

## ***Interactive comment on “Sources and formation of carbonaceous aerosols in Xi’an, China: primary emissions and secondary formation constrained by radiocarbon” by Haiyan Ni et al.***

### **Anonymous Referee #2**

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The paper acp-2019-437 by Ni et al. deals with carbon isotope measurements ( $^{14}\text{C}$  and  $\delta^{13}\text{C}$ ) measurements on carbon fractions carried out in China. The analysed samples cover 33 days throughout one year, covering all seasons and low, medium, high concentrations. The paper is clear, generally well written and the presented data are of interest for the scientific community and for future development of efficient abatement strategies.  $^{14}\text{C}$  data on carbon fractions are still relatively rare due to particular treatment of the sample and the need of accelerator mass spectrometry for isotopic ratio quantification. Nevertheless, few major concerns should be solved before the paper can be published on ACP.

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Major concerns Pag.3 line 17: “1-year 14C measurements”. From this sentence, I would expect high percentage of day coverage throughout the year. Opposite, Figure S1 evidences that 33 days are covered (less than 10% day coverage). The reviewer is aware of the difficulties related to 14C measurements and appreciates the efforts to make the analyses representative of all seasons and aerosol loadings. Nevertheless, the sentence is somewhat misleading. Please rephrase.

Paragraph 2.2: information on field blanks is completely missing and should be added

Pag.4, line 21-22: “Extraction of EC was done by heating the carbon that remained on the filters at 850 °C for 5 h”. In air or oxygen? Could you provide information on EC recovery for this kind of analysis (e.g. compared to EC quantification by TOT?). Is it similar to the one for 14C analysis?

Pag.6, line 24: “Currently, the F14C of the atmospheric CO2 is approximately 1.04 (Levin et al., 2008)”. Why do not using more updated values? (see e.g. <https://www.atmos-chem-phys.net/18/6187/2018/acp-18-6187-2018.pdf>)

Pag.7, line 15: “F14Cbb =  $1.10 \pm 0.05$ ”. Please clarify assumptions on wood age and fell date.

Pag.7, line 18: “F14Cnf =  $1.09 \pm 0.05$ ”: it seems to be fully dominated by wood burning. Please, clarify how it was obtained.

Pag.9, line 10-17: overlapping interval for expected  $\delta^{13}\text{C}$  for nearly all sources is present. This makes the analysis very weak, also considering that results are in contrast with 14C results (see pag.15, last paragraph). The reason to maintain this section and the related analyses should be better clarified. Figure S4 should be added to the text as it evidences the difficulties in apportioning coal and liquid fossil fuel contributions separately

Pag.10, line 16: “slight but consistent tendency”: what is “slight”? And in what sense “consistent”? The authors should specify the statistical approach used to verify “con-

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sistency”.

Page 10, line 22: “lower in other seasons (around 15%) with a slightly lower values in spring ( $14 \pm 3\%$ )”. Is spring really different compared to autumn and summer? As it is mentioned, it should be proved by statistical tests)

Page 10, Lines 21-29: table S1 merits to be added to the manuscript, as not all the numbers are reported in the text.

Page 11, Line 5: “ $6.8 \pm 6.0 \mu\text{g m}^{-3}$ ”. Maybe interquartile range is more significant than standard deviation, as the data distribution is not expected to follow a gaussian curve. Same comment for analogous representation of absolute concentrations in the rest of the text (e.g. pag.11, lines 6, 22)

Pag.11, line 21-23: “larger than”, “comparable with”: which are the statistical criteria used to evaluate comparability?

Pag.12, line 13: “The fossil OC is less water soluble in winter with lower (WSOC/OC)fossil ratios of around 0.5 than in the warm period”. What is “warm period”? Why indicating the value during winter and not during the warm period? Are the differences statistically significant, also considering the limited number of data available?

Paragraph 3.3: similarities in  $\delta^{13}\text{C}$  reference values for different sources affect the results presented here. The results show very high variability and this should be better commented in the text, also in the light of figure S4.

Pag.13 line 31: “moderately”. Quantify and evaluate statistical significance

Pag.14, line 1: “more constant”. Compared to what?

Pag.14, line 24: “rapid”. Please quantify (hours? Days?) and justify the sentence.

Pag.15, line 31: “Those contradictions will be discussed in the following section”. Coal is hardly mentioned in the following paragraph, thus it is unclear what the authors are

referring to.

Pag.16, line 11 and Pag.17, line 6: “slope of 1.31, and intercept of 0.32 and an R2 of 0.92”. “a slope of 0.62, and intercept of 0.01 and an R2 of 0.92”. As important uncertainties affect quantities both on x and y axis, 2-sided (Deming) regression should be attempted for better representation of these regression lines

Pag.17, line 10. “that a small fraction of primary fossil OC is water-soluble (Dai et al., 2015; Yan et al., 2017).”. This sentence should be moved more above, as it is also a justification of higher fossil POA compared to fossil WIOC.

Pag.19, line 2: “We suggest that WIOC<sub>fossil</sub> and WSOC<sub>fossil</sub> are probably a better approximation for primary and secondary fossil OC, respectively, than POC<sub>fossil</sub> and SOC<sub>fossil</sub> estimated using the EC tracer method”. This is in contrast with the sentence at the previous point.

Minor comments Page 4, line 15: “< 0.2  $\mu\text{g m}^{-2}$ ) compared to the TC loading of the samples (13–246  $\mu\text{g m}^{-2}$ ”. Replace with “< 0.2  $\mu\text{g cm}^{-2}$ ) compared to the TC loading of the samples (13–246  $\mu\text{g cm}^{-2}$ ”

Pag.5 line 7: “WSOC can be calculated as the difference between OC and WIOC”. Unclear why this sentence is here. The previous reference to radiocarbon measurements is confusing (as radiocarbon determination is not carried out as difference, as explained on page 7)

Page 5, line 19: “By water-extraction, water-soluble OC (WSOC) is removed from filter pieces (Dusek et al., 2014)”. The role of WSOC removal as a key procedure for reducing the impact of possible pyrolysis on  $^{14}\text{C}$  measurements of EC merits to be better evidenced as a key step for the correct  $^{14}\text{C}$  in EC measurement. In the years 2012-2014 three thermal treatments were developed nearly in parallel and all of them identified WSOC removal as a key step for radiocarbon measurement on EC. Suitable reference should include also Zhang et al, 2012 (<https://doi.org/10.5194/acp-12-10841-2012>) and

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Bernardoni et al, 2013 (<http://dx.doi.org/10.1016/j.jaerosci.2012.06.001>). Please note that these were the methods object of the inter-comparison reported in the mentioned Zenker et al., 2017 papers.

Page 10, lines 23-24: “Beijing shows a very different seasonal trend, where fbb(EC) was lowest in summer ( $\sim 7\%$ ) and increased to  $\sim 20\%$  during the rest of the year (Zhang et al., 2017)”. Please, introduce the sentence, e.g. “By comparison with literature data for Beijing” Page 10 line 30 (and following): change “around” with “about”

Pag.16, line 1: “Fossil WIOC (WIOCfossil) and WSOC (WSOCfossil) has been used”. Change into “Fossil WIOC (WIOCfossil) and WSOC (WSOCfossil) have been used”

Pag.16, line 28: “Thus, an overestimate of POCfossil result have two causes”. Change into: “Thus, an overestimate of POCfossil result has two causes.

Pag. 17, line 27: “An increased contributions”. Change into: “An increased contribution”

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