Thank the reviewer for the time and effort on our manuscript. We appreciate the reviewer's instructive comments and suggestions. We have responded to these in the attached file, and included point-by-point responses to the reviewer's comments for consideration.

General Comment:

This paper investigated the source apportionments of carbonaceous aerosols collected in three northern cities in China using the measurements of carbon isotopes (^{13}C and ^{14}C) and levoglucosan. I understand the sampling campaigns and the measurements are not easy and the presented dataset should be informative. However, to be honest, I cannot find exciting things in this study and there are many technical problems needed to be addressed.

Response:

Thank you for your comments! The comments are very meaningful and useful. We have checked the relevant information in detail, carried out correction and reanalysis of the data. We corrected the Lev concentrations and δ^{13}C values of sample, and adjusted the end member of the parameter used in source apportionment. The correction in sample δ^{13}C caused a change about 4% for the results of source apportionment. After the correction of Lev concentrations and end member of parameter, different sources produced a change of 0.4-4.7%. These relatively small changes did not alter the main conclusions of our study. The detailed responses are shown as follows:

Specific Comment:

1. Line 32, Line 48 and Line 179: "stable carbon" → "stable carbon isotope"

Response:

Thanks for your suggestion! We have changed the corresponding words in the manuscript.

2. Line 68-70: Can you please simply tell readers why this tracer can accurately quantify the contribution of fossil and non-fossil sources?

Response:

Thank you for your suggestion! We have added simply explanation of the principle of ^{14}C in tracing non-fossil source in the manuscript:

"The natural radiocarbon (^{14}C) is completely depleted in fossil emissions due to the age of fossil fuels well above the half-life of ^{14}C (5730 years), whereas non-fossil sources show the similar ^{14}C as environment (Szidat, 2009; Heal, 2014)."

3. Line 74: add "a" between "useful" and "geochemical marker"

Response:

Thank you for your suggestion! We have changed the word in the manuscript.

4. Line 91: Can you provide the references to support "several decades"?

Response:

Thanks for your suggestion! We have added several references in the manuscript:

"Cities in northern China have been affected by severe haze for several decades (Cao et al., 2012; Han et al., 2016; Sun et al., 2006; Wang et al., 1990)."

5. Line 129-130: I don't think data from a TV news is appropriate for a scientific paper.

Response:

Thanks for your suggestion! We have updated the data and sources in the manuscript to make it more reliable:

"According to the pollutant data released by the National Air Quality Real-time Release Platform, Ministry of Ecology and Environment (MEE) of the People's Republic of China (http://106.37.208.233:20035/), the daily average atmospheric SO_2 concentration in LF exceeded 850 μg m^{-3} on one day in January 2017."

6. Fig.1 This map is boring and less informative. I suggest the authors to add the PM$_{2.5}$ concentration at the map background.

Response:

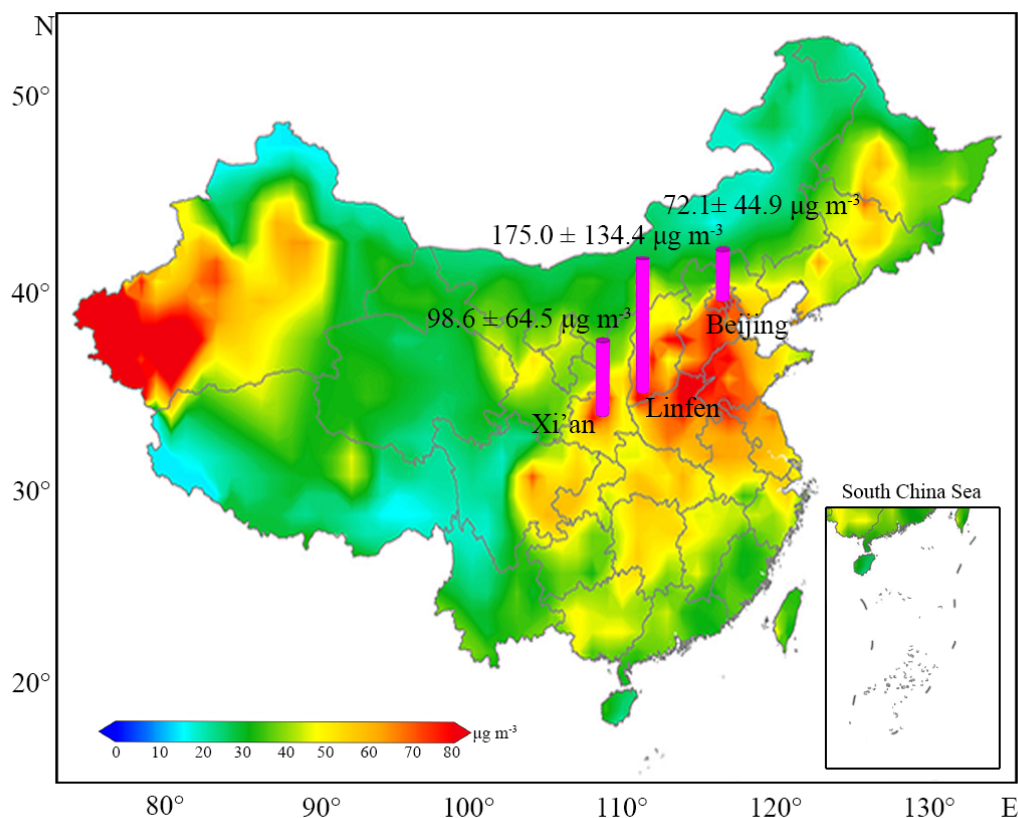Thank you for your suggestion! We have revised **Figure 1.**

Fig. 1 Locations and PM$_{2.5}$ concentration of Beijing (BJ), Xi'an (XA), and Linfen (LF). The background map shows the distribution of PM$_{2.5}$ concentrations in most of China from 2015 to 2019 (Li et al., 2021). The pink bars are the average PM$_{2.5}$ concentrations of the samples collected in this study during 2018 to 2019.

7. Line 147-150: So, how long for the sampling of each sample? 24 hours?

Response:

Yes, the samples were all collected continuously for 24 hours. We have added corresponding descriptions in the manuscript:

"A total of 124 24-hour (10 a.m. to 10 a.m. on the following day) PM$_{2.5}$ samples and 4 field blanks were obtained."

8. Line 179 Section 2.5: Please provide the corresponding reference regarding this ^{13}C analysis. Did you remove the carbonate fraction before ^{13}C analysis?

Response:

Thanks for your suggestion. We have added relevant references in the manuscript:

"The ^{13}C compositions were determined using a gas isotopic analyzer (Picarro G2131-i) in conjunction with an elemental analyzer (Elemental Combustion System 4010) at the Institute of Earth Environment, Chinese Academy of Sciences. Specifically, 0.2–0.4 mgC of sample has been placed in a precombusted tin capsule (6×10 mm) and the air was removed by squeezing. The samples were tested at 980 ℃ and 650 ℃ with 70–80 ml min^{-1} helium as the carrier gas and 20–30 ml min^{-1} oxygen as the reaction gas. The resulting gas mixture was then collected in Gas Isotopic Analyzer (Bachar et al., 2020)."

Yes, carbonate has been removed before testing by spraying with 1 mol L^{-1} hydrochloric acid. We have added this in the manuscript.

9. Line 193-194: How to remove?

Response:

Carbonate was removed by spraying with 1 mol L^{-1} HCl solution. We have adjusted the position of this description in the manuscript to indicate that all carbonate has been removed prior before measurement.

"Carbonate has been removed from the filters by spraying with hydrochloric acid (1 mol L^{-1}) before measurement."

10. Line 214-215: need to eliminate the influence of nuclear bomb excess. This correction is associated with the sampling time.

