Reply to:

Anonymous Referee #1

Referee comments in black, author replies in blue

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.

The introduction was rearranged according to the suggestion and a few sentences were added to clarify and answer the stated questions.

1. Why measure in the FT

"Knowledge of the aerosol concentrations and properties in the lower FT is important due to its impact on cloud formation and to validate model results from the FT."

2. Why long-term?

"Long-term measurements are essential to capture long-term trends, inter-season variations or other effects taking place on longer time scales. Furthermore, increasing data coverage allows for a more reliable determination of the typical aerosol conditions at a specific site."

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.

Since in many regions distinctively different sources for primary aerosol or precursors for secondary aerosol formation exist during the various seasons (especially winter/summer) it is generally advised to perform separate PMF source apportionment due to the nature of inflexible source profiles within one analysis. This is investigated on the basis of the OOA factors in Canonaco et al. (2015) (doi:10.5194/acp-15-6993-2015, in the manuscript the discussion version of that paper is referenced

because the final version was not yet available at the time of submission) and the authors conclude that "PMF analysis of multiseason data employing only two OOA factors cannot capture the seasonal variability of OOA." Additionally, a primary source which is not present during the whole period (e.g. BBOA) is likely to not be extracted properly by a multi-season PMF. It becomes more and more likely that e.g. a non-physical splitting of a factor present during the whole year reduces the PMF residuals more than the addition of a small but real primary factor making it impossible to retrieve this factor. In their guidelines for PMF on AMS data Ulbrich et al. (2009) state that factors contributing less than 5% are often difficult to extract effectively. Seasonal PMF for long-term datasets was also performed before by Minguillon et al. (2015) (doi:10.5194/acp-15-6379-2015) and Tiitta et al. (2014) (doi:10.5194/acp-14-1909-2014). Parworth et al. (2015) (doi:10.1016/j.atmosenv.2015.01.060) even took it one step further and ran separate PMF on shifting two-week windows (which they call "rolling window technique") over a full period of 19 months to capture changes in the OOA and BBOA factors. Hence, a separation into seasons increases the chances to find factors only present during part of the year. Since these limitations have already been discussed elsewhere and are reviewed in our manuscript (addition of a paragraph after P18247 L 9) we don't expect to be able to draw additional conclusions from our dataset and did not perform additional multi-season PMF.

eBC data are available. Why this hasn't been used to optimize a values, as presented in Frohlich et al., 2015?

Yes, eBC data is available but due to the very low concentrations no reliable eBC source apportionment is possible at the JFJ. Additionally, while an optimisation of the a value with correlation to external tracers like eBC usually is an adequate and reliable way to constrain a solution we do not think that this holds true in the case of the JFJ. At the JFJ, aerosol of all sources (except POAloc) is transported vertically to the JFJ at the same time resulting in similarly good correlations of all sources with eBC, see e.g. Fig1c. The JFJ specific challenges are discussed on P18247. As mentioned there, we were unable to select an optimal solution (i.e. optimal a values) and instead report the mean result of all solutions from a range of reasonable a values. This can be called a form of "extended chemical mass balance" where a range of fixed profiles is considered while leaving the OOA unconstrained.

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?

We are well aware of these guidelines and tried to follow them as much as possible but due to the specific conditions at the JFJ high elevation site not all were applicable. Below, the recommendations are listed and commented separately.

Crippa et al. (2014) report the following methodology:

1. Unconstrained run (PMF).

This was our first step, which did not result in stable and unambiguously interpretable solutions.

2. Constrain the HOA mass spectrum (MS) with a low a value (e.g., a =0.05–0.1) and test various number of factors.

The HOA was constrained and the mean result of solutions within the HOA a range given in TabS1 is reported.

3. Look for BBOA (if not identified yet): constrain the BBOA MS if f60 (i.e., the fraction of m/z 60 to the total organic mass) is above background level and check temporal structures like diurnal increases in the evening during the cold season due to domestic wood burning (suggested a value = 0.3–0.5).

f60 was investigated and elevated contributions, especially in winter, lead to the introduction of BBOA in the source apportionment but only the seasons winter 2012/2013 and summer 2013 led to contributions above 1% which is why BBOA was not considered during the other seasons. The mean results of solutions within the BBOA a range given in TabS1 are reported.

