

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

Anonymous Referee #2

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Review of the paper "Single particle characterization of black carbon aerosols at a tropospheric alpine site in Switzerland" by D. Liu et al.

This paper presents a characterization of free tropospheric background and ground sources influenced black carbon particles in the Swiss Alps (3600 m asl) using a single particle detection instrument which measures particle size, BC mass concentration, and BC mass fraction (leading to mixing state and coating thickness).

The scientific content of the manuscript is a relevant topic within the scope of ACP and with some minor changes (requested in the general and specific comments below) it is appropriate and recommended for publication.

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In general, the manuscript is well written with respect to language and structure.

The introduction provides an extensive overview about the motivation to determine mass loading and mixing state of BC particles.

General comments:

Although it is a big step forward to use the SP-2 as a single particle measurement device the comparisons with the bulk BC measurements from former CLACE campaigns at the same site should be intensified in the different chapters of the paper and not only compared at the end of the summary. These results can be found in Cozic et al. 2007 (ACP, 7, 1-11), Cozic et al. 2008 (JGR, 113, doi:10.1029/2007JDD009266) and Mertes et al. 2007 (AS&T, 41, 848-864). Beside BC mass loadings, also BC mass fractions are derived in these publications by relating the BC mass to the total aerosol mass, assuming internal mixing. Interestingly, the values of these parameters are very close to the ones obtained in this work.

The sub-division "free tropospheric background" used in the discussion section is somewhat shady. As I understood right are these the "white periods" in Fig.3. But they belong to the same weather types CA, M and CI, which were partly declared as "influenced by SE winds". Since the second main wind direction is N, those periods should be consistently labelled as "influenced by N winds" and should be influenced by pollution sources in northward direction, like the SE wind periods are influenced by pollution sources in south east direction. When these N wind periods are related to cleaner air that should simply mean that the pollution source in this direction is less strong. If this interpretation is a misunderstanding this point needs to be explained more clearly, but at the moment it looks like that there is an inconsistency between the back-trajectory analysis and the weather type classification from Tab.1.

Specific remarks

L. 62: Please indicate the size range limits of the measurements already here in the

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abstract.

L. 179, Fig.1: At least in my copy the maps are artificially stretched, which should be avoided. For a better comparison with the in-situ data, the arrival times of the trajectories at the measurement site should be given.

L. 204, Tab.1: Four of eight weather types are mentioned that did not occur at all during the measurements, so why are they listed? At least one should indicate the weather types that prevailed during the field campaign.

L. 284: According to the reference list it must be Baumgardner et al. (2008).

L. 328-336: It is difficult to follow the content of this paragraph in detail. First of all: Has the SP-2 a size-dependent detection efficiency or is it unity for the diameter range 200nm – 720 nm and zero outside of this size range? A particle loss of 34% is certainly not caused by line losses in this size range. Moreover, it would be nice to have a figure where the size distributions and not only the counts of SP-2 and SMPS are compared. This would show if the losses are size dependent or not.

Fig.2B: scatterplots in log-log presentation do not only “not show” but often hide undesired properties and should be therefore avoided (that is also true for the log plot of Fig.2A). Although 2 orders of magnitude need to be covered a linear plot would be more appropriate to see the quality of the correlation. What is the interception and significance level of the regression and how can they be motivated? However, in the log-log plot the data points are strangely distributed up to a SMPS concentration of 5 cm⁻³ showing these vertical structures, which means a large variation of SP-2 concentration for one SMPS concentration. What is the reason for this?

L. 336-337: I do not understand. Do you use two different diameter definitions for SP-2 and SMPS? If so, which is natural for the two different techniques, please indicate the difference by transferring the SMPS into the SP-2 diameter definition. Nevertheless, should this explain the 66% efficiency of the SP-2 as it is suggested by the formulation

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of this sentence.

L. 338-341: sub-micron/accumulation mode ratio: a. Are the SP-2 counts corrected for the 66% overall efficiency? b. Since the SP-2/SMPS counts ratio is obviously not constant it would be much more convincing to use the SMPS counts in the size range 200 nm – 720 nm for the accumulation mode concentration. What is the reason to use the SP-2 concentration? Moreover, in case the SMPS measures down to at least 20 nm, it would be even more justified to determine the ratio only from the SMPS in order to avoid counting differences between different sensors. c. Because the sub-micron size range includes the accumulation mode size range, it would be more illustrative to use an accumulation mode/sub-micron ratio, which directly gives the percentage of accumulation mode particles in the sub-micron size range. Why was the ratio defined in the reciprocal way?

