Response to Referee 1:

Firstly we would like to convey our gratitude to the referee for the important and encouraging comments. The following details our responds to the specific comments.

I would like to recommend that the authors change BC to rBC throughout in order to be consistent with the recent publications by Schwarz and others.

The acronym rBC is now used throughout this paper to clearly denote the SP2-measured black carbon as refractory.

Page 8775: Line 23: The comparison of the SMPS with the SP2 in the same size range is very interesting but a 36% difference is fairly high. The explanation is that the lower size cut may actually be larger than the 200 nm mobility diameter of the SMPS. How much would the SMPS diameter need to be adjusted to account for the difference in concentration? Should there also be a correction for the difference between electrical mobility and optical diameter? I assume the SMPS was calibrated for the pressure of the research site?

The referee raises a valid point on the comparison between the SMPS and the SP2. We have further investigated the particle size distribution as measured by both instruments during the experiment, finding out the SMPS/SP2 (mobility/optical diameter) differences vary significantly at different particle sizes most likely as a result of dependencies on complex morphology, chemical composition, mixing state, fractal dimension, density, equilibrium water content and vapour pressure (e.g. Hering and McMurry 1991; Heintzenberg et al., 2002 and refs therein, Park et al., 2008). Performing a thorough comparison, taking these factors into account is beyond the scope of this paper and as a result we agree with the referees that using a comparison between the SMPS and SP2 to correct particle numbers is not appropriate. We will therefore remove this section in the revised manuscript and instead report the results directly from the SP2 measurements.

Are concentrations being reported at standard temperature and pressure or local conditions? In general, for comparison with other data sets, I think the mass and number concentrations should be normalized to STP.

Values are currently reported at ambient pressure, but we agree with this suggestion and will report the STP normalized mass/number concentration in the revised manuscript.

Page 8776: Line 6: The method by which precipitation type is determined needs clarification. Backscatter and forward scatter extinction is contradictory. Extinction, I believe, is by definition the sum of absorption and scattering. Given the potential differences in scavenging efficiencies of rain versus snow, I think it is important to explain the way this instrument derives the precipitation type.

We apologise there was an error in our sentence. The following discussion will be included in the revised manuscript: “This instrument primarily determines the

precipitation type by the measured ratio of the atmospheric back scattering coefficient to the atmospheric forward scattering coefficient. A ratio greater than a specific value indicates snow while a ratio lower than a threshold value indicates rain. As a secondary approach, the size and velocity distributions of the precipitation particles complement this primary measurement to provide information on different precipitation types."

In addition, it is important to know if the precipitation is localized or wider spread in order to understand how long the particles have been in cloud and the time available for wet removal. This probably means looking at the satellite images. The alternative is to use the back trajectory analysis that provide information on the liquid/ice water content along the trajectories.

We have analysed the back trajectories to show that precipitation occurred throughout the previous 3 days during the period from 26th to 27th February, whereas from 28th February – 4th March precipitation was more likely to be localised.

Page 8776: Line 17: It is mentioned that a significant amount of the rBC mass will not be measured at the small end, but what about the mass of rBC that saturates the detectors, i.e. rBC in the large particle tail? I think that this is an important parameter to evaluate, along with the other indices like the mixing efficiency. Although you can't determine how much actual mass is, the percentage of particles causing saturation can be evaluated and could be a sensitive indicator of washout.

Thanks to the referee for this suggestion. The larger rBC particles that saturate the detectors have been evaluated to represent about 0.2-0.6% of the total population of rBC, which is of minor importance in influencing the derived rBC total mass loading, and this fraction had little variation under different conditions during the experiment. These discussions will be added in the revised manuscript.

Page 8777: Line 24: As seen in Fig. 9, sometimes the log normal fit is very good but in the high BC cases there is a larger difference, particularly in the big particle tail. This uncertainty should be quantified somewhere. From Fig. 4 there is more scatter than I would have expected between the SP2 and the MAAP. Are these comparisons only in cloud-free air? Is this indicative of the uncertainty in deriving total rBC mass from the SP2? There are very few comparisons between filter-based light absorption measurements and instruments that measure BC or EC directly. Although it is somewhat outside the scope of this paper to evaluate the MAAP, the data set is rich in the variety of rBC types. If the primary limitation of filter based absorption measurements is the interference by scattering, it could be interesting to color code Fig. 4 based on the rBC mixing efficiency to see if there are systematic difference related to the relative amount of non-absorbing coating on the rBC.

The uncertainties to determine the MAC values could arise from the random noise of the filter-based measurements and the systematic error to estimate the total rBC mass loading by extrapolating the SP2 detected mass size distribution with the assumption that the lognormal distribution could represent the rBC modes outside the SP2 detection. The

integrated area of the lognormal fitting within the SP2 detection size range is compared to the originally measured rBC mass, the difference is within ±8%, and this discrepancy is not dependent on the level of the rBC mass concentration. Moreover, the MAC values become more scattered when rBC mass is lower, which may result from the decreased instrument S/N ratio when the measured concentration is low. In addition, the high MAC ($>20 m^2\ g^{-1}$) is investigated to be more frequently associated with lower rBC mass, but independent on different conditions.

The results presented in Fig.4 are for the entire dataset. The MAC values are also investigated for the in-cloud or out-of-cloud air respectively, however no significant differences are observed.

No obvious relationship between rBC mixing efficiency and MAAP/SP2 discrepancy is observed for this dataset. The MAAP/SP2 difference may arise from the systematic uncertainties from both instruments.

Page 8779: Line 12: "The advantage of using MF to characterize the coating abundance is that this definition avoids the sphericity assumptions made when calculating a coating thickness." Doesn't the denominator of the equation for MF require the assumption of asphericity?