4. Look for COA (if cooking not found yet): check the f55–f57 plot for cooking evidence (where f55 and f57 are the fraction of m/z 55 and m/z 57 to the total organic mass respectively; see Mohr et al., 2012). Fix it in any case and check its diurnal pattern (the presence of the meal hour peaks is necessary to support it at least in urban areas).

The vertical transport pattern makes it very difficult to separate HOA and COA arriving from the valley due to the absence of usable diurnal cycles which usually present a strong argument for COA. A note that it cannot be excluded that part of the advected HOA actually is COA was added to the text.

5. Residual analysis: a structure in the residual diurnals might indicate possible sources not separated yet by the model (refer to Section 3.2.3). For each step the residual plots should always be consulted in order to evaluate whether the constrained profile(s) has(have) caused structures in the residuals. If so, the constrained profile should be tested with a higher scalar a value.

Residual analysis is a very important step and was conducted throughout the analysis. Diurnal average residual plots for all seasons have been added to the supplement.
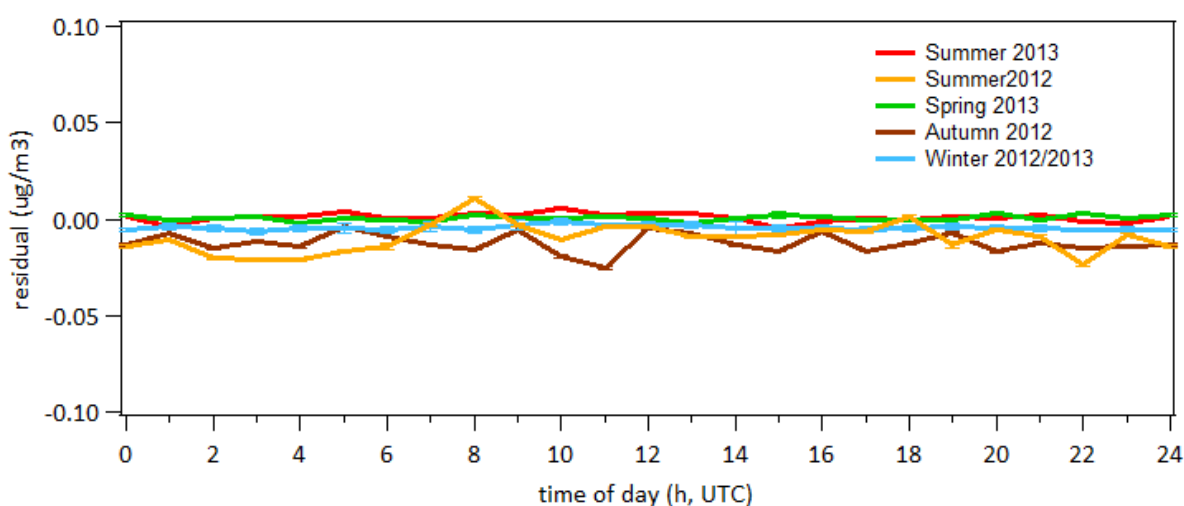

Figure 1: Average diurnal residuals in ug/m3. The mean of all ME-2 runs included in the presented solutions is shown. The error bars indicate the first standard deviation.

Residuals are close to zero during all seasons. No significant structure remained in the residuals. During some of the seasons (winter, autumn and summer 2012) slightly negative residuals are observed indicating a slight overestimation of the OA factors.

6. In general the OOA components are not fixed, but are left as 1 to 3 additional unconstrained factors.

1-2 unconstrained OOAs are reported depending on season.

Fröhlich et al. (2015) give the following recommendations

Profile constraints on the m/z 44 signal should be avoided or kept as loose as possible (high a value for m/z 44).

As mentioned in a comment below the extracted POAloc exhibits f44 of zero and for HOA (f44 < 0.02) and BBOA (f44 > 0.03) large ranges of a values were explored and taken into account in the reported solution.

