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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Source apportionment of carbonaceous aerosols in Xi'an, China: insights from a full year of measurements of radiocarbon and the stable isotope ^{13}C" *by* Haiyan Ni et al.

Anonymous Referee #3

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General comments:

This manuscript reported datasets of carbon isotopes (13C and 14C) of OC and EC in a major inland city of China, Xi'an, during one-year sampling, which were used to study the source apportionment of carbonaceous aerosols by combining 13C and 14C with Bayesian Markov chain Monte Carlo (MCMC) scheme. The data and methodology are reliable and novel. This paper shed some new light on the source apportionment of carbonaceous aerosols by distinguishing coal and liquid fossil fuel contributions to EC, C3 and C4 plant to biomass burning. The paper is relatively well-written, and it should be acceptable for publication after some moderate to major revision.

Major points: 1. Be clear re mean or median values of source contribution, E.g., P1/L23, the 45% and 31% are median values in Figure 4. Need to be consistent in the manuscript. 2. The flow of introduction is not well organized, and some part of the 14C introduction should be moved to the method part. I suggest re-organization and strengthening of the scientific objectives of this study in the introduction. In addition, you need to explain why we need to further distinguish the coal and fuel combustion in EC but not OC? Fossil sources contribute averagely 46% to OC based on your results, so it is important. 3. Provide more clear details of blank/contamination evaluation for 14C analysis, instrumental analytical precision and mention of source markers (S2) in the method part. 4. The result section needs to be better structured and written. There are many parts, specially, sections 3.4 and 3.5, should be moved to discussion; and formulas could be moved to methods part. 5. There are many comparisons without in-depth discussion in the discussion section. And comparison among different methods and different climate event seems not reasonable in 4.3. 6. The Conclusion part was too long. I suggest summarizing the key points.

Minor points and suggestions: Introduction: P2/L1-10 This part didn't emphasize the importance of carbonaceous aerosol very well. And the structure and description are very similar to the second paragraph of the introduction from Zhang et al., (2015a, Atmos. Chem. Phys). Need to revise. P2/L16: The definition and expression of fraction modern is not explicit. "The 14C content" is not a ratio as "fraction modern". I don't think the standard need normalize for fractionation to 13C=-25‰Ỳou can refer to Stuiver and Polach (1977) and modify this sentence. I also suggest move this "14C result report" part to the Methods part. P2/L25 Clarify which kind of 14C studies only have two datasets. . . . . seasonal variations? TC or other? For example, Zhang et al. (2015b, EST) also reported annual and seasonal variations of EC in Beijing. P2/L27-30 It's better to introduce 13C first and introduce 14C as a novel tool. The 13C values of distinct sources you listed overlap with each other. P3/L7: same question as above. P3/L9-13 You need to provide 13C numbers for example to make readers to remember the trend between different processes.

Methods: P4/L1 replace “pre-fired” with “pre-baked” P4/L3 what’s the standard for season’s classification? Reference? How can you classify autumn to middle day? P4/L15 instrumental analytical precision? Do you have field blank? P5/L9-13 and L21-25 The whole blank/contamination should include all blank produced during experimental process and it is very important to know if the contamination is modern or fossil for 14C analysis. For combustion process, are the stds modern and fossil? Give the mass and F14C value of the stds. Also, provide the F14C value of combustion contamination. For 14C analysis, give the mass and F14C value of contamination you got in this study. Did you have F14C value of blank filter? Provide which blank you used to correct your 14C data. P6/L18 Is the “fbb” the same as “fbb” at L1?

Results P7/L10 The number of seasonal samples in Table S2 is not the same as in sampling part, need to clarify this inconsistency. P8/L20 unify the decimal place to one in the whole manuscript. P8/L30 Is it possible that combustion of a mixture of C4 and C3 plants or liquid fuel will results in the 13CEC values of around -24‰ P9/L26 what do the grey and dark green rectangle mean in Figure 3? P10/L1 Sections 3.4.1 is not real results of this study P10/L10 Provide the four-source calculation formula in 3.4.2 section and change the name “MCMC3” in 2.6 section. P10/L28 “5 times less than in summer” should use “lower than” P10/L29-32 The proportion of liquid fossil fuel combustion in winter (more coal burning) lower than summer (more traffic emission) make sense, why is not your expectation? P11/L16 mean or median? P12/L14 Does the OCo, nf mean observed non-fossil OC?

Discussion P13/L16 Add “Characteristics” in front of 4.1 title. P13/L17 Clarify which ffossil (EC) you used in comparison, the MCMC4 or F14C? Because others use the F14C deduced values. P13/L25 should be 76% as shown in Figure 4. P15/L4-8 Discussion here is not very convincing. The contribution of biomass burning to EC is the lowest in summer, but the highest contribution of biomass burning to EC occurred in winter (most corn stalk burning in winter, Figures 4 and S5), why no significant correlation was found in winter? P15/L16-20 I don’t think it’s reasonable to directly compare

results of different methods, e.g., you got contribution of biomass burning to EC by MCMC4 with 4 sources while Zhang et al. (2015) got the fraction by 2 sources. Furthermore, taking into account of the error bar, the fraction fossil (76%)/ biomass (24%) of this study are the same to Zhang et al. (2015). Finally, Zhang et al. (2015) studied samples during the extreme winter haze episode of 2013. P15/L23-28 The same question as above. Because the PMF model didn’t use 14C, is this reasonable for comparison? P16/L7 and Figure 7 clarify the relationship between vehicular emissions and liquid fossil fuel combustion somewhere before discussion. P17/L3 Will the biogenic emission to OC result in lower 13C values than EC?

Conclusions condense and summarize the key points of this study in this part.

References Check carefully the papers of the same author, e.g., you have two Zhang et al. (2015a) and where is Zhang et al. (2014a)? I think in section 4.3, you refer to Zhang et al., (2015a, Atmos. Chem. Phys.,)

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