Reply to:

Anonymous referee #3

Referee comments in black, author replies in blue

This manuscript reports results obtained with a time-of-flight aerosol chemical speciation monitor (ToF-ACSM) during a long-term measurement period (14 months) at the Jungfraujoch research station, Switzerland. The authors described the seasonal and diurnal variations of non-refractory submicron particles (NR-PM1), and discussed the properties of NR-PM1 as a function of the air mass origin. The exceptional situation of the Jungfraujoch (a high elevation site in the heart of Europe) allowed the authors to compare on one hand free tropospheric air masses with periods influenced by the planetary boundary layer (PBL), and on the other hand air masses from different regions of Europe. They also performed a source apportionment of organics by PMF analysis, and discussed in detail the sources and processes of organics based on mass spectra (also f44 vs f43), diurnal and seasonal variations.

This manuscript is very interesting, and fits totally the scope of the journal. I recommend its publication in ACP after the authors address the following comments.

Specific comments:

1) Page 18229, line 25 to page 18230, line 2: if the authors used UTC for the entire dataset, it means that they did not take into account the daylight saving time in summer. This introduces a bias in the diurnal patterns of anthropogenic species, given that human activities are shifted by 1-hour between summer and winter.

Yes, UTC was used throughout the document. Diurnal plots however are always shown separate for all seasons. The return to standard time in 2012 was made on October 28 and the switch to daylight saving time in 2013 on March 31. Therefore no significant bias is expected in the diurnals of winter and summer. Also spring 2013 is unaffected due to the interruption of the measurements during most of March 2013 (see Fig. 1). The time shift in October took place after about half of our "autumn 2012" period has passed, i.e. the autumn diurnals might be affected. However, the emission times of the local pollution are limited by the train timetables which are also changing with season. The first train arrives at 08:52 LT (07:52 UTC in winter, 06:52 UTC in summer) year-round while in winter (October 29 – March31) the last train is leaving JFJ earlier at 16:50 LT (15:40 UTC) than in summer, when the last train is leaving at 17:45 LT (15:45 UTC). Consequently, the application of UTC prevents a bias at the end of the day but might cause one in the beginning of the day in autumn. No changes were made since we do not think a shift to LT would improve the manuscript.

2) Page 18231, lines 12-16: it would be very useful for ToF-ACSM users to include in the supplementary material a table with the details of the different calibrations for each month with the statistics (average, standard deviation, etc.). This information is important to assess the stability of these instruments, and to determine at which frequency they need to be calibrated.

We agree with the referee that such information is useful for ToF-ACSM users and added the standard deviations of the RIE values to the text. However, the mIE value does depend on the instrument hardware and TOF voltages ; changes in these may result in significant changes in the ions/pg measured. This was the first field deployment and the instrument underwent several hardware upgrades and consequent tunings of the TOF voltages. Therefore no statistics on the mIE value are reported in the paper. With the different instrument voltage profiles, mIE values between 26 and 103 ions/pg were recorded over the measurement period. After each bigger intervention (e.g. opening of the vacuum chamber, exchanging of components, transport) the mIE value was re-calibrated and adjusted in the subsequent data analysis.

3) Page 18234, line 5: I am surprised that the authors performed a back trajectory analysis over 10 days. This is too much, given that beyond 3 days, the uncertainty on the back trajectory calculation is already high. Indeed, according to Rolph and Draxler (1990), the horizontal deviations are between 200 and 700 km at 4 days, depending on the spatial and temporal resolution of the meteorological data.

We are not using single trajectory analysis in this work (as in Rolph and Draxler, 1990) but simulations of a Lagrangian particle dispersion model (LPDM). The difference being that the latter simulates the transport of thousands of air parcels for a given release time and describes their movement with the mean flow, but also turbulent and convective transport. One of the sources of uncertainties in single trajectory models is the assumption of a not dispersing air parcel. Hence, divergence in the flow field will lead to large uncertainties in the trajectory calculation. In an LPDM, divergence just adds to the overall dispersion of the particle plume through turbulence. In this context we strongly believe that running our model 10 days back in time is not using it beyond its abilities. Also others have used similar or longer backward integration times to categorise transport to measurement sites (e.g. Hirdman et al., 2010).

4) Page 18248, line 6: in the absence of a thermodenuder and information on the volatility of the particles, I think it is not appropriate to use the “LV-OOA” terminology in this study. Even “more oxidized OOA” (MO-OOA), which is found in papers with HR-ToF-AMS datasets, cannot be used, given that we do not have the O/C ratios. I would suggest to use “OOA I” and “OOA II” throughout the manuscript, which is recommended by Zhang et al. (2011) in this case.

We agree with the referee’s position that without a thermodenuder we do not have direct evidence of the volatility of our OOA. However we are convinced that we are dealing with LV-OOA. We made our decision to attribute the OOAs to the LV-OOA family on the basis of spectral similarities to the ones shown in Ng et al. (2010). There LV-OOA profiles show high f44 (0.17±0.04) while SV-OOA shows an f44 range of 0.07±0.04. Furthermore, according to Jimenez et al. (2009) LV-OOA has O:C ratios higher than 0.58 in rural areas, using the formula of Aiken et al. (2008) this corresponds to f44 higher than 0.13. Our OOA profiles show always f44 ratios above 0.20 (see Fig. 10d). Even taking into account a potential positive bias of f44 in the ACSM compared to the AMS as reported in Fröhlich et al. (2015) and Crenn et al. (2015) this indicates rather an LV-OOA than an SV-OOA. We clarified our selection in the text by an additional sentence following the introduction of OOA on P18248 L7. Also the paragraph on P18249 L6 contains a note that the decision was not made on the basis of direct volatility observations.

Technical corrections:

All mentioned technical corrections were taken into account in a revised document.

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