If constraints are applied to the m/z 44 signal, a sensitivity analysis, e.g. by manual modification of the relative amount of the m/z 44 signal is recommended.

The absence of usable external tracers at the JFJ to help optimise solutions makes it impossible to ascertain the correct f44 value. However, considering the CE = 1 at the JFJ makes it unlikely that f44 is increased a lot in the ToF-ACSM data from the JFJ, see reply concerning the f44 in POAloc.

Anchor profiles constructed from the studied data set are preferable to database profiles. These profiles can often be extracted from solutions with additional factors (e.g. this study) or from separate PMF on parts of the data set with high fractional contributions of a factor (e.g. period with nearby forest fires or high primary traffic emissions).

The POAloc was extracted directly from the dataset. For HOA and BBOA this was not possible.

The PMF results of short-term, high-resolution AMS measurements overlapping with long-term ACSM measurements can provide useful constraints on the source apportionment of the ACSM data set (e.g. number of factors, special features in a profile).

Not applicable here.

If no profiles can be extracted with the methods described above, it is advised to try and compare different database anchor profiles (e.g. by comparing SA results to external data or comparing changes in diurnal cycles). This is more crucial for factors for which the profiles typically show larger variations between sites (e.g. BBOA, see Ng et al., 2011b) as opposed to factors with more similar profiles (e.g. HOA, see Ng et al., 2011b).

The absence of usable external tracers to optimise solutions makes it impossible to judge which profile gives the more exact solutions. Therefore the averaged reference spectra from Ng. et al. (2011) were used. The reporting of the mean of many solutions spanning a wide range of a values reduces the bias towards a reference.

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?

Q values are more important for pure PMF source apportionments, by applying constraints with ME-2 the Q-value is modified (increased) and does not give much additional information compared to a close look at the residuals. Already Ulbrich et al. (2009) point out in their conclusions that the change in the Q values does not lead to strong conclusions for the real world data case they investigated. For this reason Q values are not discussed.

The profile variability also theoretically applies for air mass origins (clusters) and FT/PBL, where different kind of profiles can reasonably be expected.

Yes, but since we are not able to make a definitive selection (no optimisation of a values like e.g. in Fröhlich et al. (2015) possible) we report the mean of a range of solutions with different a values which covers small profile variations. Larger profile variations would show up in the residual plot (e.g. increased residual at m/z 44 if the air masses from one region were significantly more aged. No such increased residuals were observed. The difference FT/PBL is inherent in the separation between the two OOAs. The contribution of the OOA with the lower oxidation is increased during periods with increased PBL air injection (see FigS13). This is also discussed in the text, e.g. on P18249 (top) or P18254 (middle) or in Fig. S8.

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).

We indeed think the f44 vs f43 is affected by the vertical transport pattern, i.e. FT/PBL. The change in the strength of the vertical transport causes most of the seasonal differences. In the warmer season more of the less oxidised OOA fraction is transported to the JFJ. This is discussed in Sect. 3.2.2. Also we think that the difference FT/PBL in Fig 10 is only little smaller compared to the difference between the seasons, indicating that not all of the seasonal difference can be explained by the increased vertical transport but also by different OOAs (see squares in Fig 10d). Air masses transported from the PBL to the JFJ can already be well aged due to the increased photochemistry on days with fair weather favouring thermally induced vertical transport.

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)?

We observed frequent short-term OA peaks at the JFJ which are also evident in the particle number and only appear during the daytime and only during hours when tourists are present. These short-term spikes can only originate from a local primary source. Inclusion of the profile of these short-term peaks (resembling very much primary OA profiles containing high fractions of aliphatic compounds) into the ME2 analysis enabled a clear separation of all short-term peaks from the rest of the data. This fact represents a very strong additional evidence for a local source. Because of the mentioned reasons the factor is termed local POA. The only potential sources for local primary emissions are limited to activities related to tourism. The options are: fossil fuel combustion, cooking and cigarette smoking. By weighting the evidence we concluded that cigarette smoke was the most likely source (but still refrain from calling it cigarette smoke OA). Due to the frequency and regularity of the spikes fossil fuel combustion can be excluded (compare Fig. 1 below and Fig. S10 in the supplement) because these emissions would either be caused by scheduled construction activities,

