

Responses to Interactive comment on “New characteristics of submicron aerosols and factor analysis of combined organic and inorganic aerosol mass spectra during winter in Beijing” by Referee #2

This paper presents AMS measurements in Beijing during January 2014 and compares the results obtained with a previous study from the same group during January 2013. Results are analyzed using positive matrix factorization, where both organic and inorganic species are included. The results are very similar to those obtained in previous studies at the same location. The authors claim that the “novel” approach used including both organic and inorganic species enhances the resolving power of PMF. However, not only I do not agree with their claim about the novelty of the work, but I also strongly believe that such conclusions cannot be drawn, without comparing the results obtained to those from a more traditional PMF where only organic fragments are included, such conclusions. This is a great limitation of the paper. In general, the authors did a poor job in validating their PMF results, especially in the lack of external data supporting their interpretations. Note that the study is limited to AMS measurements only. Despite such limitations, the authors make several bold statements about the chemical mechanisms via which secondary species are formed, sources’ geographical origins and abatement strategies, which are not supported by their data and that are only based on speculations. Some technical issues regarding AMS data analysis can also be pointed out (e.g. CE determination, RIE determination). The authors mainly cite their own work and omit some important citations from other groups (e.g. from pku university, Chinese Academy of Sciences: : :). In my opinion, this is not transparent. Accordingly, I do not support the publication of this work under its current form.

We are thankful to the referee #2 for his or her comments and suggestions, which contribute to improve the quality of our paper. We have revised the manuscript accordingly. Listed below are our point-by-point responses in blue to reviewer’s comments. To make a more convenient reply, we named every comment using the red and bold Arabia number (such as **1**, **2**, **3**.....).

Specific comments –

1. main text: 0/ title: I do not see the new characteristics. The results obtained are very similar to those presented in previous studies, at the same location.

Reply: We are very grateful for this important comment.

(1) We also thought that the title of this manuscript was inappropriate. Therefore, the title has been changed to “Variation in submicron aerosols during winter in adjacent years in Beijing”

(2) Although the title has been changed in the revised manuscript, there are several novel and interesting results reported.

(a) In section 3.1, we found that the NR-PM₁ mass compositions were very different in January 2013 and 2014 and the NR-PM₁ mass concentration also decreased significantly (40 μg m⁻³). In January 2013, the contribution of sulfate was much higher than nitrate. However, the contributions of sulfate and nitrate were decreased by 5% and increased by 6% in January 2014, respectively. It is necessary to discuss the reasons for these changes. There have been

many emission reduction measures applied in Beijing and the surrounding area in recent years. Therefore, we needed to know the reasons for such variations: was it due to these measures, or differences in meteorological factors in the two Januarys? We then compared the source apportionment results in section 3.2 and carried out an in-depth analysis on meteorological factors in Section 3.4.

(b) Although the PMF was first used to analyze the high-resolution mass spectra (HRMS) of organic and inorganic aerosols (PMF_{AMS}) from AMS measurements by Sun et al. (2012), and the results are very valuable, this method has rarely been used in other areas, especially in China. Therefore, we thought it very necessary to use this method in Beijing, due to the better analysis results that could be achieved. As we know, traditional PMF only focuses on OA HRMS (PMF_{OA}) and analysis of the relationship between OA factors and other species (sulfate, nitrate etc.) is limited only to correlation analysis of their time series. Therefore, PMF_{OA} could not reflect the relationship between organic and inorganic species. However, PMF_{AMS} could reflect the relationships between them directly. For example, the sulfate and chloride mostly appeared in the highest oxidation factor ($O/C = 0.89$) and the CCOA, respectively, after PMF_{AMS} analysis. This was much better than the comparison in the previous PMF_{OA} analysis. Meanwhile, the inorganic components that appeared in the MS of different factors showed that the different characteristics, such as formation processes or diurnal variations, which could be reflected through the PMF_{AMS} analysis. These interesting results have been added for a more thorough discussion in the revised manuscript (Section 3.2).

2. 1/ Page 18538, line 8: This is not the first time that inorganic and organic mass spectra are put together in PMF and I do not consider that this is a revolutionary advancement in the field to justify the publication of this study.

Reply: In introduction part, we introduced the new analytical methods: “some alternative and innovative approaches have recently been used that take advantage of PMF analyses including these species. For example, Sun et al. (2012b)..... McGuire et al. (2014)....” and “These novel methodologies are therefore of great interest for an improved understanding of pollution sources and their evolution in the atmosphere.” Therefore, it is extremely essential to apply these new methods to other studies. However, there has been no work of this kind conducted in Beijing, despite the serious fine particle pollution and frequent heavy pollution events in this city. The detailed reply can be found in the reply for the comment 1. Therefore, we thought that the new method used in Beijing firstly and the new and interesting results found could be considered as the innovation points of a study.

3. 2/ Page 18538, line 19: I do not see any solid result to support such a bold statement!

Reply: We are very sorry for the arbitrary expression. This sentence has been deleted now.

4. 3/ Page 18539, line 10; “air quality index > 200”: Either define or remove.

Reply: According the suggestion of the reviewer, “air quality index > 200” was deleted. This is because a detailed explanation on this index needs to involve an introduction of other pollutants (such as SO_2 , O_3 , NO_2 , PM_{10} and CO) and complex calculation process. This content will occupy a certain space. Therefore, we thought it is not necessary.

5. 4/ Page 18539, line 22; “After combining the organic mass fractions observed by the AMS”: Not clear.

