Authors' Response to Editor's & Referees' Comments

We thank the editor and referees' for their critical reading of the manuscript and constructive comments and suggestions, which helped to improve the quality of the MS. The MS is revised according to all the comments.

Editor

Please address the comments from the reviewer, especially for the writing and structure of the MS.

Response: We revised the manuscript according to all the comments and suggestions of the referee #2, mainly the writing and structure of the MS. Please see our point-by-point responses below and the revisions in the revised MS.

Referee #2

General comments:

This is a quite sufficient measurement report, the amount of data from different parameters is quite large. Therefore, it is easy to mask the main idea of the story. The authors tried to reveal the source and atmospheric processes of fine aerosols in Tianjin region based on the measurements of chemical components and stable isotopes of carbon and nitrogen. However, in my opinion, it is still hard to easily capture the main idea of the story when reading through the whole MS in current version. After the revision, the MS has indeed been improved. However, I still think the writing and structure of the MS require further improvement to make it much easier and clearer for readers to understand. Detailed comments could be found as follows:

Response: We thank the reviewer once again for his/her appreciation of our work and comments/suggestions. The MS is revised according to all the comments from the referee, and the point-by-point responses are provided below.

Specific Comments:

For the whole structure of the "Results and Discussion", I think it might be better to show as the following orders?

(1)Meteorology and backward air mass trajectories; (2) Concentration and seasonal variations of $PM_{2.5}$; (3) Concentration and seasonal variations of carbonaceous components; (4) Implications for $PM_{2.5}$ sources through relationships and mass ratios of carbonaceous components. In this section, I do think the relationship between $PM_{2.5}$ concentrations and carbonaceous components should be added, because for example, the authors have explained that the "EC directly emits from incomplete combustion of fossil fuels and biomass burning", therefore, the relationships between $PM_{2.5}$ concentrations and EC should be a clear indicator for the source of $PM_{2.5}$, but such kind of relationship is not shown in current version, so as the relationship with other carbonaceous components. And, I'm quite confusing with the relationship between WIOC and SOC in current version, why only the relationship between WIOC and SOC was shown? (5) Implications for $PM_{2.5}$ sources through $\delta^{13}C_{TC}$. In this section, would it be better to summarize the $\delta^{13}C$ of different sources (Fig. 11) into several types? There are too many different sources in current version, it is hard to compare; (6) Concentration and seasonal variations of nitrogenous components and other inorganic ions. In current version of MS, the authors introduced NH_4^+ and NO_3^- concentrations in section 3.4, while introduced water-

soluble nitrogenous components in section of 3.5. This is confusing because the $N\text{-}NH_4^+$ and $N\text{-}NO_3^-$ also belong to water-soluble nitrogen. (7) Implications for $PM_{2.5}$ sources through relationships of nitrogen components and other inorganic ions. (8) Implications for $PM_{2.5}$ sources through $\delta_{15}N_{TN}$. Summarize the $\delta^{15}N$ of different sources (Fig. 12) into several types?

Response: We agree with the reviewer's opinion fully, and re-structured the 'Results and Discussion' section by dividing it into 8 sub-sections, as suggested. Please see the subsections 3.1-3.8 of 'Results and Discussion' section in the revised MS.

Following the reviewer's suggestion, we included the relationships between $PM_{2.5}$ and OC, EC and WSOC in order to assess the sources of $PM_{2.5}$. Please see Fig. 6D-F and Lines 330-334 and 354-355 in the revised MS.

Generally, it has been recognized that WIOC might be produced by incomplete combustion of fossil fuels and cooking activities and composed of long chain aliphatic hydrocarbons, ketones, alkanes and polycyclic aromatic hydrocarbons. However, in recent times, it has been suggested that the WIOC could also be produced by secondary processes in the atmosphere. Interestingly, we found high correlation between WIOC and SOC in autumn, winter, suggesting that the secondary formation the WIOC is significant in the Tianjin atmosphere. In order to show such findings, we confined to present the linear relations between SOC and WIOC only, rather than with WSOC, which is known to be mostly produced by secondary processes, as well. We noted these points in the revised MS (see Lines 385-394).