irregular helicopter flights or snowcat emissions (maximum once or twice a day). See also discussion in the text on P18250.
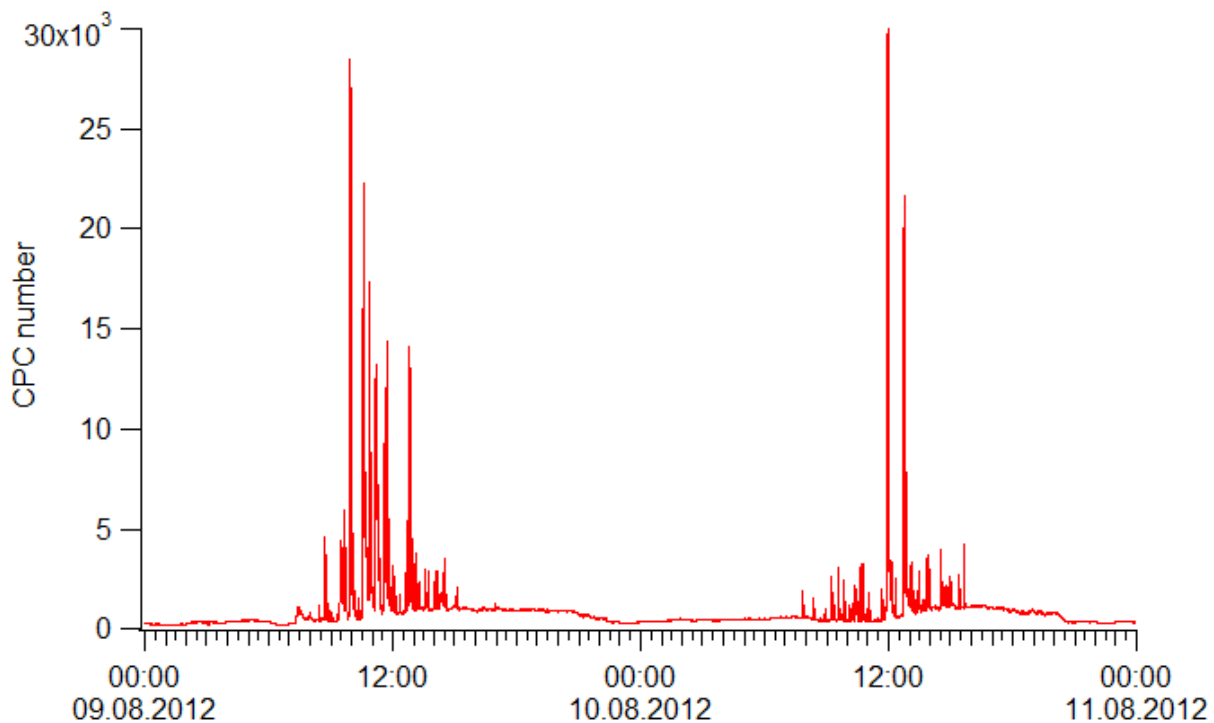

Figure 2: CPC number concentration with high time resolution. The time with peaks correlates with the timetable of the Jungfraujoch trains.