Reply: We are very sorry for the unclear expression. The first two sentences of this paragraph were changed as “Positive matrix factorization (PMF) (Paatero and Tapper, 1994), a bilinear model that constrains the factors to be non-negative, is widely used for organic aerosol (OA) mass spectra analysis (Ulbrich et al., 2009; Zhang et al., 2011). Various OA components such as hydrocarbon-like OA (HOA), biomass burning OA (BBOA), cooking-related OA (COA), coal combustion OA (CCOA), and oxygenated organic aerosol (OOA), are broadly identified depending on the site, season, and source emissions.” (line 80-86).

6. 5/ Page 18540, line 1-2: The sentence does not make sense.

Reply: We are very sorry for the unclear expression. This sentence has been changed as “These OA factors, together with measurements of hygroscopicity, volatility, and oxidation states, significantly improve our understanding of the sources and evolution processes of OA in the atmosphere (Jimenez et al., 2009).” (line 89-91).

7. 6/ Page 18540, line 5: What is the intrinsic relation between organic and inorganic species? They may have common sources, but their precursors, formation rates, partitioning properties and sinks are completely different.

Reply: We are very sorry for the arbitrary expression. This sentence has been changed as “Therefore, a PMF analysis focused on OA spectra alone may neglect the links between them. For example, some organic and inorganic substances may be emitted from the same source, or generated through a similar chemical process.” (line 93-95). In addition, although our study cannot achieve a more in-depth study of the relationship between organic and inorganic species in the atmosphere, we should not neglect it. Because a book which mainly focused on the SOA published in 2015 has found that some inorganic species (such as $(\text{NH}_4)_2\text{SO}_4$, ZnSO_4 , MnSO_4) may promote the formation of some SOA species whereas some inorganic species (such as FeSO_4) may hinder the formation of some SOA species.

Hao, J. M., Lu, Z. F., Chu, B. W., Wu, S., Zhao, Z.: Characterization, experimental study, and modeling of atmospheric secondary organic aerosol. Science Press, Beijing, 2015.

8. 7/ Page 18540, line 17: This is inaccurate; Crippa et al., could resolve the MOA, with-out the addition of the inorganic fraction. They include the latter to enable the SO₄ apportionment between marine and continental sources.

Reply: We are very grateful for this important comment. The inappropriate reference has been deleted now.

9. 8/ Page 18540, line 22: Again here I do not see the novelty of the approach used and the science presented. You mention that no work before you had applied PMF to OA and inorganic aerosol mass spectra : I am sure that this is only a slip and you are not aware of the work of Huang et al., 2014.

Reply: Please see the reply to the comment 1. In addition, we are very grateful for the reviewer for providing reference. However, it is really difficulty to query the work of Huang

et al., 2014. Although we check all query results in “web of science” (the query key words were “Huang, 2014, AMS” and “Huang, 2014, aerosol”). In addition, we also check all query results (461 items) in “Google Scholar” (the query key words we used were “Huang, 2014, AMS, aerosol”). We have not found the similar study. Therefore, if it is possible, could you please give other query information, such as the publication or the title of the paper? Then we can further revise our manuscript.

10. 9/ Page 18542, line 4: DeCarlo et al. (2006) is not a review.

Reply: This sentence has been changed as “Detailed descriptions of the Aerodyne AMS have been presented in DeCarlo et al. (2006) and Canagaratna et al. (2007).” (line 155-156).

11. 10/ Page 18542, line 8: 22.5s is very long, what was the reason for that choice? (it only worsen your data quality).

Reply: We are very grateful for this important comment.

(1) The running time of every mode was set up by the engineer of Aerodyne research Inc. He thought this running time is appropriate for our AMS.

(2) Generally, the observation results are reported in the form of hourly mean value. Therefore, we think the effect of this running time is not significant.

(3) In the revised manuscript, we compared the AMS results with other three instruments (Metone BAM-1020, TEOM, and SMPS) (line 238-249). They showed strong correlation and the correlation coefficients and slopes are consistent with many previous comparisons (Huang et al., 2010; Huang et al., 2013; Huang et al., 2012; He et al., 2011; Sun et al., 2013a). Therefore, we believe our AMS running time setting will not affect the authenticity of the observation results.

12. 11/ Page 18542, line 12: Did you determine the NH₄ and SO₄ RIEs? (Apparently not, see below).

Reply: Thank you very much for the useful suggestion. In the previous observation, we used the default value of the RIE value of sulfate, as in most previous studies, e.g., the studies in Paris (Crippa et al., 2013), the Pearl River Delta region of China (He et al., 2011; Huang et al., 2011), Changdao island (Hu et al., 2013), Beijing (Huang et al., 2010; Chen et al., 2015; Sun et al., 2015), Shanghai (Huang et al., 2012), the western Mediterranean (Minguillón et al., 2015), New York City (Sun et al., 2011), the Yangtze River delta region of China (Zhang et al., 2015), and so on. According to the suggestion of the reviewer, we determined the RIE for sulfate during the AMS calibration work several days ago. We found that the RIE for sulfate (1.21) is very close to the default value. Therefore, we believe the default value is appropriate in this study.

In addition, in order to prove the rationality of our result, we:

(1) Compared with several pervious similar studies. Sun et al. (2015) was conducted simultaneous real-time measurements of aerosol composition at two different heights at the same location in urban Beijing from 19 December 2013 to 2 January 2014. This research period partially overlapped with our study period. In their study, they also used the default value of 1.2 for the sulfate RIE, and the aerosol acidity (slope=0.65) was very close to our results (slope=0.68). Meanwhile, Chen et al. (2015) and Xu et al. (2015b) also used the

default value of 1.2 for the sulfate and found that the $\text{NH}_4^+_{\text{measured}}/\text{NH}_4^+_{\text{predicted}} = 0.56$ and 0.75.

