Response to reviewer no. 1

We would like to thank the reviewers for their comments and suggestions which greatly improved the scientific and technical quality of this manuscript. My response to each of the reviewers comments are below.

Reviewers comments (RC1) are in bold text.

Authors comments (AC) are in normal text. Please refer to the attached document to view all changes to figures.

Specific comments

Abstract

RC1.1: Page 27140, Line 5: there are different types of Time-of-Flight aerosol mass spectrometers (HR-ToF-AMS, C-ToF-AMS, CC-ToF-AMS). Please specify what kind of AMS you used in this study.

AC: Updated text.

Page 27142 Line 11: Inserted "compact" and changed ToF-AMS to cToF-AMS

Page 27144 Line 21: changed to cToF-AMS.

RC1.2: Page 27145, Lines 16-19: It would be nice to see how the composition dependent CE changes with time in your study. I suggest adding this CE vs time in the supplements.

AC: This figure is included, Figure S2

RC1.3: Page 27145, Line 21 to 29: It was mentioned in the text that the total volume from the AMS and MAAP was compared to that from the SMPS. What value of density did you use for BC?

AC: We used a value of 1.8 g cm^{-3} for the density of BC. The text is updated.

Previous text

Page 27145, Line 26: (1.72 g cm^{-3} for SO_4, NH_4, NO_3, and 1.2 g cm^{-3} for organics)....

Updated text:

.....(1.72 g cm^{-3} for SO_4, NH_4, NO_3, 1.2 g cm^{-3} for organics, and 1.8 g cm^{-3} for BC)

2.4 Positive matrix factorisation:

RC1.4: I suggest adding more discussion about the selection of fpeak and the number of PMF factors. It would be useful to add some validation plots to the supplements, e.g. fpeak vs Q/Qexpected, PMF solution with 3 factors (MS and time series), at least for some campaigns, so that the reader can see why you ended up with a 2F-solution with fpeak of 0.

AC: Additional figures and text have been included.

Updated text:

Page 27147: Line 3: A fpeak value at which a local minimum in the solution space (Q/Qexp) was achieved is used to represent each database (Fig. S5).

Page 27149: Line 8: The number of factors chosen from the PMF analysis was based on the temporal evolution of each of the organic species as well as changes in their mass spectral properties. Two different types of organic compounds were chosen to adequately represent the organic particle fraction at the pdD site. A PMF solution for three factors is shown in Fig. S7 and S8. However, we believe that there is no significant change in the temporal evolution and in the mass spectral properties of the each organic species when going from a two to a three factor solution, and therefore chose the two factor PMF solution.

3.0 Results and discussion:

RC1.5: Page 27148, line 1-2: Regarding the contribution of BC to submicron particle mass how would you estimate the uncertainty of BC measurements for submicron particles (<1 um) since MAAP sampled behind WAI inlet (with a cut-off of 30 um)?

AC: BC measurements have been performed at the pdD site over several years and demonstrated that nearly all of the BC mass is measured in the submicron mass (Sellegri et al., 2003).

The text has been updated to provide additional information.

Updated text:

Although BC species are sampled behind an inlet with a cut-off diameter of 30 μm, it has been demonstrated that nearly all of the BC mass measured at the pdD site have diameters less than 1 μm (Sellegri et al., 2003). It is therefore assumed to contribute only to the submicron mass.

RC1.6: Page 27148, lines 23 to 24: I suggest adding neutralisation plots to the supplements.

AC: These plots are now included in the supplements as Figure S4

RC1.7: Page 27149, Line 24: Why LV-OOA correlated with BC in summer? Could part of LV-OOA be related to traffic?

AC: Additional text is added to the PMF analysis section 3.2. See response to RC 1.9

RC1.8: Page 27151, Line 8 to 11: You mentioned that in autumn the correlation (r^2) between SV-OOA and BC was 0.33 but based on Table S1 it was only 0.12.

AC: This was an error. The table and text is updated.

Previous text:

Page 27151, Line 11: ....with the time-series of BC (r^2=0.33).

