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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Black carbon concentrations and mixing state in the Finnish Arctic" *by* T. Raatikainen et al.

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We would like to thank Referee #3 for the comments. Below are the issues raised by the referee (quotes shown in italics) and our replies (plain text).

General comments:

1. The reason at the root of the discrepancy between the aethalometer and MAAP with respect to the SP2 is not clear. The explanations in the paper are not too convincing to me; therefore, it is difficult to assess the goodness of the estimation of the LAC volume fraction used at the end of the paper. Most aethalometers have multiple wavelengths, some of the proposed ideas for why the SP2 and the aethalometer do not agree could be tested using the information provided at the different wavelengths, for

example through the Angstrom exponent. Could it be that the mass density chosen for rBC plays also a role into these discrepancies? Finally, it might help to discuss some papers that already reported issues with filter-based instrumentation such as those by Cappa et al., Lack et al. and Subramanian et al. (1. Bias in Filter-Based Aerosol Light Absorption Measurements Due to Organic Aerosol Loading: Evidence from Laboratory Measurements, Christopher D. Cappa , Daniel A. Lack , James B. Burkholder , A. R. Ravishankara, Aerosol Science and Technology Vol. 42, Iss. 12, 2008; 2. Bias in Filter-Based Aerosol Light Absorption Measurements Due to Organic Aerosol Loading Evidence from Ambient Measurements, Daniel A. Lack , Christopher D. Cappa , David S. Covert , Tahllee Baynard , Paola Massoli , Berko Sierau , Timothy S. Bates , Patricia K. Quinn , Edward R. Lovejoy , A. R. Ravishankara, Aerosol Science and Technology, Vol. 42, Iss. 12, 2008; Yellow Beads and Missing Particles: Trouble Ahead for Filter-Based Absorption Measurements, R. Subramanian , Christoph A. Roden , Poonam Boparai , Tami C. Bond, Aerosol Science and Technology, Vol. 41, Iss. 6, 2007)

We do not have complete explanation for the difference between Aethalometer/MAAP and SP2. We have modified this section so that instrument bias (due to inaccurate instrument parameters) is one potential explanation and the presence of light absorbing material that cannot be detected by the SP2 is the other explanation. It is clear that the instrument and other parameters (including mass density of rBC) have some uncertainties, but we are using the best available information, which means that the results are as accurate as they can be. We have calculated the Ångström exponent from the Aethalometer data and found nothing exceptional; the exponent is close to 1.2, which indicates relatively weak wavelength dependency. This is now mentioned in the paper.

2. An abundant body of literature on single particle microscopy and single particle numerical calculations of the optical properties of rBC mixed with other material and the effects this mixing might have on radiative forcing has been ignored. Including a discussion of some of this literature might improve the discussion of the results found by the authors and the relevance of the paper.

In the introduction we have added a reference to the Bond et al. (J. Geophys. Res., 118, 5380-5552, 2013) review, which summarizes several single particle microscopy studies and studies examining the effect of mixing state on aerosol radiative properties. We have also clarified our aim, which is to compare different mixing representations based on the new information about the mixing state and concentrations from the SP2 and MAAP measurements. To our knowledge, there are no similar studies that could be directly compared with our study.

Specific comments:

Abstract:

1. "SP2 is a unique instrument that can give..." several instrument might be considered "unique". I would suggest to just write "SP2 provides..."

Done.

2. I suggest removing "As expected"

Done.

3. "...the number fraction of particles containing rBC..." this value is calculated only in the range of sizes detected by the SP2? Maybe it should be noted here.

The size range is now given.

4. "Comparison of the measured rBC mass concentration with that of the optically detected equivalent black carbon (eBC)..." add "...using an aethalometer and a MAAP"

Done.

5. I think that the sentence "(separate non-absorbing and coated rBC particles)" is not very clear here until one reads the rest of the paper, so my suggestion is to just remove this. If the authors prefer to keep it, they might want to explain it a bit more.

We have now removed the sentence.

Introduction:

- Line 39: "For example, it can be distributed" what "it" refers to might be confusing, I suggest to specify.