(2) In the revised manuscript, we compare the AMS results with the PM_{10} mass concentration monitored using a beta-attenuation method (Metone BAM-1020, USA), the $\text{PM}_{2.5}$ mass concentration was monitored using Tapered Element Oscillating Microbalance (TEOM series1400a, Thermo Scientific), and the particle volume variation was measured with a collocated Scanning Mobility Particle Sizer (SMPS, TSI Inc.). Comparisons revealed that the results were very close to those reported in several previous studies, further proving the correctness of our observation results (line 238-249).

13. 12/ Page 18543, line 3: Having a constant bias in Figure S3 - i.e. a constantly acidic aerosol - is seriously iffy: To make any conclusion about the acidity of your aerosol you have to determine the RIE of sulfate. 1.2 is a generic number, but we have observed RIEs as low as 0.8 or as high as 1.5. In addition, a key assumption here is that Cl is bound to NH_4 ; there is no way this can be proven... This nonsensical parametrization is in my opinion complicating your mass determination, as it won't matter whether you use a $\text{CE}=0.45$ or a $\text{CE}=1-0.73*(\text{NH}_4/\text{NH}_4, \text{ predicted})=1-0.73*0.68=0.5$. An indication that your total mass determination (or CE) is not completely off is the comparison between the AMS and Metone results. Now, there could be compensating errors, as for example the use of a wrong SO_4 RIE or a high contribution of refractory species (e.g. black carbon) or particle bound water not measured by the AMS; therefore you should be careful when providing quantitative numbers. In this regards, where there any black carbon/elemental carbon measurements? If yes, why did the authors omit any discussion about it? You know there are instruments other than the AMS. Also, did you have any SMPS measurements? This would help constraining the CE.

Reply: Thank you very much for the useful suggestion.

(1) The reply for the RIE of sulfate has been showed in the reply for comment 12. We read a lot of previous study results, which observed the aerosol based on the AMS in many sites. Meanwhile, we focused on the comparison with some study results in Beijing in similar study period. Then we believe our AMS data is rationality and could reflect the true situation of the atmospheric PM in Beijing.

(2) The $\text{NH}_4^+_{\text{measured}}/\text{NH}_4^+_{\text{predicted}} = 0.68$ is the global aerosol acidity of the whole study period. However, the aerosol acidity is changing with the composition of aerosol and sometimes higher (or lower) than 0.68. Therefore, we used the **variable CEs** which calculated using the equation $\text{CE}_{\text{dry}} = \max(0.45, 1.0 - 0.73 (\text{NH}_4/\text{NH}_4, \text{ predict}))$ in this study, but not used the constant CE 0.45 or $\text{CE} = 1 - 0.73 * 0.68 = 0.5$.

(3) It is really unfortunately that the black carbon was start sampling in June 2014. Therefore, there is no available black carbon data in our study. Meanwhile, there is no available EC data in January 2013, because it was start sampling in March 2013.

(4) As we replied above, we added the comparison of AMS results with TEOM and SMPS results in the revised manuscript. The detailed information can be found in line 238-249.

14. 13/ Page 18543, line 14-15: I am not convinced that this can be generalized.

Reply: Yes, the determination of PMF analysis range of m/z needs to be according to the actual situation of the AMS. In previous studies, the PMF focused on the $m/z < 100$ (Allan et

al., 2010; Cottrell et al., 2008; DeCarlo et al., 2010), $m/z < 120$ (Dall'Osto et al., 2013; Xu et al., 2015), or $m/z < 150$ (Huang et al., 2012; Huang et al., 2010). In our study, as the number of possible ions at each integer m/z increases with the m/z value, the W-mode resolution becomes insufficient to resolve multiplets in the $m/z > 120$ regime. Thus only smaller ions ($m/z < 120$) are fitted and used for PMF analysis in this study. Meanwhile, Sun et al. (2009) also thought this approach should have little effect on the final results and they also found most of the organic signals (~95%) appeared at $m/z < 100$. After analyzing the data in our measurement, we thought the smaller ions i.e., $m/z < 120$ are best suited for the analysis in this study.

15. 14/ Page 18545, line 1-3: This is very vague! Please explain. I would expect the mass determined with the AMS to be lower, for the same reasons I have mentioned above (a lens cutoff lower than 1 μm , the presence of particle bound water and refractory species).

Reply: We are very sorry for the vague expression. In order to further prove the reliability of our data, we also compared AMS results with TEOM ($\text{PM}_{2.5}$) and SMPS. Then we found the comparison results are very close some previous results (Huang et al., 2010; Huang et al., 2013; Huang et al., 2012; He et al., 2011; Sun et al., 2013a) (line 238-249).

16. 15/ Page 18545, line 4-7: "Figures 1d and e compare the composition of NR-PM1 between January 2013 and January 5 2014. The contribution of organics in January 2014 accounted for 54% of the total mass, which was slightly higher than in January 2013." Then you say "In contrast, the contribution of total inorganic species was reduced." Redundancy: : :

Reply: The redundant content "In contrast, the contribution of total inorganic species was reduced." has been deleted.

17. 16/ Page 18545, line 7: How subtle is that? Please specify.

Reply: We are sorry for the inappropriate expression. The "subtle" has been deleted now. Then the sentence is more concise and easy to be understood (line 252-254).

18. 17/ Page 18545, line 20: "This implies that there was a striking increase in the oxidation of OA in January 2014". This contradicts your previous statement in the same page, that secondary processes were more important during more extreme haze conditions in 2013.

Reply: Thank you very much for the important comment. Another reviewer also thought the discussion in this part is not very reasonable. Therefore, we deleted this part. However, this will not reduce the richness of the article. Because we added the PMF_{OA} results and discussed more in-depth, this makes the research focus more prominent.

19. 18/ Page 18545, line 22: compared to O_3 , OH is much more efficient.

Reply: We also thought a comparison with OH is more efficient. Unfortunately, there are no available OH data can be used in our study. Meanwhile, this part has been deleted now.