Correlations of POAloc with COA and cigarette smoke OA showed larger similarities with the cigarette smoke OA profile provided by Faber et al. (2013) Additionally, environmental tobacco smoke markers were measured at the same site before by Morrical and Zenobi (2002)and nicotine is one of the major ions seen by colleagues on site with an API-TOF (personal communication with Carla Frege, Paul Scherrer Institute). Further evidence for tobacco as the source of POAloc is given by the increased frequency of signal spikes on days with agreeable weather (see Fig. S9), no night-time events and the increased response of the near-ultraviolet wavelength signal of the Aethalometer (370nm, see Fig. 3 below) during POAloc spikes. On many occasions (prerequisite is that the ToF-ACSM is not in filter mode and the Aethalometer is measuring at the correct wavelength), the POAloc spikes appear concurrently with an increased Aethalometer signal at the 370 nm channel. Tobacco smoke is known to have increased optical absorption at the near-ultraviolet wavelength of 370 nm (see Aethalometer manual: Hansen (2005)). Fig. 3 was added to the Supplement and a paragraph was added to the text. However, as mentioned in the text, the application of the cigarette smoke OA profile of Faber et al. (2013) or other COA profiles both did not result in good separation of the short-term peaks, i.e. in increased mixing with the HOA factor. For comments concerning the absence of f44 in the POAloc, see the replies below.
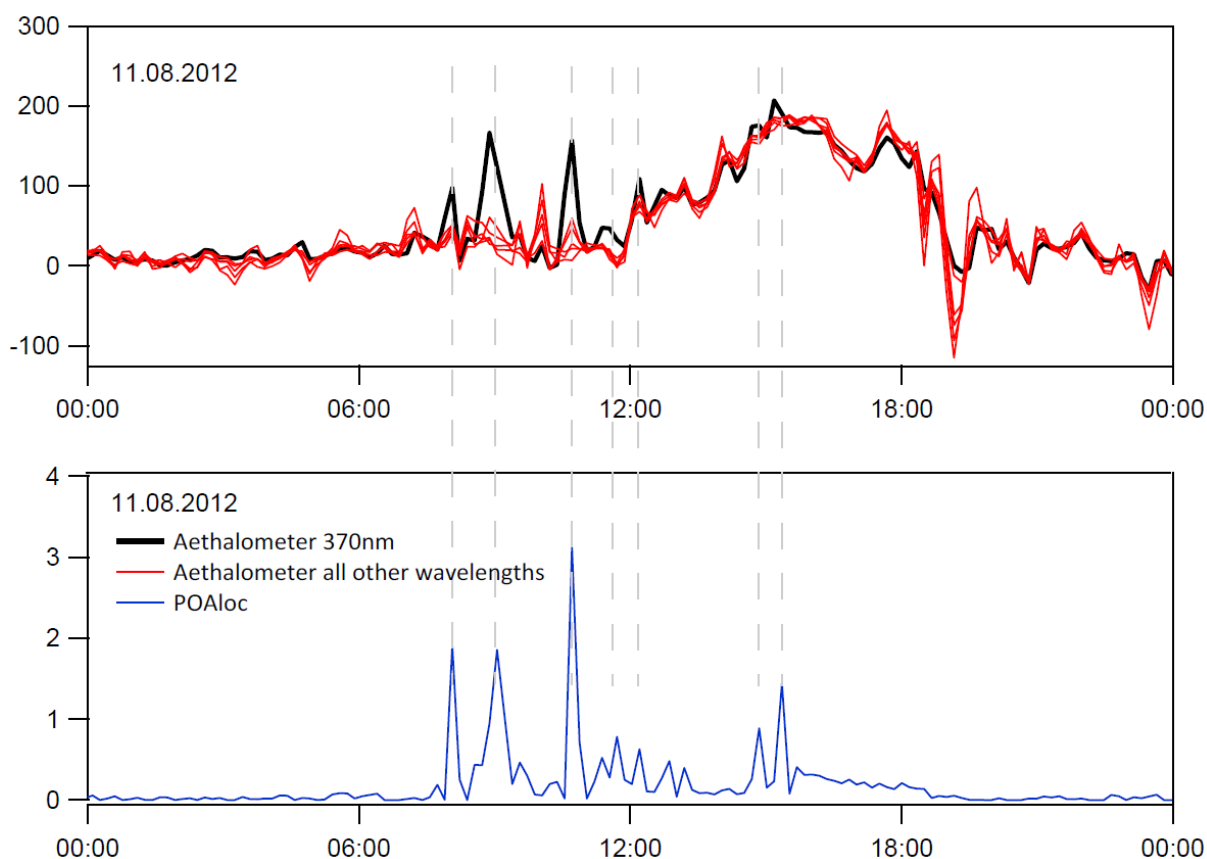
Figure 3: Top: example time series of the Aethalometer Aug 11, 2012, black: 370nm signal, red: signal of all other wavelengths. Bottom: time series of POAloc during the same day. Dashed grey lines are added manually to guide the eye. Cigarette smoke is known to have increased optical absorption at the near-ultraviolet wavelength of 370nm.

