

Response to Reviewers' Comments and Suggestions

Reviewer's Comments

Authors' responses and revisions

Comments from Referees

Author's response

Author's changes in manuscript

Reviewer: 2

Comments:

This manuscript applied a powerful radiocarbon source tracer to apportion fossil fuel and biomass/biofuel contributions to carbonaceous aerosols in ten cities of China. The method was well established. Although the sample numbers are limited for each city (two samples), the result contain new message for sources of organic carbon, elemental carbon, water soluble organic carbon, primary and secondary aerosols in Chinese cities. These carbonaceous aerosols are included as major concerns for climate changes and human health. The conclusion therefore is important for air pollution mitigation in China. Before publication on ACP, some technical improvements are suggested.

Line 46, "fossil fuels " changes to fossil fuel combustion.

Response and Revisions: "fossil fuels" has already changed into "fossil fuel combustion."

Author's changes in manuscript: "EC is formed either from biomass burning (BB; e.g., wood fires, heating) or fossil fuel combustion"

Line 56, 2007b;Docherty et al., 2008;Mayolâ RBracero et al., 2002;Weber et al., 2007a); (Huang et al., 2014). Error.

Response and Revisions: Thank you for pointing out this. We have already made the correction in the revised manuscript (line 56-57).

Author's changes in manuscript: "(Weber et al., 2007b;Docherty et al., 2008;Mayol - Bracero et al., 2002;Weber et al., 2007a;Huang et al., 2014)"

Line 57, Several methods have been introduced to identify and quantify OC emission sources. Please show more methods for aerosol source apportionment; other methods like receptor models (PMF, CMB), and dispersion models.

Response and Revisions: Thank you for your suggestion. The references have already added in the revised manuscript (line 59-60).

Author's changes in manuscript: "Several methods have been introduced to identify and quantify OC emission sources, such as the use of organic molecular tracers (Simoneit et al., 1999), receptor models (PMF, CMB)(Singh et al., 2017;Bove et al., 2014;Marcazzan et al., 2003), and dispersion models (Colvile et al., 2003);"

Line 65 14C level (Szidat et al., 2009) Hence, 14C measurements can provide information about the. Full stop had been omitted.

Response and Revisions: Thank you for pointing out this. Full stop has already added in the revised manuscript.

Author's changes in manuscript: "shows a high contemporary 14C level (Szidat et al., 2009)."

Line 66: Numerous studies have been performed on the regional background of carbonaceous aerosols at urban sites. I prefer to change this sentence to: Numerous studies have been performed at urban sites to assess carbonaceous aerosol sources at the regional scale.

Response and Revisions: This sentence has been changed.

Author's changes in manuscript: "Numerous studies have been performed at urban sites and background sites to assess carbonaceous aerosol sources."

Line 68: contemporary carbon was the dominant pollutant in carbonaceous aerosols at a background site; The references should be cited for this conclusion at a background site (which one, it is better to detail the background site).

Response and Revisions: We have already added references and pointed out the detailed background sites in the revised manuscript (line 70-73).

Author's changes in manuscript: "For example, contemporary carbon was the dominant pollutant in carbonaceous aerosols at a background sites such as Ningbo and Hainan stations (Liu et al., 2013a;Zhang et al., 2014c)"

while a significant difference was found among seasons at urban sites (Yang et al., 2005;Chen et al., 2013;Liu et al., 2013a;Zhang et al., 2014b;Liu et al., 2014a). This is a new/independent sentence which suggests seasonal variations at urban sites. The conjunction word "while" is not suitable since the seasonal variations have no clear relationship with the previous result from a background site.

Response and Revisions: We are sorry for the misunderstanding and thank you for your suggestion. We have already revised the sentence.

Author's changes in manuscript: "In urban, the relative carbon contributions have shown a significant seasonal difference (Yang et al., 2005;Chen et al., 2013;Liu et al., 2013b;Zhang et al., 2014a;Liu et al., 2014a;Zhang et al., 2017)"

Line 72: aerosols (Gelencsér et al., 2007;Ding et al., 2008;Lee et al., 2010;Yttri et al., 2011). It is better to add one or two latest references. The combination of organic tracer and radiocarbon diagnosing is the main advantage of this research. Therefore, it should have one or two latest literatures to support the hot topic of this method.

Response and Revisions: Thank you for pointing out this. We have already added new literatures in the revised manuscript (line 76-77).

Author's changes in manuscript: "A combination of 14C analysis and organic tracer determination allows for more detailed source apportionment of carbonaceous aerosols (Gelencsér et al., 2007;Ding et al., 2008;Lee et al., 2010;Yttri et al., 2011;Zong et al., 2016;Liu et al., 2015;Zhang et al., 2014b)"

Line 74: the beginning of the period of widespread hazes. Where? Probably it may be specified

in China.

Response and Revisions: We are sorry for the misunderstanding and thank you for your suggestion. We have revised the sentence.