20. 19/ Page 18545, line 22: A similar variability between increased temperature and SOA production is only correlation and not causation. Aging does not occur through heat!

Reply: The unreasonable discussion has been deleted.

21. 20/ Page 18545, line 24: use the present tense: shows instead of showed. In general the English/the grammar need polishing.

Reply: Thank you very much for the suggestion. This manuscript has been polished by the professional polishing company.

22. 21/ Page 18545, line 26: You are considering changes in the composition, in particular O/C, and photochemical extent against supposedly independent variables (e.g. temperature as a surrogate of photochemical aging, or RH). However, these variables are not independent and I think that much of this is correlation but not causation. Therefore this analysis cannot be used to determine the mechanism by which OA ages. I give some examples. At higher temperature, I would expect less contribution from combustion sources and hence higher OOA contribution and O/C ratios. The analyses you are presenting do not take into account any change in primary emission contribution, but your conclusion is that at higher temperature aging is more important. A lot of the statements are not fully justified and need toning down. Haze events are often associated with higher RH, where secondary processes are more important (due to stagnation and higher cloud processing). That does not mean that cloud processing is a major contributor to SOA as you claimed. Are O/C ratios of SOA from aqueous phase chemistry and from homogeneous photo-chemistry the same? The argumentation you are presenting is based on such assumption. Can you validate this assumption?

Reply: We are very sorry for our inappropriate discussion and this part has been deleted now.

23. 22/ Page 18548, line 24: Is that normalized to OA in NO₃-OA factors (right) or to the total spectrum (wrong)?

Reply: We are very sorry for the unclear expression. This sentence has been changed now (line 328-331).

24. 23/ Page 18549: It would be useful to also normalize the contribution to OA, for a comparison with previous source apportionment data.

Reply: Thank you very much for the useful suggestion. We have added the PMF_{OA} results and compared with the results in January 2013 (section 3.2).

25. 24/ Page 18549, line 12: From where did you obtain the time dependence of the equilibrium constant? This is not shown here and not explained. K_p is dependent on temperature and RH, but the formation of NH₄NO₃ is also dependent on the available NH₄. In this regards, the formation of NH₄NO₃ is not at all favorable in acidic conditions with no gas-phase NH₄ available (maybe check Seinfeld and Pandis 2008), while this is what you are suggesting (recheck your RIE): : :

Reply: We are very grateful for this important comment.

(1) We cited the discussion reported by Sun et al. (2012), because the diurnal variation of NO₃-OA₁ in our study is close to the NO₃-OA in their study. However, we maintained a cautious attitude to the discussion on this part and revised the content in this part according to the comment (line 349-352).

(2) About the sulfate RIE used in our study, please see the reply for comment 12. Meanwhile, we have added the comparison of AMS result with TEOM and SMPS results in the revised manuscript. The detailed information can be found in line 238-249.

26. 25/ Page 18549: The interpretation of the nitrate factors is very speculative/dubious to say the least. You are suggesting that NO₃-OA1 formed via the N₂O₅ hydrolysis follows the expected partitioning behavior of nitric acid. Whether HNO₃ is formed by the heterogeneous hydrolysis of N₂O₅ or by the reaction of NO₂+OH, its partitioning into the particle phase is driven by temperature, RH and the availability of NH₄ (particle acidity). Further, I would not expect that the nitrate resulting from the 2 processes have distinguishable mass spectral profiles to be separated in PMF. Therefore, this separation is most-likely driven by the organic mass spectra. As you have mentioned Sun et al. (2014) have resolved similar factors without including the inorganic ions, but here it is not clear how did the inorganics help in resolving the factors. For the case of the nitrate factors, you may check the neutrality in these spectra, which would give you an indication whether some of the nitrate signal detected is not related to ammonium nitrate (may be organic nitrate or some other primary salts: NaNO₃ or KNO₃). You can also look at the ratio of NO/NO₂ compared to that from nebulized NH₄NO₃. I believe in general that the organic mass spectra helped the resolution of several inorganic containing profiles and not vice-versa. But, to convince me you would need to run an OA-only PMF.

Reply: We are very grateful for this important comment.

(1) PMF components were identified by their MS signatures and the correlation of their time series with tracers, and were then confirmed using additional information, such as diurnal cycles, and elemental composition. In addition, we also analyzed the MS signatures and time series of factors for factor numbers lower and higher than we chose.

We acknowledge that nitrate resulting from the two processes does not have distinguishable mass spectral profiles to be separated in PMF. This is why we combined the HRMS of inorganic aerosol with organics to perform the PMF_{AMS} analysis in this study. As we know, PMF_{OA} analysis can identify several factors that showed different characteristics. Therefore, when positive matrix factorization (PMF) is applied to the merged HRMS of organic and inorganic aerosols, the inorganic aerosols will appear in different factors and may show similar characteristics to the organics in the same factor mass spectrum, such as the diurnal variation and similar sources or formation processes. For example, we found the NO⁺/NO₂⁺ for NO₃-OA1 and NO₃-OA2 were totally different to the ratios for HOA and COA (line 324-328, 444-448).

In addition, the contribution proportion of ammonium nitrate in NO₃-OA1 and NO₃-OA2 reached 53% and 34%, respectively. Therefore, the difference in the characteristics between these two factors was not only determined by the organics. On the other hand, we assumed that the formation mechanisms of the nitrate in the two factors were the same. Then, we believe the diurnal variation will not be obviously different, like in our study.

