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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Single particle characterization of black carbon aerosols at a tropospheric alpine site in Switzerland" *by* D. Liu et al.

Anonymous Referee #3

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Single-particle characterization of BC aerosols at a tropospheric alpine site in Switzerland D. Liu et al. ACPD

The paper describes measurements of BC aerosol measured at Jungfraujoch, with very interesting observations of cloud removal events and BC mixing state evolution. It includes detailed discussion of the single particle soot photometer (SP2) measurements, upon which most of the conclusions are based. I applaud the authors' willingness to include this discussion, but feel that they are secondary to the main contributions of the paper. Thus I hope the authors will concentrate on addressing my main point below:

The paper will benefit from careful editing throughout for readability; although much information is presented, the reader is not provided with clear guidance as to the value of the information or the implications of the results. Many of the graphs are very information dense, as are the tables. An example of this, I point to the detailed catagorization of different airmass types, yet in reading the manuscript I only was made aware of the importance of a small subset of these. I hope that editing will remove any sensation that this is a merely a collection of observations, and provide a better flow of information to support the conclusions (which should also be made clear). To my mind the conclusions about BC removal should certainly be amongst the main conclusions of the paper. This is my most significant comment overall.

I had many technical questions/points, largely centered on the experimental discussion, which I include below.

Figures are not cited in order.

Abstract: - Lines 50-54: is the MAC value(and those it is compared to) appropriate for all the air sampled, or only the FT air? - Line 64: The statement that non-refractory materials are thickly coated "around the BC core" is likely overly specific. The incandescence method used cannot determine physical arrangement. - Line 66: The term "enhancing BC mixing state" is not clear, but I believe the point of this sentence is not explicitly stated: as it is self-evident that condensation will lead to decreasing BC mass fraction in particles, I read this as an indication that, in the airmasses sampled, condensation was a more significant route than coagulation or cloud processing in accomplishing this decrease. A similar sentence should be revised in the conclusion/summary

Introduction:

- Line 90: BC aerosol is removed through wet and dry deposition: i.e., it is not an "ideal tracer". Please correct the discussion - - The introduction, which may be too far ranging, includes brief description of many questions "related to the size distribution

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and mixing state of BC". What is the subset of these are addressed in this paper? (Line 129). Providing an answer in the text is a step to addressing my larger comments above. - The introduction did not include much discussion of the importance of the BC size distribution – what is it?

Line 203: Please add "mixed" to the appropriate extended weather type in table 1.

Line 236: is the laser intensity measured here, or is this the value published in Stephens et al? Please clarify in the text.

Discussion of lines 240-252: if any strong absorber (line 240) emits thermal radiation, why is this sufficient to identify particles as BC or not (line 252)?

L252: Calling a particle "BC" when an incandescence signal was observed suggests that the mass of the particle is then the mass of BC, which is not the case. I suggest calling these particles by a more general term (e.g., BC-containing) when specifically referring to an internally mixed particle containing BC . Then when you refer to "BC", it will be clear that you refer only to this component of the aerosol. (The discussion later in the paper of "when a particle has an absorbing core" is what brought this to my attention).

Line 263: what was the minimum calibration mass used? What is the detection range of the SP2? Please clarify in the text.

L273: What is the meaning of "a range of uncertainty"? Does this value represent all the uncertainty with the mass determination, or just the extraction from the incandescent signal?

Line 275: the shape of the BC does not affect its void-free volume

L283: presumably variation in laser intensity results in a proportional change in the scattering signal, which probably scales very strongly with particle diameter. Why does diameter vary so much 6% for such small (10%) changes in laser intensity?

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L284: the Baumgardner 2007 citation is not included in the references. (I assume it is the B. 2007 paper on Mexico City measurements.) This paper does not present a confirmation of lack of refractive index dependence (See figure 2 for a narrow range of RI). Additionally Figure 8 of the Gao 2007 citation shows a strong dependence of scattering signal on index of refraction. Please correct/clarify in the text.

L291: Is the scattering signal of the absorbing material measured at the laser center?

L309: how is uncertainty for the mass fraction only 23% if the uncertainty in “Do” is as high as 55 %? (l296)

Paragraph at L329-345: I am surprised that there are such significant particle losses associated with diffusion at such large sizes: typically diffusion loss is at smaller particle diameters. This raises several questions: First, how good is the SMPS at quantifying particle number? Presumably the SMPS assumes an efficiency for the electrostatic neutralizer, but if it was not as efficient as expected (i.e. an older polonium source) this would quickly impact number determinations. Also, diffusion losses would lead to a shift in the size distributions measured by the SP2: large particle number would not be affected as much as small particle number. This would determine the impact on the BC mass concentrations determined by the SP2 [which I presume is the main reason to include this analysis]. Please comment on the feature of the sample line that suggests the losses are caused by diffusion.

In lines 334-336, this statement is incorrect: the slope of the scatter-plot does not indicate the total number ratio. The discussion of the paragraph is aimed at the efficiency of the SP2 in counting total aerosol in the accumulation mode, but does not deal with the question of its counting of BC particles (and hence the implications for bias in the “bc number fraction” [l341-342]). I suggest adding an estimate of this bias, if possible, and explicitly stating that it is not addressed by the previous discussion. Ah. Now I see some of this discussion in the results section. Perhaps it is more appropriate here?

L367 Please explicitly state that the total number fraction of BC observed is based on

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the assumption of all BC in a single mode.

The log-normal fits in figure 9 look like they were constrained, perhaps the center-position value? If is this is the case, this should be stated, and the rational for choosing a center value explained. In either case, please clarify in text.

L402: I don't understand: in the Slowik study, MAAP measured more absorption (with some intermediate MAC that's reasonable?), so would this not suggest that high MAC is correct?

L412: Delayed with respect to the peak of the scattering signal?

P423: Please state that this analysis assumes a laser with uniform intensity, and thus there is no expectation that the bare BC core can scatter more light than the coated particle.

L454: Which Moteki et al citation is appropriate? In the Schwarz et al reference, bounding assumptions were made about the coatings on undetermined particles – not what is stated here. Please correct.

L464 "more likely to be thickly coated.". . . if they are detected?

L494 How does the pollution levels in an air mass change, if it's history doesn't change?

Figure 7: please consider overlaying smoothed/averaged with slower time resolution BC,BC ME, and CO/BC ratio to make average values of these quantities easier to make out.

For discussion of BC/CO ratio, it seems that CO background has not been removed for the discussion/analysis. As CO has such a large background compared to its enhanced values, this hides the BC/CO relationship shown in Figure 8, and discussed. Perhaps you can plot BC vs CO to show the correlation between the two species. At present, I don't think this is convincingly displayed. Please improve.

L582: please comment on the bimodal distributions associated with SE winds in Figure

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8.

L598-600. Please add the subject of the comparison: x is more important than y for. . .. .

L626: BC is not soluble

L635: .23 = 23%

L717 BC not BE. In this section it seems that there is mixed discussion of ice phase clouds: CCN and IN are different, and the impact of coatings of BC on its IN/CCN activity will be different. This discussion should thus be edited to remove this confusion. L721 – I don't understand how mixed particles homogenously nucleate.

Figure 5: is the histogram on the right normalized for the different log-widths of the vertical axis? Please include the answer in the paper, as it makes it easier to know whether the area under curve scales as total particle number or not (for the different regions).

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 8765, 2010.