L. 341-343: Has the SP-2 a (size dependent) detection efficiency for BC particles? If so, is this taken into account for the determination of the BC number fraction? It would be more interesting to determine a BC number fraction for the same size range. So what about to use the SP-2 or SMPS concentration in the same size range as the SP-2 BC particle measurement to determine a BC number fraction?

L. 362-371: It is unclear whether all presented SP-2 data (where relevant) is treated with this log-normal extrapolation to smaller sizes, which significantly increases BC number and mass concentration. Particularly it is not clear whether this extrapolation approach is applied for the determination of BC number fraction defined before (L. 341-343). Since this approach is a kind of data correction it should be shifted forward into the “instrumentation” sub-chapter and should not be presented in the results chapter. In order to further evaluate the extrapolation approach, BC mass concentrations with and without this correction could be compared to bulk BC mass measurements at the same site (e.g. Cozic et al., 2007, Cozic et al., 2008, Mertes et al., 2007, Petzold et al., 2007).

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L. 371: Fig.4A: Again a regression plot with linearly scaled axes would be more instructive. In such a plot it is much easier to evaluate the strength of correlation between BC mass and particle absorption coefficient, like it is usually done, e.g. Rosen et al. (1978, Applied Optics 17: 3859-3861), Petzold et al. (1997, Atmospheric Environment 31(5): 661-672), Mertes et al. (2004, Journal of Aerosol Science 35: 347-361). In addition the regression parameters should be given as well as their significance level. Maybe it might be useful to carry out separate regressions for the different discussed weather types. The resulting regression slopes (may be forced through zero) are another approach to determine MAC.

L. 387: Figure 4B: The frequency distribution of MAC show a non insignificant amount of MAC values above 40 m²g⁻¹. Could the measurement uncertainties mentioned later on (L.399-408) indeed explain such high MAC?

L. 389: A reference for the summer MAC is missing.

L. 401-406: This argumentation is not convincing, since the MAAP is only used to provide the absorption coefficient, which is a correct measurement, and not the BC mass. The arguments given here only affect the resulting MAC, but only up to values of about appr. 20 m²g⁻¹. Larger values are difficult to explain by enhanced absorption caused by coated and internal mixed BC particles, cf e.g. Fuller et al. (1999, Journal of Geophysical Research 104(D13): 15941-15954) and Martins et al. (1998, Journal of Geophysical Research 103(D24): 32041-32050).

L. 425-431: Is this only dependent on MF? One could suspect that the size of the BC particles should have an impact too. A large BC particle might have a thin coating, but the mass of the coating might be larger than a more thick coating on a small BC particle. Thus the large particle should experience a longer delay time as the small one, although the coating thickness and MF are smaller. Please comment.

L. 490-492: According to Fig.3 the period started with SE wind and did not change to SE. Moreover AW is an advective weather type where vertical exchange should be

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unlikely due to the authors explanation, but the SE influence is visible. This seems to be a non-consistent argumentation.

L. 551-557: Since the N wind periods have the same convective or mixed weather type with vertical exchange with the boundary layer, one should not call it free tropospheric background only, but "influenced by N wind" in the same way as for the SE wind periods. Or why should only the SE wind be able to vent anthropogenic pollutants to the measurement site? Why is CO not enhanced (according to Tab.2) during the "SE winds" compared to "background"?

L. 578: How is the concentration of precipitation particles measured? And what is the (size) definition of a precipitation particle used here? This should be given in the paper.

L. 613-616: Not all scavenged particles are lost by wet-deposition. According to the inlet description by the authors also residuals incorporated in to cloud particles are sampled, i.e. not only interstitial particles remain. This should be more precisely and carefully described.

L. 633-638: So what are these nearby sources in SE direction which creates low ME and high NO_x values in contrast to the other sources in SE direction which causes the high ME of 48 %?

L. 717: BC not BE.

L. 695-697: The results of Fig.12A have been already discussed before and given in Tab.2, so that this figure should be taken out.

Fig.12B: Neither in the plot nor in its caption is a description of the y-axis. Most likely it should be concentration of precipitating particles. Is it indeed cm⁻³ and not L⁻¹?

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

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