Author's changes in manuscript: "In this study, sampling was conducted in 10 typical Chinese cities during early winter when heavy haze pollution frequently occurs in this season."(line 78-79)

Line 75: carbon fractions such as WSOC, WINSOC and EC, along with water-soluble inorganic ions (F-, Cl-, SO₄²⁻, NO₃⁻, NH₄⁺, Na⁺, K⁺, Ca²⁺ and Mg²⁺) and anhydrosugars (levoglucosan, galactosan and mannosan). The details of water-soluble inorganic ions and anhydrosugars in brackets should not be showed in the introduction, while they should appear in method or result.

Response and Revisions: Thank you for your suggestion. We have revised the sentence.

Author's changes in manuscript: "Carbonaceous aerosols, including different carbon fractions such as WSOC, WINSOC and EC, along with water-soluble inorganic ions and anhydrosugars, were analyzed in PM_{2.5} samples." The details of compounds have already shown in the method section.

The last paragraph of Introduction, authors may include some information for the advantage of the combination of radiocarbon and anhydrosugar tracer. In introduction, authors should clarify what are target sources for organic tracer.

Response and Revisions: Thank you for your suggestion. We have already added the sentence as following.

Author's changes in manuscript: "In particular, anhydrosugars such as levoglucosan are used as a molecular marker to indicate biomass-burning emissions. The combination of ¹⁴C analysis and the concentration of levoglucosan has offered new insights into the detailed sources of carbonaceous aerosols. So, source apportionment of carbonaceous aerosols was performed using a source apportionment model based on the ¹⁴C results and measured chemicals."

Fig.1, I suggest to include annual or winter aerosol optical depth to display the representative of the 10 cities for air pollution hotspots in China. Alternative, a literature for PM_{2.5} map in China may be helpful to show the relative high levels of the 10 cities. An example can be found in figure 1 of a publication: Light absorption enhancement of black carbon from urban haze in Northern China winter, Environ. Pollut., 221, 418-426, doi: <http://dx.doi.org/10.1016/j.envpol.2016.12.004>.

Response and Revisions: Thank you for your suggestion. We have already added new figure 1 into the revised manuscript.

I am interesting on the thermal and FID signal of the EC isolation of radiocarbon analysis of this method. This method is similar to CTO-375, but different from SWISS-4 (i.e. Zhang et al.) and NIOSH870 protocols.

Response and Revisions: These methods utilize the difference in thermal stability between OC and EC, which is different from the method of SWISS-S using thermal-optical approach. C₁₄ signal in

the EC fraction in this method was performed by evaporation of OC in a muffle furnace at 375°C in air with reaction time of 4h. More detailed method development of ¹⁴C analysis of WINSOC and EC please see at <http://pubs.acs.org/doi/abs/10.1021/es401250k?journalCode=esthag> (Title: The use of levoglucosan and radiocarbon for source apportionment of PM_{2.5} carbonaceous aerosols at a background site in East China). In addition, detailed information of ¹⁴C analysis of WSOC, WINSOC and EC can be found at <http://pubs.acs.org/doi/abs/10.1021/es503102w> (Title: Source Apportionment Using Radiocarbon and Organic Tracers for PM_{2.5} Carbonaceous Aerosols in Guangzhou, South China: Contrasting Local- and Regional-Scale Haze Events).

Line 308: PM_{2.5}, OC and EC levels were highest in northern China, with maximum concentrations of 482 µg m⁻³, 75.9 µg m⁻³ and 19.3 µg m⁻³, respectively. Please show the detail site of these highest levels.

Response and Revisions: We have already added details in the revised manuscript. (line 329)

Author's changes in manuscript: "PM_{2.5} samples were collected continuously from 10 Chinese urban cities during early winter 2013. PM_{2.5}, OC and EC levels were highest in northern China, with maximum concentrations of 482 µg m⁻³(Taiyuan, n=31), 75.9 µg m⁻³(Taiyuan, n=31) and 19.3 µg m⁻³(Beijing, n=31), respectively."

Line 309: OC and EC were the major components of PM_{2.5}, accounting for 13 ± 8% and 2 ± 1%, of total PM_{2.5}, respectively. This is not suitable conclusion of this study. Author did not analyze several major chemicals such as sulfate, nitrate. I do agree that OC and EC are very important species of particulate matter, considering the health and climate impacts of the carbonaceous aerosols.

Response and Revisions: Thank you for pointing out this. The sentence has already been deleted.

Line 320: while SOC contributed more in cities in other regions of China. What is the meaning of other regions in China? Please specify the exact regions.

Response and Revisions: The sentence has already been revised into "while SOC contributed more in cities in other regions of China, such as Nanjing and Wuhan."

Line 321-322: however, the contribution of POC from both NF and NF increased significantly in these periods. This sentence should be corrected and improved.

Response and Revisions: This sentence has been changed into "During haze days, there were no dramatic changes in carbon sources or carbon compositions in the sampled cities, but the contributions of POC were relatively higher than the non-haze days."

Final sentence: This indicates that synoptic conditions promote the accumulation of particles derived either from local or regional sources. This is not an informative conclusion for the scope of this research.

Response and Revisions: Thank you for your suggestion. We have deleted the sentence.