(2) We are very sorry for the unclear expression. In fact, this comparison only focused on the diurnal variation of these factors. The sentences have been revised now (line 393-395). Although the two factors (SV-OOA and LSOA) in the study of Sun et al. (2014) are similar to the two NO₃-OA factors in our study, the PMF_{OA} could not reflect the relationship between

organic and inorganic species. However, the PMF_{AMS} could reflect it directly. For example, the sulfate and chloride mostly appeared in the highest oxidation factor ($O/C = 0.89$) and the CCOA after the PMF_{AMS} analysis, respectively. This is much better than the comparison in the previous PMF_{OA} analysis, i.e., which only compared the correlation of the time series of OA factors and other inorganic species. Meanwhile, the inorganic components that appeared in the MS of different factors showed different characteristics, such as formation processes or diurnal variations, could be reflected through the PMF_{AMS} analysis.

(3) The NO^+/NO_2^+ ratios for NO_3 -OA1 and NO_3 -OA2 were 2.69 and 2.21, which were close to the value of 2.8 for NH_4NO_3 determined in the AMS calibration. They were also in the range of 1.5 to 2.9, reported in some previous studies (Farmer et al., 2010; Fry et al., 2009; Bruns et al., 2010; Hao et al., 2014). However, the ratios for HOA and COA were much higher than that in the NO_3 -OA1 and NO_3 -OA2, with the values of 12.5 and 14.5, which corresponded to the ratio range of 10–15 measured for organic nitrate derived from reactions of monoterpene and the NO_3 radical in laboratory chamber experiments (Fry et al., 2009; Bruns et al., 2010; Hao et al., 2014). Thus, the nitrate distributed in these factors corresponded to organic nitrate. This discussion has been added into the revised manuscript (line 324-328, 444-448).

(4) In the revised manuscript, we have added the PMF_{OA} results and discussed them in greater depth (Section 3.2).

27. 26/ Page 18550: The discussion about aerosol acidity and nitrate formation is all biased by the presence of organic-nitrate, other nitrate salts from combustion, and the SO_4 RIE determination....

Reply: We are very grateful for this important comment.

(1) This aerosol acidity calculation method is only based on the NH_4 and three other anions (i.e., sulfate, nitrate and chloride). However, the effect of metal ions and organic acids on the aerosol acidity should not be neglected in the actual atmospheric environment. Therefore, the real aerosol acidity in the atmosphere is not as strong as the calculation results in this study. However, the components that can be observed by AMS is very limited (Org, NO_3 , SO_4 , NH_4 , and Chl) due to the principle of AMS. Therefore, the calculation results based on the AMS results should be mainly used for a preliminary understanding of aerosol and comparison with other AMS observation results. It is inappropriate to use them to evaluate the actual acidity of aerosol in the atmosphere.

(2) About the sulfate RIE used in our study, please see the reply for comment 12.

(3) As we replied above, we have added the comparison of AMS result with TEOM and SMPS results in the revised manuscript. The detailed information can be found in line 238-249.

(4) In addition, the strong aerosol acidity in Beijing were also reported by Sun et al. (2015) ($NH_4^+_{measured}/NH_4^+_{predicted} = 0.65$), Xu et al. (2015) ($NH_4^+_{measured}/NH_4^+_{predicted} = 0.75$), and Chen et al. (2015) ($NH_4^+_{measured}/NH_4^+_{predicted} = 0.56$ and 0.62). The study period was partially overlapped with our study.

28. 27/ Page 18551, line 13-17: If Sun et al. (2014) were able to distinguish the same factors than here using OA spectra only, why was that not possible in the current study. In general,

without a comparison between the OA-only PMF and all-species-PMF, the reader does not get how this inclusion affects the capability of PMF to resolve additional sources.

Reply: We are very grateful for this important comment.

(1) We are very sorry for the unclear expression. In fact, this comparison only focused on the diurnal variation of these factors. The sentences have been revised now (line 393-395). Although the two factors (SV-OOA and LSOA) in the study of Sun et al. (2014) are similar with the two NO_3 -OA factors in our study. But the PMF_{OA} could not reflect the relationship between organic and inorganic species. However, the PMF_{AMS} could reflect it directly as we mentioned above. Meanwhile, the inorganic components which appeared in the MS of different factors showed the different characteristics, such as formation process, diurnal variations, that can be reflected through the PMF_{AMS} analysis.

(2) According to the reviewer's suggestion, we have added the PMF_{OA} results in the revised manuscript. Then we compared them with the PMF_{AMS} results and the PMF_{OA} results in January 2013. The detailed information can be found in section 3.2.

29. 28/ Page 18553, line 21: I did not understand you expect that all primary emissions to have the same composition and the difference in their O/C ratio is only due to aging?

Reply: We are very sorry for the unclear discussion.

(1) The POA, such as HOA and COA, emitted from different sources and showed completely different features, such as diurnal variation and the time series. The MS signature is only one recognition feature to identify the OA factor. This is why the identify of OA factors need to take into account several factors, such as MS signature, elemental composition, diurnal variation, time series, the relationship with other species and the actual situation of observation site.

(2) O/C ratio is only one OA factor recognition feature, which has been considered as a good reference for the oxidation state and photochemical age of OA (Jimenez et al., 2009; Ng et al., 2010). However, it is not only determined by the aging process, but also affected by other factors, such as the mixing of OA from different sources and with different degrees of oxidation can lead to O/C changes as well. For example, a decrease of O/C can be observed simply because of a larger contribution from chemically reduced POA from vehicle emissions and cooking sources, which often happens at peak hours for traffic and meals.

30. 29/ Page 18554, line 1-3: That cannot be assessed because we do not see how OA only PMF performs.

Reply: We added the PMF_{OA} results and carried out the detailed comparison with the PMF_{AMS} and the PMF_{OA} results in January 2013. The detailed information can be found in section 3.2.

31. 30/ Page 18555, line 7-11: This is not as straightforward as presented here. Caution has to be taken; consult Chhabbra et al. (2011-ACP) and El Haddad et al. (2013-AE).

