

## **Interactive comment on: “Biomass burning contribution to regional PM<sub>2.5</sub> during winter in the North China”**

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Air pollution from PM<sub>2.5</sub> is a major health and environmental concern in China. Accurate PM<sub>2.5</sub> source attribution is therefore critical for the development of successful air pollution control measures in the region. The study by Zong et al. investigates the sources of PM<sub>2.5</sub> at a background site in Northern China during the winter of 2014, utilizing different source apportionment techniques. By reporting detailed measurements of the elemental composition of PM<sub>2.5</sub> collected on 76 quartz filter samples, the manuscript provides interesting new data, which is further grouped into three source regions based on back trajectories analysis. In addition, radiocarbon measurements are used to apportion the carbonaceous fractions of the aerosols into fossil and biomass combustion contributions. These results are combined with a PMF model to derive the major sources of PM<sub>2.5</sub> in the region. While the combination of these different techniques is a very good approach for achieving accurate PM<sub>2.5</sub> source apportionment and can provide important insight into what is driving the elevated concentration in the region, there are several major problems in the reasoning, explanation and interpretation of the methods and the data reported in the manuscript.

One concern is the use of only one station at a background location, to interpret sources of PM<sub>2.5</sub> over a very large area without any discussion of the possibility of local sources affecting the measurements. In one part of the manuscript the authors hypothesize that the elevated PM<sub>2.5</sub> at the sampling site are due to the proximity of sources (p.10, line 3-5). But later based on their cluster analysis they say only 11% of the air masses were from the Shandong Peninsula (p. 11 line 18).

Further, measuring PM<sub>2.5</sub> concentrations based on filter mass is not accurate due to variability within the filter area. Each punch might weigh differently due to the inhomogeneity of the filter itself. There are no references about this technique being used anywhere else.

Radiocarbon was only measured on two samples, each representing an air mass coming from a different region. One sample cannot capture any variability in sources and is not a true representative of regional sources. It is also not clear what the uncertainties in the results are and how they could affect the source apportionment. When using radiocarbon, it is particularly important to accurately represent the uncertainty in aerosol samples, as they are very small and usually have high degree of uncertainty, which needs to be considered when interpreting the results.

With respect to the back trajectory analysis and clustering, the manuscript did not provide enough detail to explain how this was performed and the model for the clustering. Also the authors did not include the reference, giving credit to the original HYSPLIT model development.

In general, the manuscript was very hard to read and understand due to the language (and multiple typos and errors), which needs significant improvement. The title of the manuscript is also misleading, since the biomass-burning signal is not the main focus of the manuscript. The lack of appropriate citations makes it hard to track the accuracy and reliability of the statements and hypothesis throughout the manuscript and makes the interpretation of the results difficult. The details provided in the methods section are not enough to fully understand the analysis or to allow traceability or reproducibility of the results. The results and discussion section was hard to follow too due to the lack of structure within the text and paragraphs summarizing the result from the different experiments and drawing conclusions together.

A list of specific comments and questions are provided below:

- Missing citations throughout the introduction
- Spaces missing p. 3 line 20; p. 4 line 10, p. 13 line 20, p. 27, line 22
- The introduction (and the rest of the manuscript) does not explain how  $^{14}\text{C}$  provides information about fossil and modern sources.
- Page 5 line 9: Longkou district and Tianjin are not shown on map. Is Longkou the closest urban region and can it be considered a local source of urban emissions?
- Page 5, line 18: Why do the filters undergo a 24h equilibrium period at 25C? Wouldn't that potentially introduce contamination with the filters absorbing volatile organic compounds during that period?
- OC and EC page 6 lines 6-7: The DRI currently uses an updated protocol – IMPROVE\_A (*Chow et al., 2007*). Which one was used for this manuscript? <http://www.dri.edu/eaf-projects>
- Page 7, line 14: Radiocarbon methods are not cited properly (p. 7, line 14 Liu et al 2014 is not explaining the process) – correct citation is Zhang et al. 2015 (*Env. Sci. & Tech*) Radiocarbon-Based Source Apportionment of Carbonaceous Aerosols at a Regional Background Site on Hainan Island, South China
- Page 7 Line 16: which TOR protocol? (also acronym was not defined anywhere)
- Graphitization at CAS and AMS analysis – can you provide citation, sizes, black corrections?
- Page 8: HYSPLIT and EPA PMF v5.0 citations
- Page 9, line 15 The text does not follow the order reported in Table 1 (notes on tables and figures will be provided at the end)
- Page 9, line 17 What is the uncertainty of the max PM<sub>2.5</sub> value? Also, there is no need to report significant digits, as the range is so large
- Page 10, line 2: It is not clear where Tuiji Island is and how it is relevant in the comparison.
- Page 10, line 8-9. Write chemical formula for sulfate, nitrate and ammonium and use them throughout the text. The lack of consistency with the names of the chemical elements and radiocarbon vs  $^{14}\text{C}$  makes the text harder to read.
- Page 10, line 20-1. There are major issues with the chemical elements here – The ones written out are not the correct elements!
- Page 11, line 22: Mean test?
- Break next section in paragraphs. It is very hard to read.

- Page 12, second paragraph – write out chemical elements
- Page 12, line 21. Cite the sources for sea salt composition
- Page 13: It is really hard to follow and it might be helpful to break it up in a couple paragraphs
- Page 14: It is not clear how the Enrichment Factor (EF) method works, if/how it has been applied in other studies and how reliable it is. Original citations are missing. More description is necessary. It will also be helpful to plot the EF calculated for all elements.
- Page 14: Using OC/EC ratio and NO<sub>3</sub>/nss-SO<sub>4</sub> a traces :
  - Lack of citations OC/EC p. 14 line 21
  - Mean test (p. 15 line 2)?
  - When calculating OC/EC ratios what is the uncertainty in the OC and EC and the ratio.
  - Lack of citations SO<sub>2</sub> and NO<sub>x</sub> p. 15 line 7
- Page 15, line 18: Where is Chengde and how is it relevant?
- Source app of carbonaceous PM<sub>2.5</sub> (pages 16-19)
  - The back trajectory analysis is not clear in the description of how the samples were combined. The figure does not help either.
  - Page 16 line 11 – 19 unclear and belong in the methods
  - Page 18 line 2: What is the WSOC and WIOC concentration uncertainty? The reported numbers have two significant digits, but are the methods accurate enough to report concentrations of this precision?
  - M1 and M2 are significantly different, but could this be due to high level of uncertainty in the measurement? Can it possibly be affected by local sources?
- Page 19, line 15: F<sub>peak</sub> and Q are not defined
- Page 20, line 6: Citations missing
- Page 24, line 3: PMF and 14C results are compared within 3-4% precision, but it is likely the uncertainty is higher.
- Implications for PM alleviation – since it was mostly focused on biomass burning it seemed to not fully capture the scope of the work.
- It would have been very helpful if there were a section in the manuscript where all the results were drawn together so the reader can logically conclude how the different techniques complemented each other and what the final result of the different analytical techniques was.
- There are a lot of typos in the references.

#### Tables and Figures:

Table 1: It is not clear why the table is split in two and why the units are different. It would be much easier to interpret the content if everything is aligned into 3 columns and the units are consistent.

Table 2: Make into one page

Table 3: Uncertainty needs to be reported for each measurement

Table 4: Rows 1, 3, and 4 do not add up to a 100% when horizontally summed. The difference is minimal, but what is the reason for it? Also the significant digits are probably not necessary, based on the accuracy of the analysis.

Figure 1. Some locations that were mentioned in the text are not included in the figure.

Figure 2. This is very difficult to follow, a more detailed description of what the figure is showing will be helpful

Figure 4. The figure caption should explain why the B&B and Fossil emissions from the PMF do not add to a hundred in the bars. It is described in the text, but a note in the figure will make it much easier to understand.