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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Fossil vs. non-fossil sources of fine carbonaceous aerosols in four Chinese cities during the extreme winter haze episode in 2013" *by* Y.-L. Zhang et al.

Anonymous Referee #2

Received and published: 28 November 2014

This paper describes a source apportionment of the carbonaceous component of 24 h samples of PM2.5 collected in four major cities in China in January 2013, when total PM2.5 concentrations reached very high levels (up to 100s μg m-3). The source apportionment is principally based on the proportion fossil/non-fossil carbon in the TC, and in the OC and EC fractions, as determined from accelerator mass spectrometry (AMS) measurements of the amount of the radiocarbon isotope, 14C, in the carbon. These data were supplemented by measurements of the levoglucosan, mannosan and water-soluble K+ concentrations in the PM2.5 which provide additional information for the source apportionment of biomass burning.

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Interactive Discussion
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Both the analytical and data-interpretation methodologies for this study follow very closely that of a number of previous studies, particularly in Europe, undertaking similar source apportionment of the carbonaceous aerosol. This has the advantage of use of methodology that has already been through the peer-reviewed literature. The novelty here is its application to PM2.5 samples in very large Chinese cities that have experienced PM2.5 levels up to an order of magnitude greater than in many European urban locations. Poor air quality in China is clearly a major cause for concern and it is important for all, particularly policy-makers, to have insight into the constituent components and sources of the PM2.5.

Key results from this study include the finding of substantial non-fossil contribution to OC (in common with similar studies globally) and the inference that a substantial fraction of this non-fossil OC is primary rather than secondary in nature. The authors also compared their source apportionments between the most heavily-polluted days and moderately polluted days and noted that despite the increase in absolute masses the proportion of secondary was even slightly higher.

The paper describes thorough experimental procedures and appropriate data analysis methodologies. The writing is generally fluent, although occasional grammar and comma punctuation usage requires amendments. I have a couple of points regarding scientific interpretation, and the remaining points are largely concerned with presentation. I recommend this paper as suitable for publication in ACP following attention to these issues and any other relevant issues raised by other reviewers.

(1) The authors could likely gain some greater insight into the origin of their various carbon fractions by undertaking an air-mass back-trajectory investigation for the days of their samples, particularly through a comparison of the high-pollution vs. moderate-pollution days.

(2) The description and nomenclature of the divisors used in Equations (3) and (5) was not immediately clear to me, i.e. the terms fM(bb) and fM(nf) in the two equations,

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respectively. I interpret these terms as being the values used to correct the fM(EC) and fM(OC) values to yield a fraction contemporary carbon in EC and OC, rather than the fraction modern carbon in EC and OC. In my opinion, the terminology fM(bb), and the phrase "a 14C reference value for biomass burning" do not make it clear that the reference value is the percent modern in EC emitted from burning contemporary carbon-containing fuel. Likewise, for lack of clarity in Equation (5). The application of these terms does not become clearer until the text in point #1 on P26267.

Technical corrections:

Abstract: State the collection duration of each PM2.5 sample (24 h).

Abstract: State the number of samples analysed for 14C, i.e. the number of sample values that underpin the mean and standard deviation of source apportionment proportions presented in the abstract.

P26259, L6: Rephrase end of the sentence as "...was conducted at the four major cities of Xian, Beijing, Shanghai and Guangzhou." (The fact that the study was conducted in several large cities in China has already been stated in the previous sentence.)

P26259, L7. Delete "An effective" and start the sentence directly as "Statistical analysis of..." Remove the words "An effective" from in front of similar phrasing elsewhere in the paper where the Latin Hypercube technique is mentioned; it is a redundant adjective.

P26259, L11: Rewrite as "across all sites."

P26259, L19: Delete "rather".

P26260, L1: Delete both the two commas.

P26261, L9: Delete comma.

P2621, L22: Please provide a quantitative indication of what is meant by "extremely high concentrations of PM2.5"

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P26263, L4: “Six filters were selected. . .”

P26265, L12: Sort out the formatting of the citation in this sentence.

P26267, L21: Insert “for” to read “To correct for the. . .”

P26271, L16: Correct the sentence containing the phrase “. . .with an equally enhancement. . .” which doesn’t make grammatical sense.

P26272, L13: Should this read the “ratio of ECf to OCf”?

P26273, L6: Insert comma after “marker”

P26273, L7: Insert comma after “)”

P26277, LL6&&: Provide a definition here of the two acronyms MPD and HPD.

P26295: Delete the word “below” from the last line of the caption of Figure 6.

Supplementary information, caption to Table S1: Should read “The sampling dates for the. . ..”

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 26257, 2014.