We thank the referee for this point and have removed the sentence.

Page 8780: Section 4.3: Previous studies of air mass aging, for example many of those published in the MILAGRO special issue, normalize the particle mass concentrations with the CO minus the background CO. Doing this type of normalization with these measurements might help to highlight the differences in the air mass histories and the various degrees of aging.

The rBC mass has a relatively weak linear correlation with CO mixing fraction for this dataset compared to the other urban studies because of the low concentrations for both rBC and CO, leading to the difficulty in determining the background CO concentration. However, we have reanalysed the data by estimating a background CO concentration from the frequency analysis in Fig.8 B: The peak of the lowest mode of the CO density function is about 120ppbv and likely represents the background state. This value has applied to the dataset to obtain the ΔCO. This will be discussed and the BC/ ΔCO will be analysed in the revised manuscript.

Page 8781: Line 9: "The pollutants were rarely removed by wet deposition during this phase, as shown in Fig. 3, and hence BC mass loading reached its maximum during the experiment." I don't understand how this can be concluded from the available measurements. I also think that dry deposition needs to be at least acknowledged as a possible source of removal.

We agree what the referee comments, will amend the texts in the revised manuscript.

The other parameter that I think is very useful is the mass equivalent diameter (MED). This is one of the statistics that can be easily derived from the mass size distribution and may also be related to the age and mixing state of the aerosols. I would really like to see if it changes significantly depending on the air mass history.

The SP2 operation was not under the optimum condition during the experiment due to the systematic triggering issue, causing a <90% collection efficiency for the rBC below 200nm. The lognormal fitted rBC size distributions consistently peak around 200-210nm under different conditions. The geometric mean MED without the lognormal fitting is also calculated within the SP2 detectable size range (190-720nm). A slightly increased rBC MED is observed when influenced by SE wind compared to the background condition or influenced by precipitations. The revised manuscript will have a discussion on this.

Page 8782: Line 22: "The measured results in the free tropospheric background are statistically analyzed and presented in the first row of Fig. 8.". Maybe I am just arguing semantics but this isn't really a statistical analysis. I would call it more a graphical analysis since these are frequency histograms. I am also quite surprise how high and variable the CO values are for being free tropospheric. How do these compare with published values from other mountain site or airborne measurements?

The text will be revised as: "Frequency distributions for black carbon mass and CO are shown in the first row of figure 7."

A detailed analysis of the perennial CO time series and an intercomparison of 4 standard techniques for CO measurements at Jungfraujoch have recently shown that the CO instrument used here (Horiba APMA360; based on non-dispersive infrared (NDIR) absorption technique) is well suited for CO measurements also at remote sites such as Jungfraujoch (Zellweger et al, 2009). Zellweger et al. have also shown that the agreement with other measurement techniques for CO is very good and that the NDIR technique provides reliable data for hourly averages and higher aggregates. The 4-instrument-intercomparison in 2006 also revealed short episodes with enhanced CO variability observed by all techniques (Zellweger et al., 2009). Thus, the strong short-term variability seen in Fig.7 at the end of period III is most likely a real effect and not related to a poor instrument performance.

The mean annual CO cycle at Jungfraujoch shows a distinct seasonal cycle with a peak-to-trough amplitude of about 50ppb with highest monthly means in February to April (mean monthly averages 2000-2007 period: February 152.2ppb, March 151.8ppb, April 160.6ppb) and lowest values in July to October (mean monthly averages 2000-2007 period: July 111.3ppb, August 112.2ppb, September 112.6ppb, October 110.6ppb). The mean CO mixing ratio during the illustrated period in Fig. 7 (February 25 to March 14) is 151.4ppb and is thus higher than the average for February (140.4ppb) but lower than March 2007 (164.6ppb) (Zellweger et al., 2009).

Continuous CO time series of high quality at elevated sites for a direct comparison is rare. However, the CO data at Jungfraujoch are comparable with CO data from flask analysis at other elevated sites. Mean CO data at Niwot Ridge (40 deg N, -106 deg E, 3523m asl, i.e. similar latitude and altitude) range from 143.5ppb (monthly mean in March, 2000-2007 period) to 104.6ppb (August). These are data from the NOAA/ESRL Cooperative Air Sampling Network and are thus based on analysis of discrete flask samples that are taken once a week. The NOAA flask sampling network aims at identifying long-term trends, seasonal variability, and spatial distribution of trace gases. Thus, the flasks are usually filled around noon local time and cover only sporadically regional pollution episodes. This can explain the slightly lower mean CO mixing ratios observed there.

The Jungfraujoch CO data do also fit very well with the marine boundary layer reference data from the Globalview Project (GLOBALVIEW-CO, 2009). The Globalview Project generates these background CO values as a function at latitude based on all CO data from the NOAA/ESRL Cooperative Air Sampling Network.

Page 8784: Line 26: It is difficult to tell from these curves that they are the same shape or not. This is where having additional parameters like the MED and maybe even standard deviation would be better indicators. Also, the number of particles saturating the detector I think is also important. One way to compare the curve shapes is to normalize them by the area under the curve.

The answers to the comments of referee 1 in [Page 8781: Line 9], [Page 8776: Line 17] and [Page 8777: Line 24] apply to the comments here with respect to MED, particles saturating the detector and underneath area of the curve respectively.

References

GLOBALVIEW-CO – Cooperative Atmospheric Data Integration Project - Carbon Monoxide. CD-ROM, NOAA ESRL, Boulder, Colorado [Also available on Internet via anonymous FTP to ftp.cmdl.noaa.gov, Path: ccg/co/GLOBALVIEW], 2009.

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