Atmos. Chem. Phys. Discuss., 15, C5573–C5578, 2015
www.atmos-chem-phys-discuss.net/15/C5573/2015/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Fourteen months of on-line measurements of the non-refractory submicron aerosol at the Jungfraujoch (3580 m a.s.l.) – chemical composition, origins and organic aerosol sources" *by* R. Fröhlich et al.

Anonymous Referee #1

Received and published: 4 August 2015

This article presents a fourteen-month dataset of the chemical composition of submicron aerosols at a high altitude site in the Swiss Alps. Long-term datasets at high time resolution are rather new, as well as the instrumentation used (TOF-ACSM), which makes this study unique and very interesting. The topic is obviously within the scope of ACP. I recommend publication in ACP after all major and minor comments listed below are addressed by the authors.

Major concerns.

First, I suggest the authors to reorganize the introduction. In its current form, I find it pretty hard to follow. I would have expected a thought process like: Why is it important to measure in FT? Why long term measurements? Why MS techniques rather than filters? Why ACSM? Why TOF? All of these questions were not clearly answered.

Second, I have serious concerns about the organic mass spectra analysis, which I found too weak to be published as is.

- ME-2 was performed seasonally, because the use of fixed factor profiles over long term periods is conceptually inappropriate (and I fully agree with this). It would have been appreciated to clearly see the benefit of this approach compared to one single PMF run (leading to unidentified factors, or high seasonal residuals for example?). Seasonal PMF of OA is a new topic, the authors need to provide more information.

- eBC data are available. Why this hasn't been used to optimize a values, as presented in Frohlich et al., 2015? More globally, some papers (Crippa et al., 2014; Frohlich et al., 2015) clearly give guidelines for OA source apportionment, that don't seem to be followed in this study. Is the JFJ site too specific, so that these guidelines don't apply? Also, Q values doesn't seem to be used here, but it is an essential mathematical parameter of PMF analysis; is there a reason for that?

- The profile variability also theoretically applies for air mass origins (clusters) and FT/PBL, where different kind of profiles can reasonably be expected. From Fig10, the f44 & f43 are mostly influenced by seasons, and not much by the air mass origins or FT/PBL (which is somehow surprising, but is a very important result that is not really discussed). This means that doing PMF differentiated by clusters or FT/PBL (which would have been appreciated in this study) would not help much; but in Fig10, POA factors were subtracted, which leads me to another major concern :

- While I don't disagree on the method used to extract POA profile, I have serious doubts on the outcome. How do the authors justify the fact that no m/z44 is present in the profile, as opposed to the CSOA profile from Faber et al. (2013)? From the

ACPD
15, C5573–C5578, 2015

ACSM intercomparison, the TOF ACSM has the highest f44. It wouldn't thus have been illogical to find higher f44 in POAloc than CSOA; but the total absence of m/z44 cannot be justified by the f44 instrument-to-instrument variability. Could it be that the high S/N of 44 and a non-optimized downweighting can cause the quasi exclusive apportionment of 44 into OOA profiles? One simple test would be (if some of the authors are smokers) to smoke just below the ACSM inlet to have a clear instrument-specific signal of cigarette smoke; have the authors tried that? In its current form, I don't believe the POA profile to be real. If the authors think it is, then they need to better prove it. Thus, I am not convinced with Fig10 (although the idea is excellent) and its interpretation.

- I also have concerns with BBOA factor. From Fig7, f60 has a nice seasonal variability with highest values in winter 2012/2013 (and also widest distribution, so we except significant temporal variations). However, from Fig.S6, BBOA during this period looks like noise, has no significant temporal variation, and overall level is very near the LOQ (if LOQ=10*LOD/3) of OA. . . More importantly, still in Fig.S6, it is in summer 2013 that highest BBOA are measured. But from Fig.7, summer 2013 does not exhibit high f60 values. These two figures thus provide contradictory information. Can the authors provide an explanation for that?

Minor comments.

- P18227, l1: While I don't see how history can be combined with future, this first sentence is way too navel-gazing. Peace around the world, starvation, poverty, are also some great challenges for humankind. Yes, our work is important, but please moderate.

- P18227, l24: what is a "good" time resolution?

- P18228, l10: please provide an approximative number of measuring locations for the observation of free tropospheric aerosols. Give some examples.

ACPD
15, C5573–C5578, 2015

- P18229, l21: talking about representativity is important. How representative is the presented dataset compared to several-decade trends? This would be a very interesting input to the paper, as it helps interpreting the results.

- P18229, l25: please clearly state how seasons were differentiated

- P18231, l14-15: how did the calibration values (IENO3, RIENH4 & RIESO4) vary over time? As monthly/bimonthly calibrations were performed, please provide a standard deviation.

- P18231, l20: the CE=1 is expected at JFJ, but is unexpected some lines later (p18232, l2). Please revise

- P18231, l20: a good correlation (r or r^2) will still be obtained with other CE values. Only the slope of the correlation will change. Please be more specific

- P18231, l25-26: please clearly state how SDE were identify, or add a link to section 2.6.

- P18231, l29: SO4 and OM are not always > 80% of total PM1 (see p18238, l8). Please revise

- P18232, l25: saying that eBC concentrations are subject to uncertainties is way too vague. Please provide an estimate of the uncertainties associated with this measurement. If not possible, please discuss it a bit more in details. By the way, why eBC data haven't been corrected?

- P18235, l19: the recommendations from Griffiths et al. (2014) is 0.75 – 2 Bq.m-3. So why did the authors choose 1.5? Is it an empirical determination?

- P18236, l6-8: so what is the take-home message of section 2.5? How does the choice of the methodology change the interpretation? Why didn't the authors perform a "careful exploration" as in Herrmann et al. (2015)?

- P18237, l3-4: please revise Fig1b. There is supposed to be a continuity of seasons

ACPD

15, C5573–C5578, 2015

throughout the year.

- P18237, l5: “in the summer months”. Months have not been used to differentiate the seasons. Please revise

- P18248, l8: m/z60 is not the principal fragment of BBOA. It is rather the most specific fragment of its profile.

- P18248, l19: what is the influence of small variations of a value for POAloc (+ or – 0.05)?

- P18251, l20-21: the summer 2013 eBC concentrations is similar to summer 2012. Diesel engine generators don’t emit OA? This explanation is too vague, and in case these engines occur to be a significant eBC source, why didn’t these periods deleted from the PMF analysis?

- P18251, l26-27: this explanation is very subjective and is not illustrated. From what I can see in Fig. S6, the temporal variations of BBOA in winter 2012/2013 looks like noise and doesn’t really follow OOA general trend. Then, BBOA is much higher in summer, but is not discussed. (see major concern)

- In the beginning of section 3.2, the authors wrote that “Figures 7a-e show the fractions of total OA). But in the caption of the figure, it is stated that local primary OA have been subtracted. If it is the case, then see my concern for POA above; if not, then delete this from the caption.

- Table S2 : this table has nothing to do with HOA – eBC ratios. Please revise

Technical comments.

While the general language is correct (and appreciated), many typos are present in the text. Some obvious ones are listed, but I suggest the authors to thoroughly proofread the manuscript.

- P18226, l5: please replace a.s.l. by above sea level

ACPD
15, C5573–C5578, 2015

- P18227, l13: please replace real time by near real time

- P18227, l14: near real time and high time resolution is equivalent. Please choose one or the other

- P18228 ,l17: please remove one “and”

- P18228, l17: please insert a “–“ in “gasphase”

- P18238, l15: please remove one “which”

- P18238, l22: “In autumn, winter and spring”

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 18225, 2015.