From the 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.

Indeed, the ToF-ACSM exhibited the highest f44 during the ACSM intercomparison. But this does not necessarily imply that the f44 measured with the ToF-ACSM at the JFJ would have been relatively elevated. Firstly, the extent of the elevated f44 in the ACSMs compared to the AMS depends on instrument history and increases after exposure to high mass concentrations. The ToF-ACSM was brand new at the JFJ and measured only low concentrations; in contrast, before Paris the instrument was exposed to large loadings during laboratory testing and then experienced high loadings during the study. Secondly, the additional f44 in the ACSMs is believed to originate from delayed further oxidation of other organics in the vaporiser region (this is still under investigation and several publications are in preparation) but at the JFJ we already see collection efficiencies close to one also with the AMS, leaving only very few material to be further oxidised and increase f44.

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?

It is possible that the separate PMF on only the spikes is prone to larger uncertainties because of the limited number of points and underlying variability. The big competition of the f44 by the OOA may lead to the underestimation of the f44 in POAloc but also in Faber et al. (2013) f44 is only a minor fraction of the laboratory measured CSOA (~3-4% in Fig. 1 of Faber et al. (2013)). There are other published cigarette smoke OA spectra showing lower f44 than Faber et al. (2013) (e.g. Fig. 4 in

Northway et al. (2007), DOI: 10.1080/02786820701496587 where m/z 44 is about 1/12th of m/z 43, or the sidestream smoke case of Fig. 4 in Tang et al. (2012), DOI:10.1080/02786826.2012.663949 where m/z 44 is about 1/5th of m/z 43). A paragraph was added to the text of Sect. 3.2.1 and 3.2.2 discussing the discrepancies and a possible underestimation of f44. However, a possibly slightly incorrect apportionment of f44 does not significantly alter the results (absolute and relative concentrations) and does not change the conclusions of the section. If one assumes that the "real" f44 was about 3.5% as in the Faber et al spectrum, this would change the total contributions of POAloc to the total OA by less than 1%.

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?

No, unfortunately we never tried that because at the time of measurement we were not expecting to find cigarette smoke OA to such an extent at the JFJ. The prototype ToF-ACSM used in that campaign however does not exist anymore in the same state. In the meantime many components were upgraded and exchanged and the vaporiser was exposed to different aerosols potentially affecting the additional f44. It has to be noted that also the mode of operation has changed with faster valve switching cycles more closely resembling the AMS chopper timescales in an effort to diminish discrepancies in observed f44 between the different instruments.

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.

We are very much convinced that the POA is real because of the above mentioned points. The spikes are very effectively separated from the rest of the data. We agree that the MS profile may not accurately represent the MS of the source (in particular for CO_2^+) as a consequence of the limited number of spikes available to extract the profile. But the effect on absolute and relative factor concentrations is small and within the expected errors (see Fröhlich et al. (2015)). Furthermore we believe that also considering the newly added Figures in the Supplement (Fig. 3 above showing the 370nm Aethalometer signal) we presented convincing evidence for our interpretation that the POAloc is connected with cigarette smoke.

Thus, I am not convinced with Fig10 (although the idea is excellent) and its interpretation.

The results of Fig. 10 would not be affected significantly by an inappropriate f44 in the POAloc factor because the f44 contribution of that factor to the total f44 would remain small in any case. If one assumes that the correct f44 was similar to cigarette smoke OA of Faber et al. (2013) then the f44 that came from POAloc was about 3-4% (f44 in CSOA) times 7-12% (POAloc in OA) = 0.2-0.5% of total OA while the f44 contributed by OOA is 30% (f44 in OOA) times 71-88% (OOA in OA) = 21-26% of total OA. The relative change of the vertical position of the (f44) in Fig. 10 therefore would be on the order of 1-2% (i.e. about 5 times smaller than the width of a marker in Fig. 10a) with even smaller additional variation between seasons since (not considering f44 in HOA and BBOA which are still included in Fig. 10, which would reduce the effect even more). Thus we conclude that also in the case of a possible underestimation of f44 in POAloc the overall picture of Fig. 10 would remain within the order of the shown accuracy and the interpretation remains valid.