Reply: We are very grateful for the useful references.

These two references are very valuable for our study. The Chhabbra et al. (2011-ACP) investigated the variability of SOA formed in chamber experiments from twelve different precursors in both "triangle plot" and Van Krevelen domains. The El Haddad et al. (2013-AE) using a multiple regression analysis, absolute concentrations of functional groups (carboxylic

(RCOOH), carbonyl (RCOR'), and nitro (RNO₂) were combined with those of organic carbon derived from different sources in order to infer the functional group contents of different organic aerosol fractions. These fractions include fossil fuel combustion emissions, biomass burning emissions and secondary organic aerosol (SOA). Results clearly highlighted the differences between functional group fingerprints of primary and secondary OA fractions. Then we have a deep understanding of the characteristics of the OA factors at different aging stages and the changes of their characteristics with OA aging, etc. Some useful information has been cited in our study now. However, our study is a overall study on the aging of PMF factors as reported in Heald et al. (2010) and Sun et al. (2012). Therefore, we focused on the analysis of the whole trend of all PMF factor although we have combined the studies of Chhabra et al. (2011-ACP) and El Haddad et al. (2013-AE) (line 533-542).

32. 31/ Page 18558, line 10-20: What is considered as a regional scale? Is it 20-40km or greater than 100Km? Your diurnals seem to suggest that most of the sources are rather local or at least close; it depends on the wind speed, but for winter winds in china below 10Km h⁻¹, it needs at least 5h to transport primary emissions 50km away from the sampling site, e.g. in the Hebei region. This has to be clarified. Specific comments

Reply: (1) The fine PM pollution is regional, and has not the obvious boundaries. Up to now, there are many studies have found that the regional transport is an important contributor to the PM pollution in Beijing, especially the pollutants originated from southern Beijing (Jia et al., 2008; Zheng et al., 2014b; Sun et al., 2014; Wang et al., 2014b). During the heavy pollution event in winter 2013, the regional transport contributed 66% to PM₁ on average (Sun et al., 2014). The hybrid single-particle Lagrangian integrated trajectory (HYSPLIT) model is a useful air trajectory model, especially for studying the long-range transport of air masses. It has been used in many cities, including Beijing (Huang et al., 2010; Sun et al., 2010), Shenzhen (He et al., 2011) and New York City (Sun et al., 2011b), to explore the influence of regional transport on PM₁ loading and composition at sampling sites.

(2) The purposes of analysis the regional transport in this study are: (a) compare the air mass in the two January studies. Because we know that the air masses that originated from the direction between south and west (180 °-270 °N) belonged to the pollution air mass because there are a large number of polluting industries, such as power plants, steel mills, cement mills and the petrochemical industry, in this area. However, the air masses that originated from the direction between west and north (270 °-360 °N) belonged to the clean air mass because there are almost no pollution sources in this area. Therefore, we can obtain the information about the changes of the regional transport in the two January studies. Then we can analysis the reasons for the decrease of fine PM mass concentration in January 2014. (b) In our study, we analyzed the 48 h back trajectories and calculated every 6 h (at 0, 6, 12 and 18:00 local time (LT)) throughout the campaign. Meanwhile, the our observation was duration about one month. Therefore, we believe that there was enough time for the transport of pollutant from the surrounding areas of Beijing even the wind speed is slow in winter. Meanwhile, the Fig. 8c and d also showed that the slowest air mass was passed Hebei, Shandong, and Shanxi provinces and other air masses passed longer distance. Therefore, the pollutants that transmitted from other areas (such as Hebei province) would seriously affect the air quality in Beijing.

33. – Supplement: You claim that the factor extracted is related to coal because you expect that coal emissions are important. If you already know the important sources, then why take the trouble to perform the source apportionment. You cite the studies that extracted a coal burning factor in Beijing, but this shows on the one hand that what you did in the current study is not new and on the other hand the studies cited are all from your group, which do not really support your case. In general, as I have mentioned above, it appears that you did not examine and validate systematically the PMF results. This validation should be based on a systematic examination of the mass spectral profiles, the diurnals, the correlation between tracer and factor time-series. The study seriously lacks external data to support factor identification, all the more since the inorganic species were included in PMF. Amongst the 6 factors identified, the only factor supported by external data is traffic. The CCOA mass spectrum looks like that of BBOA, and additional data are needed to support your assignment. For the secondary factors, it would be more appropriate to compare a traditional organic only PMF to the all-species-PMF presented here, as in the case of organic only PMF more species are available for validation. Also, please remove the following statement: “Most importantly, the 6-factor solution was in agreement with the actual situation in Beijing”.

Reply: We are very grateful for this important comment.

(1) There are many studies that have proved coal combustion is a very important PM source in winter in Beijing. However, we won't know its characteristics, such as its contribution proportion, the relationship with other PMF factors, diurnal variation, etc. if we don't carry out the source apportionment work. For example, in many cities, we can make a preliminary judgment on the PM sources, such as the traffic source, dining activity, industrial source, secondary sources, etc. However, we can only carry out in-depth research based on source apportionment. This information is very important for assessing the effectiveness of emissions reduction policies and offering guidance as to their next steps.

(2) There were two purposes to citing other studies that extracted a coal burning factor in Beijing. First, we wanted to prove the rationality of our results. Second, we wanted to see if there were any some new changes in the contribution of coal combustion. Because the government has introduced many emissions reduction measures, focused on coal combustion, in Beijing and the surrounding area in recent years. Therefore, analyzing the changes in coal combustion sources in winter in different years was a very effective way to evaluate the effects of emission reduction measures.

(3) In this part, we cited the results reported by Sun et al. (2013a) to prove the rationality of our PMF results. In fact, our group and Sun's group are completely different research groups. Therefore, it was rational to cite their results here.

