

Dear Authors:

After careful consideration of your revised manuscript, I think that several of the points brought up by the referees must be addressed more thoroughly prior to publication. Please consider my comments below. Thank you.

Eleanor Browne

Major comments:

Referee 2 expressed concerns regarding the removal of highly polluted periods prior to PMF analysis. I agree that this needs to be addressed in greater detail in the manuscript – it appears to me that nothing has been changed. Additionally, I do not understand the authors' reply to this comment. Mainly, I do not understand what is meant by "ion fitting in this period is bad." With high signals, it is typically easier to do ion fitting. Do the authors mean that new ions appear? If so, those should be fitted and addressed. The discussion on PMF results with and without this time period should at least be included in the supplementary material.

Referee 2 also expressed concerns regarding the conclusions drawn from the ¹⁴C analysis and the representativeness of the samples. I think that the authors should reconsider their interpretation of these results. In particular, I am hesitant to agree with the statement that the "...air mass and aerosol source are pretty stable..." when applied to the ¹⁴C measurements. One of the ¹⁴C measurements (January 23) was performed on a day that was removed from the data analysis due to "highly polluted" conditions, and another (January 3) was during a period before the start of the AMS measurements. Given that 50% of the filters are from time periods where the AMS data is not even considered in the manuscript, how much can we really learn from this analysis and is this analysis even appropriate?

The comment from Referee 2 regarding the CCOA factor also needs to be addressed further in the manuscript. Given the similar time trends of CCOA and BBOA, how certain is it that *m/z* 91 and the PAH-related ions are related to CCOA and not BBOA, especially given that the BBOA spectrum is somewhat constrained? Please include discussion addressing these points in the manuscript.

Specific Comments

Line 120: "...applying thousands of individual species..." Given that vaporization and ionization in the AMS result in extensive fragmentation, the AMS does not really measure "individual species." I recommend revising since this is somewhat misleading.

Line 131-132: I think a few more references would be appropriate. Particularly some that represent the more recent advancements in models.

Line 135: "...appear to be advance..." This does not make sense. Please revise.

Line 156-157: The aerosols are also influenced by very different meteorological processes between the two seasons.

Line 219: "ionic path" This is non-standard. Please consider more standard wording.

Line 231: If a background was determined only once in the study, how was the gas-phase CO₂ correction determined for other times during the campaign? Using a constant value is likely not appropriate, and given the emphasis put on CO₂⁺, this needs to be explained.

Sect. 2.2.1: How was the size measurement of the AMS calibrated? Significant time is spent discussing diameter later in the manuscript so this must be addressed.

Line 248-249: "...refractory mass of the particle quantified by detection of the main light-absorbing component is rBC." This is unclear and should be rephrased.

Line 283: What density was assumed in making the comparison with the SMPS? Is a slope of 1.48 really good? Or was no density applied?

Line 335: "pretty stable" is not very meaningful and should be quantified better. Please see general comments for other concerns regarding this section.

Line 344: Where does the factor of 1.03 come from?

Line 363: Should f_{NF_BBOC} be less than 1? That is my understanding from the wording. Please clarify.

Line 376: Some of the average values (particularly temperature) are not very convincing in support of the statement of stable conditions. I assume this is due to diurnal variation. Perhaps it would be better to present average lows and highs.

Lines 394-396: Do you mean to say that you have measured a lower limit?

Sect 3.1.4: The size distribution of chloride seems more similar to the size distribution of the organics than to the other inorganics. What are the implications of this and is it consistent with your factor analysis?

Line 460-462: I find this highly speculative since the oxidizing capacity is greatly reduced in the winter. Is the increase in SO₂ really enough to account for changes in sulfate given lower oxidizing conditions?

Line 479: NO₂ is not formed from the reaction of NO with OH. Please correct.

Line 486-489: These comments regarding the inorganic species can be constrained (at least somewhat) with the appropriate analysis. I recommend either performing this analysis, or removing some of this from the paper as the major point of the paper is the analysis of the organic aerosol.

Line 533: If the HOA and BC are thought to be mainly from the same source than why is the HOA peak in the evening small than the morning while the BC ones are about equal?

Lines 597-598: There are so many points in Fig. S10 that I cannot clearly see at all any time of day dependence. Please consider some sort of data reduction to make this clear. For Fig. S10, where do the COA and HOA lines come from and why does HOA fall on the COA line in panel B? It would also be helpful to see the plot for the C_xH_y⁺ ions at *m/z* 55 and 57 as well.

Line 638: The CCOA is not "high and left." This was pointed about by Referee 2 as well. Please fix.

Line 660: Please correct the spelling of Jimenez.

Line 673-674: "These results indicate that the atmospheric oxidation capacity during the winter was still very strong." This statement is very vague (what precisely is meant by "very strong"?) and I think somewhat strong given that there is no exploration of the oxidative budget. Please reconsider.

Line 716-717: It is not at all clear to me that a correlation exists for POA less than 15 $\mu\text{g}/\text{m}^3$. At 15 $\mu\text{g}/\text{m}^3$ SOA varies between ~ 4 and 12 $\mu\text{g}/\text{m}^3$!

Lines 798-803: This part still seems rather speculative (as pointed out by Referee 2). I do not know what we are really learning from this discussion/comparison since there is no analysis of gas-phase organics in the present manuscript.

Lines 824-826: I encourage the authors to remember that the boundary layer explanation is still rather speculative since simultaneous measurements of the boundary layer or vertical structure do not exist. Even in Fig. S8 only early morning and evening temperature profiles are shown and no data is given for noon.

Table 2 & multiple places in the text): Please be consistent and use either r or r^2 .

Fig 1: I find this very difficult to read due to the small size and the large amount of information. Please consider making more figures. It would also be useful to see the wind direction as a wind rose.

Fig 5: How is there 50 ppbv of NO at night and still 10 ppbv of O₃? I would have thought that O₃ would have been titrated away.

Fig. 7: Please use the standard AMS colors for the ion families. Please also identify them as ions by including "+" in each name. Also, the y-axis labels are overlapping and difficult to read.

Fig. 8: I am unsure how useful this figure is given that most (if not all) of this data already appears in Fig. 4.

Fig. 9: What is the R² value representing? A value of 0.46 seems unrealistically high for any sort of line in panel A. Is OOA supposed to be SOA?

Fig 13: Would a van Krevelen plot be more meaningful? Also, please update LV-OOA and SV-OOA to their correct names. Also Fig. 13 is referenced in the text before figures 9-12 are referenced. Please reorder the figures to be referenced in order.