Response:

Thanks for your comment! We have noticed that and explained why the effect of nuclear bomb excess has been excluded in this study, which was shown in detail in the manuscript:

"Atmospheric nuclear bomb tests in the late 1950s and the early 1960s released a large amount of ^{14}C, and the ratio of $^{14}C/^{12}C$ in atmospheric CO_2 roughly doubled in the mid-1960s (Hua & Barbetti, 2004; Levin et al., 2003, 2010; Lewis et al., 2004; Niu et al., 2021). However, f_M in the atmosphere has been decreasing because of the dilution effect produced by the absorption of marine and terrestrial biospheres and the release of fossil fuels. In recent years, studies on background $^{14}CO_2$ in China and other countries have shown that the f_M value in the atmosphere has decreased and approached 1 (Hammer et al., 2017; Niu et al., 2016). This means that the impact of the nuclear explosions has almost disappeared for current atmosphere, and the change in current atmospheric ^{14}C was mainly influenced by the regional natural carbon cycle and fossil fuel CO_2 emissions. Thus, the f_M values were not corrected in this study, because the material used for biomass burning in China was mainly from crop straw (Fu et al., 2012; Street and Yarber, 2003; Yan et al., 2006; Zhang et al., 2017b), and the influence of atmospheric nuclear bomb test has basically vanished for the annual plants."

11. Line 224: This equation is questionable. A large part of OC in the air is formed by the oxidation of gaseous compounds, while some OC would be decomposed. It is impossible to make a source apportionment for OC or TC using the measurements of stable carbon isotope due to the unavoidable occurrence of isotopic fractionation.

Response:

Thanks for your comment! We have consulted the references on this issue.

The formation process of OC can cause the fractionation of ^{13}C, with a range mainly in 0.03–1.40‰ (mean 0.2‰) (Aggarwal and Kawamura, 2008; Cao et al, 2011; Ho et al., 2006; Zhao et al., 2018). Therefore, a small correction (0.2‰) was made for the $\delta^{13}C_{sample}$ before it used to estimate the contributions of coal and liquid fossil fuel combustion. The correction of $\delta^{13}C$ in TC was found to cause a relatively small change (<5%) in source apportionment. The description has been added in the manuscript:

" $f_{nf} \times \delta^{13}C_{nf} + f_{coal} \times \delta^{13}C_{coal} + f_{liq.fossil} \times \delta^{13}C_{liq.fossil} = \delta^{13}C_{sample} + \beta$ (6)

$$f_{\text{coal}} + f_{\text{liq.fossil}} = f_{\text{f}} \tag{7}$$

where f_{nf}, f_{coal}, and $f_{\text{liq.fossil}}$ represent the proportions of non-fossil source, coal and liquid fossil combustion, respectively, $\delta^{13}C_{\text{nf}}$, $\delta^{13}C_{\text{coal}}$, and $\delta^{13}C_{\text{liq.fossil}}$ represent $\delta^{13}C$ from the corresponding sources, $\delta^{13}C_{\text{sample}}$ is the $\delta^{13}C$ of the samples at each site, and β is a small correction.

Since the formation process of OC can cause the fractionation of ^{13}C, with a range mainly in 0.03–1.40‰ (mean 0.2‰) (Aggarwal and Kawamura, 2008; Cao et al, 2011; Ho et al., 2006; Zhao et al., 2018), a small correction (0.2‰) was made for the $\delta^{13}C$ sample used in Eq. 6."

New source apportionment results have been revised throughout the manuscript.

12. Line 239, Line 247: Two important issues need to be addressed when you try to employ Lev to calculate the contribution of biomass burning. 1) You need to eliminate the interference from the emission of non-biomass burning sources. Lev is not a unique tracer of biomass burning (Wu et al. 2021); 2) You need to consider the atmospheric degradation of Lev in the air (Li et al. 2021).

Response:

Thanks for your suggestions! We have reviewed the relevant references and corrected the concentration of Lev. The specific content is showed in the manuscript and **Figure S3**. The new source apportionment results have been changed in the text and figures throughout the manuscript.

"Recent studies indicated that Lev was degraded to some extent during atmospheric transportation, and about 25% of them came from other non-biomass burning sources (Hoffmann et al, 2010; Wu et al., 2021). Therefore, correction of the biomass burning source lev (Lev_{bb}) is required before the source apportionment:

$$Lev_{bb} = \frac{Lev \times 0.75}{p} \tag{1}$$

where p (0.4–0.65) is the degradation rate of Lev, which has different characteristics in each seasons. For specific p value in each season, please refer to the research of Li et al. (2021)."
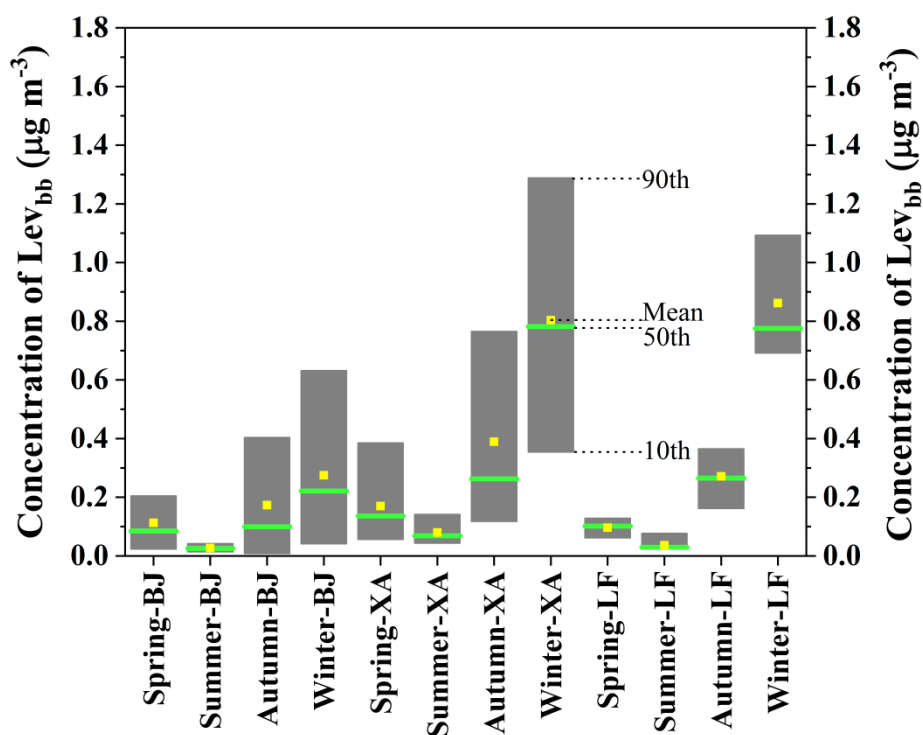
Fig. S3 Distribution of biomass-burning source Lev concentration in carbonaceous aerosols at the sampling sites in Beijing (BJ), Xi'an (XA) and Linfen (LF).

13. Line 297-309: The problem is that the wood for burning may be 20-year-old or ever older, which means that you still need to correct the effect of nuclear bomb test.

Response:

Thank you for your suggestion! According to the researches about biomass type, crop straw rather than wood are mainly used as biomass fuels in China (Street and Yarber, 2003; Yan et al., 2006). Therefore, the influence of nuclear bomb test can be ignored. The description has been added in the manuscript:

"According to the researches about biomass burning type, perennial biomass fuel was less frequently used in China (Fu et al., 2012; Street and Yarber, 2003; Yan et al., 2006; Zhang et al., 2017b), the impact of nuclear explosions on ^{14}C data can be ignored, and the $f_M(nf)$ and $f_M(bb)$ of the local station should be close to the atmospheric value."

14. Line 338-341: I would say the occurrence of biomass burning can lead to an

enhanced OC/EC ratio as well.

Response:

Thank you for your suggestion! We have reviewed the relevant researches and made a supplementary description in the manuscript:

"Additionally, the use of biomass fuels can also enhance the OC/EC ratio (Popovicheva et al., 2014; Rajput et al., 2011). Therefore, the high OC/EC ratio indicates that carbonaceous aerosols contained a large number of SOCs or biomass burning sources, especially in XA."

15. Fig.2. Please improve the quality of this figure. We can't see the variations. Please add a Yaxis for EC and readjust Y-axis scales.

