Response to Referee 2

Firstly we would like to thank the referee for the important comments to improve this manuscript, the following details our responses to each specified comment.

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.

Some discussions will be added in section 4.4 of the revised paper: "A median value 18 ng m^{-3} of BC mass loading from Jungfraujoch site for winter conditions was reported by Petzold et al. (2007), which falls within the results in this study. However, the results are lower than the long term averaged BC mass measurements reported by Cozic et al. (2007) during the winter time in 2004 and 2005 (54.2 ng m^{-3}), which may result from the different techniques for the quantification of BC mass, although it may also be that the site experienced fewer strong pollution events during this study." and "The mass fraction of rBC was also calculated by dividing the rBC mass loading with SMPS-determined total aerosol mass assuming an average aerosol density of 1.5 g cm^{-3} using the method of Cozic et al., 2008b. The calculated rBC mass fraction has only a weak dependence on SE wind or precipitation, ranging from 0.01-0.07 during this experiment, which agrees with the out-of-cloud results of BC reported by Cozic et al. (2008b)."

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.

The referee raises a valid point that the pollutants may come from the northern Swiss plateau, however the pollutants from northward were less intense and lasted only about 5hours during this study, which will not significantly influence the statistics of the entire dataset. Therefore this condition is not defined and discussed separately. This will be mentioned in the revised manuscript to clarify the confusion.

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

Corrected.

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.

Fig.1. will be modified accordingly in the revised manuscript.

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.

The weather types shown in Tab.1 are classified by Wanner et al., (1998). The occurrence of each weather type during this study will be presented in the revised manuscript.

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

The missing reference Baumgardner et al. (2007) will be added in the revised paper.

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 of this sentence.

The revised manuscript will include the following discussion: "Particles that can be optically sized by the SP2 are in the diameter range 200nm-720nm, and the optical size below or above the SP2 detection limit will not be triggered or will saturate the detectors respectively".

The referee has some questions regarding the comparison between the SMPS and the SP2 size distribution. As we have discussed according to referee 1, we agree and have removed this section

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 20nm, it would be even more justified to determine the ration 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?

See responses to referee 1 above.

We have chosen to plot the ratio as sub-micron/accumulation mode particle numbers because this highlights periods of high numbers of fresh particles, indicative of relatively fresh pollution. This can be related to the mixing state of BC, as is discussed section 4.5.

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?

Please refer to the answer to the comments made by referee 1 [Page 8775: Line 23]: a detection efficiency derived from SMPS/SP2 comparison will not be used in the revised manuscript. A scaling factor is applied to the measured rBC number concentration by a lognormal fitting on the number size distribution (section 4.1). This correction on the rBC number concentration will be more clearly illustrated in the revised manuscript.

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 etal., 2007, Cozic et al., 2008, Mertes et al., 2007, Petzold et al.,2007).

Please refer to the answer to the comment above regarding the rBC number fraction.

This part will be moved into the instrumentation part in the revised manuscript.

The extrapolated rBC loading is definitely higher than the uncorrected one, however, only the average value for the whole dataset can be compared with the other reports at this site (section 4.4), which means a real-time evaluation of the extrapolation methodology is not applicable.

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.

The linear plot along with the regression parameters will be shown in the revised manuscript.

Please refer to the answer to the comments by referee 1 [Page 8777: Line 24]: no obvious dependence of MAC on the weather type is observed.

L. 387: Figure 4B: The frequency distribution of MAC show a non insignificant amount of MAC values above 40 $m^2 g^{-1}$. Could the measurement uncertainties mentioned later on (L.399-408) indeed explain such high MAC?

The MAC above $40 m^2/g$ could arise from both the lower S/N ratio of the filter-based measurements and the systematic error to estimate the total rBC mass loading by extrapolating the SP2 detected mass size distribution, and the presence of light absorbing organic aerosols may also positively bias the filter-based measurements.

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

Added.

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 $m^2 g^{-1}$. 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).

We agree with this comment, the coating enhancement cannot solely explain the very higher MAC value, but may lead to a fraction of overestimated MAC. The texts will be amended in the revised manuscript.

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.

The definition of the MF includes the rBC mass as the numerator. Hence the MF will be positively correlated with the rBC mass and a BC particle with smaller rBC mass is more likely to be thickly coated, extending the coating evaporation time (T). An investigation on the relationship between the T and rBC mass validates this assumption, and the large BC particle is rarely linked with a longer T. The instrument was operated in a scattering triggering mode so that a small rBC with thin coating cannot scatter sufficient light to be detected by the SP2.

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

We thank the referee for pointing out the inconsistencies. The text will be amended accordingly in the revised manuscript.

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"?

Please refer to the answer to the second response.

The enhanced CO was indeed observed when SE wind, as the enhanced 75%, 90% percentiles show. However the overall CO concentration becomes more variable leading to reduced 10%, 25% percentiles. The CO concentration at this site is significantly controlled by the changeable background values, in addition to the variety of pollution sources, leading to a variable CO concentration when influenced by SE wind.

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.

Please refer to the answer to the comments of referee 1 [Page 8776: Line 6] and the end of section 4.2.

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.

The corresponding texts will be corrected as: "The aerosol present during periods when air masses were affected by precipitation were biased towards smaller sizes compared to the average particle distribution for the period, suggesting the larger aerosols are preferentially removed by scavenging of precipitation".

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 %?*

Solid fuel fuel combustion (wood) is the primary source of home heating in central Switzerland (e.g. Alfarra et al., 2007; Targino et al., 2009). Such sources are considerably closer to the site compared to the major industrial sources in the Po Valley and hence NOx concentrations have not yet reduced nor have BC particles become extensively coated. This will be detailed discussed in the revised manuscript.

L. 717: BC not BE.

Corrected.

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.

This figure will be amended in the revised manuscript.

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}*?*

It is L^{-1}, the plot is corrected.