(4) In fact, we have provided a detailed discussion on the determination of the optimal number of factors. According to the suggestion of the reviewer, we performed the PMF_{OA} analysis and compared the time series of every factor and other tracers (section 3.2). Although no other tracers (except the nitrate, sulfate, chloride, EC, NO_x) could be used in our study, the analysis results were still sufficient to guarantee the correctness of our results. Because many studies have used the similar tracers for comparison. For example, figures 1 to 3 compared the time series of PMF_{OA} components and their tracers in Beijing (Sun et al., 2013a), Shanghai (Huang et al., 2012), and New York City (Sun et al., 2011). The PMF_{OA} results have been

added in the revised manuscript and are compared with the PMF_{AMS} and PMF_{OA} results in January 2013.

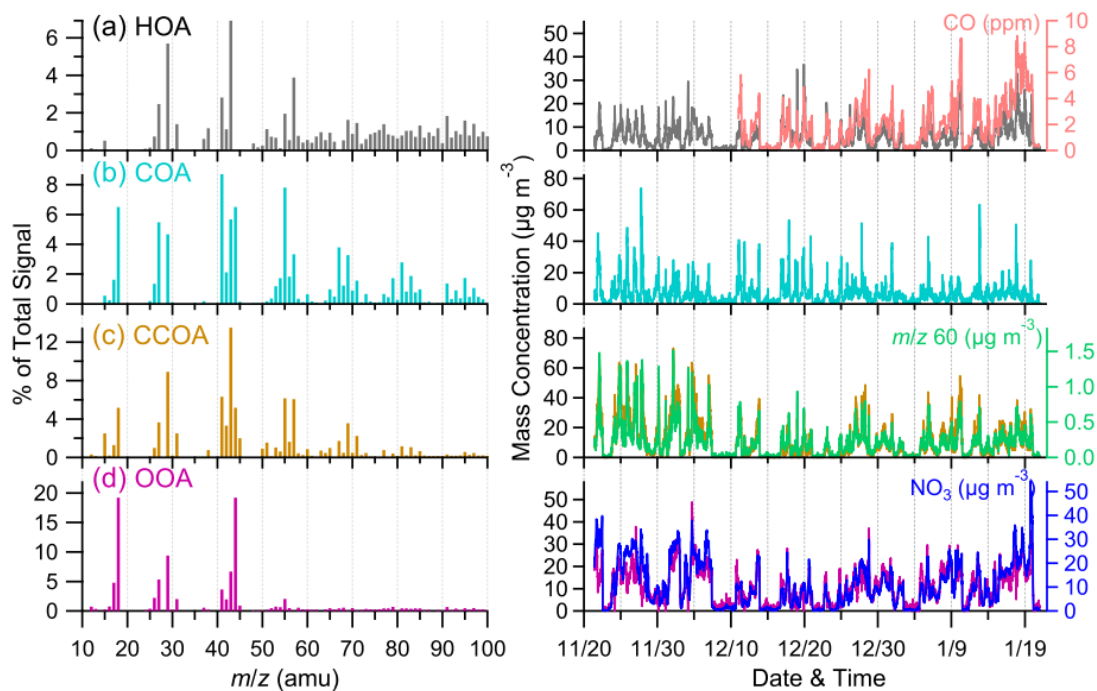


Fig. 1 The PMF results in Beijing reported by Sun et al. (2013a)

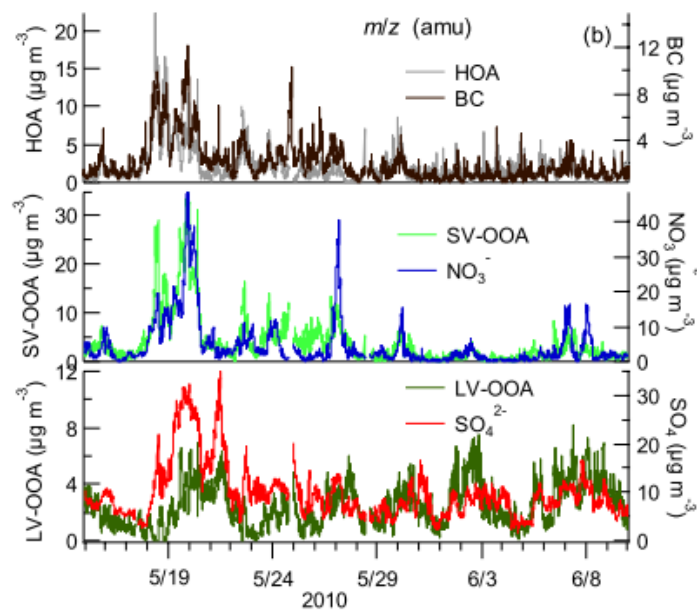


Fig. 2 The PMF results in Shanghai reported by Huang et al. (2012)