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).

With f60 Fig. 7 shows the relative contribution of m/z 60 to the total OA in the different seasons. Because in winter the concentrations are very low (median about 0.1 µg/m^3), the influence of the noise is much bigger, i.e., the wider distribution is caused by the noise. All fractions show a wider distribution in winter although the real amplitude of the temporal variations is typically bigger in summer due to the frequent injections of PBL air with different OA mixtures. A note was added to the figure caption explaining the width of the distribution.

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.

Yes, in winter the time series are much more affected by noise (also visible for all the other factors) because of the very low concentrations. If the time series is averaged to a longer time step (see below for 3h) then clear temporal variations and significant non-zero values are better visible. BBOA constitutes 7% of total OA in winter which is above the 5% of the guidelines from Ulbrich et al. (2009)
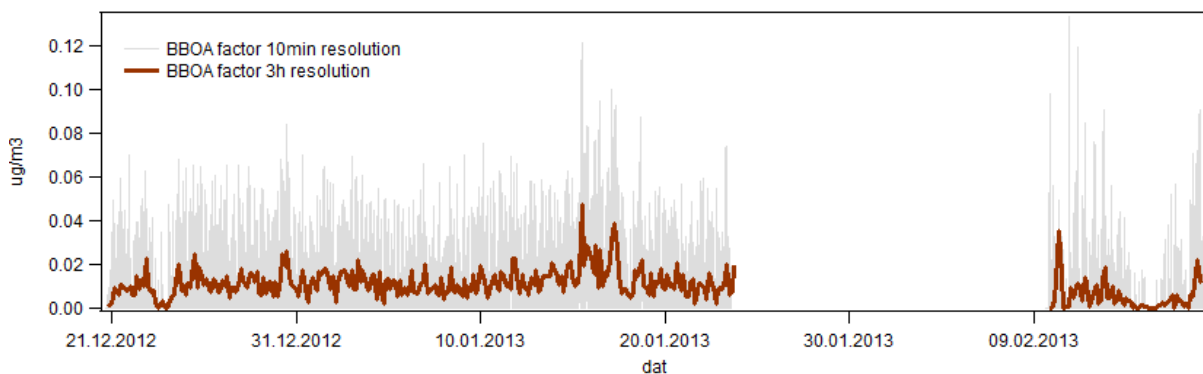

Figure 4: Fig. 4. BBOA factor of the ME2 analyisis at 3h time resolution.

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?

We do not think that these plots display contradictory information. In the whole summer we see three events with elevated BBOA concentrations (27.06, 05.07 & 06.07) which we attribute to air masses advected from the valley carrying aerosol from agricultural waste burning which does happen occasionally in summer. However, these events only cover a very short period of the total summer, meaning elevated f60 values would not show up in Fig. 7 whose whiskers are showing the 10th to 90th percentile only. Additionally, in the same air masses also a lot of other OA not carrying molecules that show up at m/z 60 was measured (LVOOA I: 4ug/m3, LVOOA II: 1.5ug/m3, HOA: 0.25ug/m3 vs BBOA only 0.25ug/m3,). This overlies the amount of m/z 60 in the BBOA when the fractions are calculated from the total OA.

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.

Revised. New text: "Many environmental topics which are becoming increasingly important in the future are closely linked to aerosols…"

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

Revised. Text changed to: "…with time resolutions on the order of minutes to hours on a long-term basis…"

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

We removed that statement since we are unable to provide an exact number here and in theory there are many locations (almost every mountaintop) that would allow the probing of FT air but only at a few of them the facilities exist. If one queries the data base of the Global Atmosphere Watch (http://gaw.empa.ch/gawsis/find.asp) only 16 operating observatories above 3000m are listed worldwide. In Europe the list is limited to only three: Jungfraujoch (CH), Sonnblick (A) and Plateau Rosa (I).