We have now clarified that it refers to absorbing material.

- Lines 41-42: "By definition, an aerosol population is externally mixed when not all particles are absorbing and internally mixed when all particles are absorbing." This could be confusing as an externally mixed population of particles could be just made of non-absorbing aerosols at all, just with different composition. I think it should be clarified that this refers to absorbing aerosols only.

We have now clarified that this is an optical definition.

- Lines 56-57: "This means that non-refractory absorbing material such as brown carbon cannot be detected by the SP2." I would think another main reason the SP2 would not detect directly brown carbon is that it operates with a NIR laser at a wavelength where brown carbon would not absorb.

This wavelength limitation is now mentioned.

Experimental:

- Lines 97-98: I think that at this point in the paper, this sentence is not clear, what is meant becomes clearer later on, but it might be good to either remove or clarify this sentence here. In general I found all this paragraph until line 106 confusing and unclear. I would suggest clarify it a little bit more maybe by adding some details.

We have now clarified this paragraph.

- Line 107: Specify the model of the aethalometer. Is this the 7-wavelength model? In general provide model and maker also for the other instruments such as MAAP, aethalometer, DMPS and gases.

Done.

- Lines 122-123: The particles actually pass through the laser cavity.

Fixed.

- Line 123: "...absorbing particles are identified..." not all absorbing particles, but mostly rBC. For example, brown carbon is probably not detected.

This part of the text is updated.

Results:

- Line 194: "Ambient temperature..." is this the daily average, the min the max, something else?

It is hourly average, which is now mentioned in the text.

- Line 226: "number fraction of particles containing rBC is 0.24..." as for the abstract, please define clearly the size range used for this calculation.

We have now added size ranged to the previous paragraph where the parameter time series (shown in Fig. 3) are described.

- Line 309: Please explain how the Δ are calculated, in other words, how is the reference value estimated? In addition, it might be interesting to provide also the slopes for the other gases.

We have now explained the fit and the calculation of the reference value. Since our focus is on rBC mixing state, we have shown only carbon monoxide (CO) which is the most commonly used tracer gas. Giving the other slopes would require proper description, interpretation and comparison with literature values; this would make this section too long.

- Lines 329-331: it might be interesting to look at the $\Delta rBC/\Delta CO$ versus distance from the source as given by the hysplit.

Note that ΔrBC/ΔCO is a single value calculated from the campaign data. However, we have examined if the deviation of the individual data points from the linear fit depends on the HYSPLIT parameters, but clear dependencies were not found.

- Line 352: That's true but MAAP I believe measures at 670 nm where brown carbon should absorb very little. Also the sentence "Secondly, MAAP detects practically all absorbing particles" can be deceiving as one might interpret it as if the MAAP could detect also all BrC and all dust, which would not be true if not absorbing at 670 nm. Maybe the authors mean "all rBC" instead of "all absorbing particles"?

Brown carbon is not mentioned anymore. The second sentence was referring to the detection range (limited size range for SP2, but not for MAAP), which is now clarified.

- Lines 371-376: This paragraph is a little bit hard to read and understand. Either I do not understand clearly the different models, or the internally mixed homogeneous model seems very unrealistic. A cartoon of the different mixing scenario might help much more than the written explanation.

A cartoon has been added.

- Line 382: the placement of the right ")" seems incorrect.

This was intentional, but we have now removed the parenthesis.

- Line 406: the surface albedo used is fairly low here. Please justify the choice; for example, considering that at the site the albedo might be much higher if snow is present.

The used surface albedo is typical for forests seasonally coved by snow. It is a fact that the absolute values of the surface albedo and most other parameters in this equation are not well known. Since our purpose is not to give exact RFE values, but to compare different mixing state models, slight inaccuracies can be tolerated. The equation itself is also a great simplification of a difficult radiative transfer problem, but good for our purpose. We have now clarified this.

Caption of figure 5: I would change "correlation" to "slope" or "line" in "The dashed line shows 1:1 correlation."

Done.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 15621, 2015.