In fact, we summarized the $\delta^{13}C$ and $\delta^{15}N$ of various source types such as marine and continental including biomass burning and fossil fuel combustion in Introduction Section in the revised MS (see Lines 90-95, respectively). Here, we confined to compare our results with the isotopic signatures of point sources and other literature values., in order to identify the potential specific sources.

Both the inorganic ions including NH_4^+ and NO_3^- and nitrogenous components are combined into subsection 3.6 in the revised MS. Therefore, such confusion doesn't arise now.

The section of "Ionic balance" in current version is better to delete, because I did not see any importance of this section on revealing the source and atmospheric processes of $PM_{2.5}$ in current description.

Response: Following the reviewer's suggestion, we removed the 'Ionic balance' section in the revised MS.

Technical corrections:

Lines 36-53: The authors introduced the EC, OC, SOC and WSOC in order, and then introduce EC and OC again, it is kind of circle, why don't put the two sections of EC and OC together?

Response: Following the reviewer's suggestion, we restructured this paragraph by first describing different sources of EC and OC followed by their atmospheric loadings and impacts, and then introducing the sources and impacts of SOC and WSOC. See Lines 36-54 in the revised MS.

Lines 85-88: Better show the range of the sources.

Response: Following the reviewer's suggestions, we included the range or average values of the sources in the revised MS (see Lines 90-95).

Lines 88-90: Better explain how the isotopic fractionation affects the isotope values of carbon and nitrogen.

Response: Following the reviewer's suggestions, we described how the isotopic fractionation occurs during the occurrence of chemical reactions and phase transitions and thus influence the corresponding isotope

ratios with aging in the revised MS (see Lines 96-101).

Line 92: Add reference after "……are significant".

Response: Following the reviewer's suggestions, we cited the appropriate references in the revised MS (see Line 103).

Lines 92-101: Kind of confusing, better make it clear, especially how to use $\delta^{13}C$ and $\delta^{15}N$ to investigate the aging process.

Response: We made it clear by adding a phrase "---, which could accelerate the enrichment of 13C and 15N in the particles, ---" in that statement in the revised MS (see Lines 106).

Lines 107-108: I don't understand why there are two different area of forest in Tianjin (2039 and 1364)? Better shown in percentage instead.

Response: In fact, the 2,039 km^2 area is the total forest land area and the other areas meant for developed and natural forest areas. However, in order avoid any confusion to the reader, we provided only the total forest area, including its % in the total Tianjin land area in the revised MS (see Lines 117-120).

Line 120: Add "SO42-, Ca2+, Mg2+……" after "inorganic ions"

Response: We added the list of inorganic ions measured (Cl^-, SO_4^{2-}, NO_3^-, Na^+, NH_4^+, K^+, Mg^{2+} and Ca^{2+}) in the revised MS (see Line 131).

Lines 147-148: I didn't buy it, cause the temperature could be more than ~30℃ in summer, this will still have minor effects on the samples?

Response: We agree with the reviewer's opinion that there will be minor effects of sampling artifacts at ~.30℃ in summer. In order to address such discrepancy, we toned down our statement by adding another phrase: "----, although we do not rule out them completely", in the revised MS (see Lines 156-159).

Line 184: Explain which ions?

Response: We added the list of inorganic ions: Cl^-, SO_4^{2-}, NO_3^-, Na^+, NH_4^+, K^+, Mg^{2+} and Ca^{2+}, in the revised MS (see Line 194).

Table 1: No units.

Response: We added the units of all parameters in both Table title and in the Table in the revised MS (see Table 1).

Lines 287-289: Seems this sentence belong to section 3.2.

Response: Following the reviewer's suggestions, we moved this sentence into the sub-section 3.2 in the revised MS (see Lines 262-265).

Lines 438-439: Seems belong to section 3.4.3.

Response: Such discrepancy doesn't arise now, because this section (3.4.3) is combined with the previous section and they appearing under the section 3.7 in the revised MS.

Line 466: Should be Fig. 8.

Response: We regret for the typo, and corrected it in the revised MS (see Line 520).

Lines 498-499: Why don't put the δ13CTC of fatty acids into Figure 11?

Response: Because this study is focused on $\delta^{13}C$ of TC and all the data provided in Fig. 9 is of only $\delta^{13}C$ of TC, we preferred to provide the $\delta^{13}C$ of fatty acids in the text rather than in the Fig. 9.