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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.

Anonymous Referee #3

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The paper discusses SP2 measurements in arctic regions. Overall the paper is well written with only a few unclear sections. The measurements are interesting and worth publication. The novelty of the paper is limited; however, I still believe that these data should be available to the community as there is a strong need to know the concentrations and mixing state of rBC in different locations around the globe. The paper should be published after addressing a few issues.

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)

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.

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. . ."

2. I suggest removing "As expected"

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.

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”

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.

Introduction:

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

- 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.

- 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.

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.

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

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

Results:

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

- 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.

- 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.

- Lines 329-331: it might be interesting to look at the ΔrBC/ ΔCO versus distance from the source as given by the hysplit.

- 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”?

- 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.

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

- 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.

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

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