

Interactive comment on “Diurnal variations of organic molecular tracers and stable carbon isotopic compositions in atmospheric aerosols over Mt. Tai in North China Plain: an influence of biomass burning” by P. Q. Fu et al.

Anonymous Referee #2

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This manuscript presents measurement results of aerosol particles from a mountain site in east China as part of the MTX2006 campaign. A large number (~130) of individual organic compounds was quantified in the ambient particulate matter which was collected with rather high time resolution (compared to typical filter-based sampling). The detailed chemical speciation included numerous molecular source tracers, which allowed for a qualitative assessment of different types of source contributions. Specifically, the strong influence from biomass burning was demonstrated with the chemical aerosol characteristics along with fire counts and air mass history analysis. Diagnostic

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tracer ratios confirmed that the biomass burning activities occurred in form of agricultural residue (i.e., wheat straw) combustion, which resulted in a substantial increase in the aerosol loadings and was characterized by different diurnal patterns. Stable isotopic composition of total carbon in the aerosol samples revealed additional insights into the sources and transformations of aerosol particles during atmospheric transport, highlighting the importance of secondary organic aerosol formation.

As measurements of individual organic compounds and stable isotopic carbon ratios at high-elevation sites are rare, the findings from this study are very valuable in the context of regional-scale pollution sources, transport and effects. The results presented in this manuscript are obtained from experiments that had a sound scientific approach and were carried out with sufficient QA/QC measures. The interpretation of the data is reasonable and the discussion of the implications is logical, except for the attempt to quantify the relative source contributions, as discussed in more detail under the specific comments. In conclusion, I highly recommend publication of this manuscript in ACP upon consideration of the comments and suggestions given below.

Specific comments

Page 9088, lines 10-11: The measurement period during this study was rather short, while this limitation is, to some extent, compensated by the relatively large number of samples (due to the high sampling frequency of 3 hours), yet caution should be used with the interpretation of the resulting data due to the limited number of sampling days.

Page 9088, lines 10-11: The statement that "the concentrations of WSOC are well correlated with those of OC, suggesting a similar source" is obvious, as WSOC is, by definition, a portion of OC, and thus the implication is self-explanatory. On the other hand, it would be more meaningful to state that WSOC constituted an important fraction of OC based on both the good correlation and high percentage.

Page 9088, lines 14-15: It seems as if the authors mistakenly wrote "black carbon" instead of "WSOC", or else this statement is out of place here. Coincidentally, this

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raises the question regarding the BC or EC content in the TSP samples in early and late June – while the ambient EC concentrations are listed in Table 1, it would be helpful for the readers to have a short statement regarding the EC patterns in this paragraph. In addition, the authors should discuss the OC/EC ratios here, which can be a useful indicator of biomass burning (and/or biogenic) source influence.

Page 9090, lines 1-6: Indeed, the L/M ratio can be a useful indicator of specific types of biomass that were burned. Therefore, the authors should compare their findings with those from other studies, such as those reported in Fabbri et al. (2009) or Sheesley et al. (2003). In fact, the L/M ratio calculated from the data by Sheesley et al. for rice straw smoke aerosol is very similar to that reported in this study. Moreover, Engling et al., (2009) discuss the use of these diagnostic ratios with a specific focus on emissions from agricultural residue combustion. The high ratios observed in this study in early June (with enhanced biomass burning activity) agree very well with those for other types of straw (especially rice straw), confirming the utility of these characteristic ratios as indicators of straw burning.

Page 9091, lines 5-12: The discussion on the ambient temperature dependence of phthalates is not logical to me. First, how is the presence of these species in particulate matter explained if they are released due to evaporation (i.e., going into the gas phase) from polymers? Second, if the concentrations show a noon time peak, wouldn't that indicate local emission sources rather than regional-scale transport? Presumably, there are no (or insignificant) local sources at this mountain site, and therefore the origin is expected to be from upwind source regions, which are not expected to show a clear diurnal pattern.

Page 9092, lines 13-14: Even higher contributions were found in the study by Zhang et al., (2010) at a remote site in South China, which can be mentioned here, especially as their findings are from the same country and a remote site (even though at lower altitude and latitude) as well.

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Page 9092, lines 16-19: Another very recent study also showed a good correlation between biomass burning and fungal spore tracers (Yang et al., 2012), and can therefore be cited here.

Page 9098, lines 10-17: It is not clear how the authors derived these numbers – please specify! Merely adding up the individual tracer compound concentrations and calculating the relative abundance does not indicate source contributions in a quantitative manner, in my opinion. If this is what the authors did with the percentages listed here, then there is little meaning to those numbers. On the other hand, estimating source contributions based on emission factors and ambient tracer concentrations is a valid method that has been used in a number of studies, although there are, without doubt, uncertainties associated with this approach, as indicated by the authors. Thus, the contributions from biomass burning, fungal spores and SOA presented here are warranted, whereas the other percentages should be omitted. One more comment/question (related to page 9098, line 22): while there have been at least a few source profiles published for agricultural residue burning, why do the authors use factors from a study in the Amazon basin (which may have been strongly influenced by biomass burning activities, yet it doesn't provide true source emission factors)?

Technical corrections:

1. The definite article "the" is not used properly throughout the manuscript, i.e., it is missing in many cases, such as on p. 9081, line 5 (before "Mount"); p. 9081, line 6 (before "North"); p. 9082, lines 22 (before "Asian"); p. 9083, line 1 (before "North"); etc.
2. Please, check the correct use of singular and plural forms throughout the manuscript and make corrections, such as for "plays" on p. 9082, line 9, or "compositions" on p. 9083, line 23.
3. Page 9082, line 14: Change "comprise" to "are comprised".

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4. Page 9083, line 1: Delete ", which".
5. Page 9083, lines 11-13: It would be helpful to see references providing a general overview of the Mt. Tai Experiment 2006 (MTX2006) and some related measurements during MTX2006, such as those by Yamaji et al. (2010).
6. Page 9089, line 6: Change "in" to "on".
7. Page 9089, lines 19-20: While levoglucosan is released during the thermal decomposition of both cellulose and hemicelluloses, these isomeric anhydrosugars (mannosan and galactosan) are actually only emitted from hemicelluloses (i.e., from their parent sugars, mannose and galactose, which are not present in cellulose).
8. Page 9092, line 6: Change "innumeros" to "numerous".
9. Page 9093, lines 9-11: Additional references which are relevant to this statement are the recent papers by Despres et al. (2012) and Froehlich-Nowoisky et al., (2012).
10. Page 9094, line 17: Change the sentence to "... an indicator of primary pollutants in motor exhausts ...".
11. Page 9096, lines 17 and 20: Add "to" before "each".
12. Page 9096, lines 21-22: The meaning/point of this sentence is not clear.
13. Page 9097, line 17: Change "occurred" to "occurring".
14. Page 9097, lines 22-25: This sentence sounds awkward and should therefore be reworded.
15. Page 9100, line 13: Change "of" to "the".
16. Fig. 6: Is the number of significant figures used for the correlation coefficients really warranted by the measurement certainty? Probably not, and thus they should be reduced, e.g. similar to Fig. 13.

Here is a list of references used in this review, which should be added in the manuscript:

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- Despres et al., *Tellus B*, 64, 2012 - Engling et al., *Aerosol Science & Technology*, 43, 662-672, 2009 - Froehlich-Nowoisky et al., *Biogeosciences*, 9, 1125-1136, 2012 - Yamaji et al., *Atmospheric Chemistry & Physics*, 10, 7353-7368, 2010 - Yang et al., *Science of the Total Environment*, 431, 68-77, 2012

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