torres et al., 2013 Aerosol Sci & Tech; Hadley et al., 2008 Env. Sci& Tech) .

    We have added both references in the discussion.

37. Page 3573, line 25 "The rBC concentrations ranged between ~35 and 70 ug L-1, and remained constant at around 5 %." This is a confusing sentence that at first seems contradictory. This can be clarified by rewriting this sentence as well as the previous one as "Different samples with rBC concentrations ranging from 35 to 70 ug L-1 were each measured at regular intervals over 15 minute. The reported concentrations remained constant (standard deviation <5%) over each measurement period."

    The reviewer is correct. The sentence was not clear, and was we thus rephrased it as suggested.

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