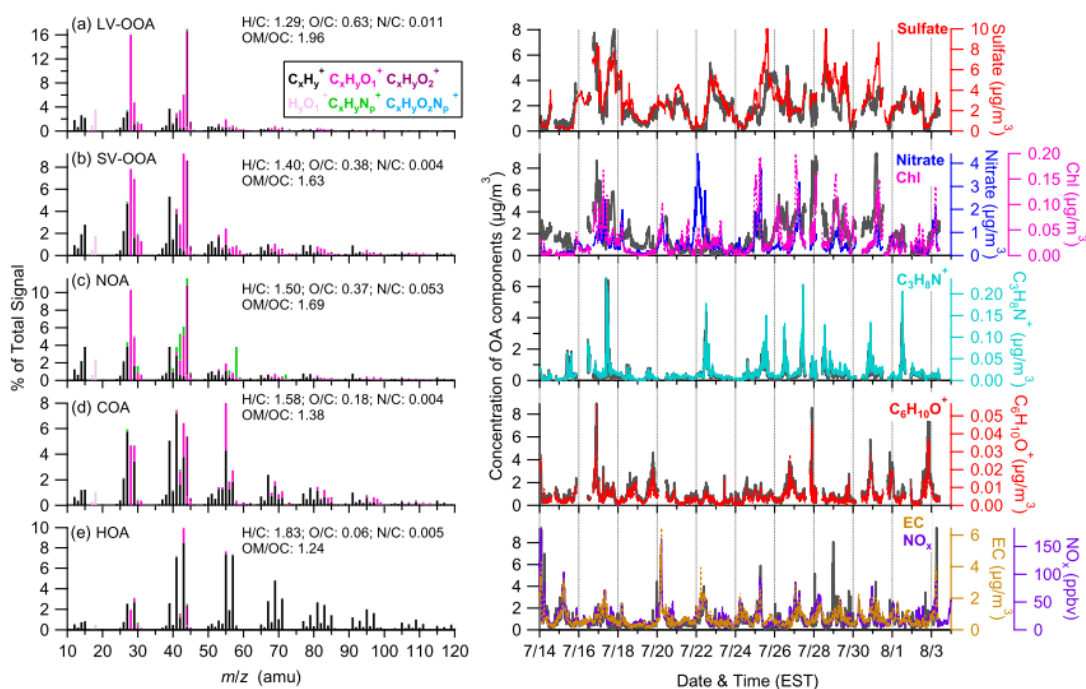


Fig. 3 The PMF results in New York city reported by Sun et al. (2011)

(5) The identified of CCOA factor was based on the following considerations: (1) The biomass burning mainly occurred in June for wheat and in late September to early October for corn. Then the biomass burning presented high contributions to fine PM (Yu et al., 2013; Sun et al., 2013b). While in winter, compared with the coal combustion for heating in Beijing, the contribution of biomass burning is much weaker. Figure 4a and b are the distribution of the fire spots, which are mostly caused by the biomass burning, in autumn (1 to 30 September 2014) and winter (1 to 31 January 2014), respectively. It can be found that the biomass burning activity was frequently occurred in the surrounding areas of Beijing in autumn. However, biomass burning activity in winter was much weaker than that in autumn. (2) Our observation site located in urban Beijing and the biomass burning is prohibited in this area. Therefore, coal will become the main fuel for heating in winter in Beijing. In addition, Zhang et al. (2015a) also thought that the contribution of biomass burning is highly seasonal and may be important during the harvest seasons (i.e., spring and fall) in Beijing. Therefore, we believe this OA component mainly emitted from the coal combustion in Beijing. Some useful information has been added in the supplementary information now.

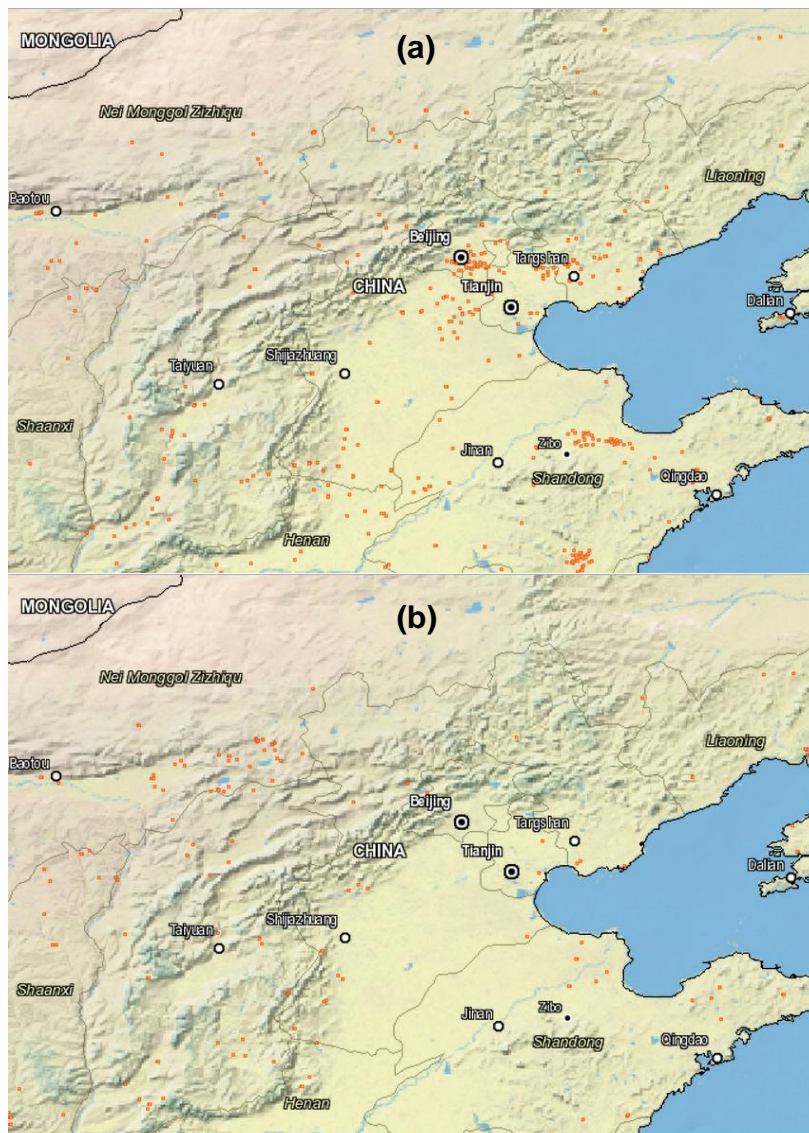


Fig. 4 The distribution of fire spots in North China in (a) autumn, and (b) winter (the study period of this study)

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