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

The results in this paper are in good agreement with previous measurements of chemical composition making the findings representative. A note about representativity and a reference to the review article of Bukowiecki et al. (2015) which compiles aerosol measurement results at the JFJ of the last 20 years was added to the text.

P18229, l25: please clearly state how seasons were differentiated

Revised. See also P18237 L3 and Fig. 1b.

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.

Revised. Calibrated RIE values were 3.2+/- 0.4 for NH4 and 0.6 +/- 0.1 for SO4. More detail about IE can be found in the answers to referee #3.

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

Revised.

P18231, l20: a good correlation (r or r2) will still be obtained with other CE values. Only the slope of the correlation will change. Please be more specific

Revised. This was caused by a language misconception. The word correlation should only refer to "plotted against each other".

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

Revised.

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

Revised.

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?

The Aethalometer data used in this study was treated according to the recommendations of the World Data Centre for Aerosols: "For all instruments manufactured by Magee, please use the corresponding level 0 template only for all types of data reporting, since there are no accepted Standard Operating Procedures for producing higher data levels available for these instruments." (http://www.gaw-wdca.org/SubmitData/RegularAnnualDataReporting/FilterAbsorptionPhotometerregular.aspx). Furthermore, for very low attenuation values the loading correction does not always improve the results (see Weingartner et al., 2003). At the JFJ the attenuation remains low for a considerable time after a filter change even if the highest possible flow rate is applied. Thus the application of the loading correction is subject to bias in this case. An estimation of the site specific uncertainties of MACabs (used to calculate eBC from the raw attenuations) is still under investigation and a publication is in preparation by Zanatta et al. (2015). The text was changed accordingly.

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?

Yes, this value was determined empirically and it agrees with the value found by Herrmann et al. (2015) in a long-term (6 years) investigation of the aerosol size distributions at the JFJ.

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)?

In this subsection of the methods description the three methods are merely described. Hence, the take home message is that several approaches exist and were applied to the dataset. The results are then discussed in detail in Sect. 3.1.3 (Basically all three methods deliver similar results for the median FT aerosol composition and concentration). The more detailed exploration was not performed because it was deemed to be out of the scope of this publication. On the one hand Herrmann et al. (2015) published this in a separate paper and on the other hand the back dispersion raw data necessary for a deeper investigation was not available to us at the time when the work was performed.

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

Due to the measurement interruptions visible in Fig. 1b no continuous data was collected. The regions in Fig. 1b show the periods that were used to calculate seasonal concentrations etc. Gaps do

not mean that data was omitted, just that no data was available before and after. The figure caption was revised to avoid misunderstandings

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

Revised. Here and elsewhere.

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

Revised.

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

See table S2 showing the variability of the averaged results for all factors due to the applied a value range. Table S2 contained an incorrect table caption which was revised.

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?

Yes, absolute eBC concentrations are similar (see Table 1) but relative eBC contributions to the total aerosol are higher in summer 2013 (4% vs 6%, see Fig. 2 or sum in Table 1), i.e. the ratio OA to BC is different. The HOA to BC ratio of on-site diesel generators potentially differs for the HOA to BC ratio of aged fossil fuel combustion emissions transported to the JFJ from the surrounding valleys. The mentioned construction activities were taking place over a longer period and no significant BC spikes can be seen in the eBC data. But since this cannot be excluded as a potential contamination it is mentioned here together with a second possibility. An underestimation of HOA in the source apportionment would be a third possibility and was added to the paper.

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)

See replies and BBOA time series in major concern section. The missing BBOA winter diurnal variation plot was added to the supplement.

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.

The POAloc was subtracted, i.e. the caption is correct. The text in section 3.2 was changed accordingly. For comments about the concerns dealing with POAloc and the OA fractions see above.

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

Revised. A wrong table caption was displayed for Table S2.

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

Revised.

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

Revised.

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

Revised.

P18228 ,l17: please remove one "and"

Revised.

P18228, l17: please insert a "–" in "gasphase"

Revised. Here and elsewhere.

P18238, l15: please remove one "which"

Revised.

P18238, l22: "In autumn, winter and spring"

Revised.

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