Updated text:

...with the time-series of BC (r^2=0.12)

RC1.9: Page 27149 to 27152, PMF in general. I suggest adding more discussion about the sources and characteristics of LV-OOA and SV-OOA based on Table S1, e.g. LV-OOA correlates better with inorganics in all seasons, chloride correlates very well with LV-OOA in winter (does that mean biomass burning?). How about correlations with the reference mass spectra? Earlier you said that the resolved PMF factors were similar during all the seasons. If that is true why do the correlations with the reference MS change that much from season to season (Table S1)? Could you describe shortly how MS for LV-OOA and SV-OOA varied from campaign to campaign?

AC: Several additions are made to the text.

Added text:

Page 27150: Line 27. During the autumn and winter the LV-OOA component showed good correlations ($r^2 > 0.50$) with both NO_3 and NH_4.Correlations between the organic and the inorganic species during the winter suggest that these species have similar sources. However, during the winter boundary layer dynamics are likely influencing the time series of all compounds. Unlike all other seasons the LV-OOA showed a strong correlation with Chl in the winter (>0.70), suggesting that the aerosol is related to biomass burning emission sources (deCarlo et al., 2008). This is confirmed by the similarities of the LV-OOA mass spectra with the reference mass spectra of biomass burning organic aerosol (BBOA), which is due to contributions from m/z 60 and m/z 73, characteristic marker peaks for levoglucason. The weaker correlation between LV-OOA and SO_4 during the autumn experiment suggests that some of the SO_4 particles may have different sources to that of the organic species. Weak correlations between the organic and inorganic species throughout the summer experiment are a result of the very high mass concentration of the organic species suggesting different sources of the organics to the inorganic species. However, in addition to high organic mass concentration the lack of correlation between the organic and nitrate species is also a result of nitrate chemistry, with nitrate mass concentration decreasing during the day time hours, and the photochemical formation of organic species. Correlations between LV-OOA and BC (r^2=0.44) suggest that some of the LV-OOA may be related to anthropogenic sources.

Page 27151, Line 8.The time series of this SV-OOA correlates with the time series of NO_3, BC, and gas phase species CO_2 (r^2=0.27) and NO_2 (r^2=0.48) suggesting that particles arrived from an anthropogenic source.

Page 27152, Line 16..... This is in agreement with our previous discussions where the LV-OOA time series measured during the winter is strongly correlated with that of Chloride (r^2=78%) and has similar mass spectra to reference BBOA mass spectra. For the summer campaign, the high correlations of the measured SV-OOA with the mass spectra of secondary organic aerosol formed from alpha-pinene is in agreement with the hypothesis that the organic mass spectra situated on the right hand side of the graph are likely related to the biogenic precursor sources. However, given its correlation with the time series of BC, the LV-OOA component measured during the summer is likely to also have an anthropogenic source.

Technical corrections

RC1.10. Title: I suggest adding France in title.

AC: The title has been updated from

"Seasonal contributions of aerosol chemical composition at the puy de Dome research station"

To

"Seasonal contributions of aerosol chemical composition at the puy de Dome research station in France"

RC1.11: Results and discussion Page 27147, line 10: Parentheses are missing for BC.

AC: This is corrected.

Page 27147, Line 10: Insert sentence.

Tables and Figures

RC1.12: Table 1. Page 27162: Check the labels for the columns and units. They are mixed now.

AC: This has been corrected.

RC1.13: Fig 3. Page 27166: (a), (b), and (c) are missing in Fig.3.

AC: (a), (b), and (c) are now inserted into Fig.3

RC1.14: Fig 5. Page 27169: (a) to (f) are missing in Fig.6.

AC: (a) to (f) are now inserted into Fig.6

Supplements

RC1.15: Fig. S2: Comparison for summer is missing. There are (a) and b) in Figure but (c) and (b) in Figure caption. In Figure caption add...measured by the AMS and MAAP.

AC: The comparison with summer is included and the figure caption is updated.

RC1.16: Figure S5: (a) to (d) are missing in Figure.

AC: (a) to (d) are now included.

RC1.17: Table S1: Change Table 1 to Table S1.

AC: The text is updated.