Response:

Thanks for your suggestion! We have increased the axes for clearer viewing and increased the resolution of the **Figure 2**.
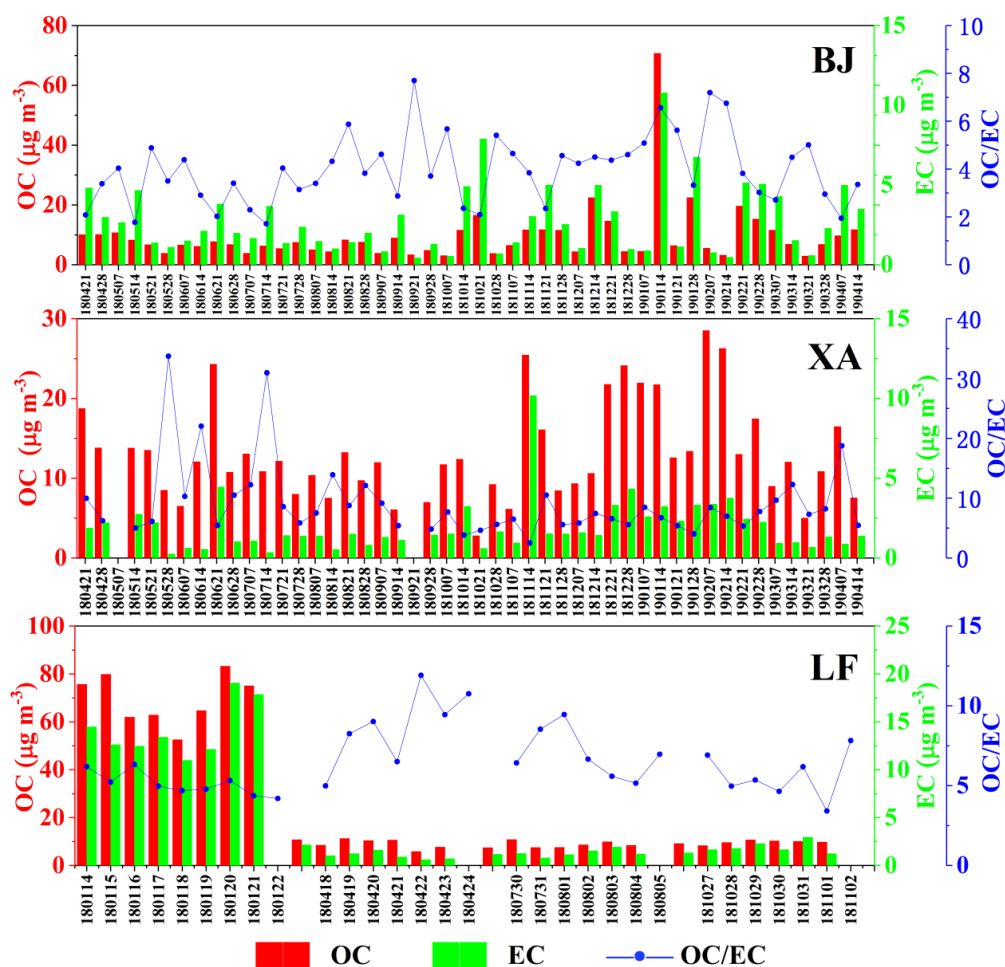
Fig. 2 Variations of elemental carbon (EC), organic carbon (OC) and their ratios in PM$_{2.5}$ at the sampling sites in Beijing (BJ), Xi'an (XA), and Linfen (LF) (date, "yymmdd").

16. Fig.5. I think this comparison is inappropriate because only few samples were performed for 14C measurements in some studies of this figure. For example, Huang et al. (2014) only analyzed 6 samples collected in the 2013 winter season, however, this paper analyzed ~ 50 samples for all seasons in Xian city.

Response:

Thanks for your comment! Due to the different sampling periods in studies, direct comparisons would be inappropriate. To eliminate this effect, we have changed the figure for a clear comparison of the same sampling periods. The results showed that the fossil sources in Beijing show a downward trend after the Action Plan for each season/period. The corresponding results were corrected in the manuscript.

"As can be seen in **Fig. 5**, the contribution of fossil sources in Beijing decreased by about 6-15% for the different sampling season/period after the implementation of Action Plan, based on previous studies (Fang et al., 2017; Lim et al., 2020; Liu et al., 2016a, b; Ni et al., 2018, 2020; Shao et al., 1996; Sun et al., 2012; Yang et al., 2005; Zhang et al., 2015, 2017a) and this study. Among them, fossil sources decreased significantly in autumn and winter after the Action Plan, which were 15% and 14%, respectively. The contribution of fossil sources in our study decreased by 16% in winter compared with the previous results. For the polluted and clean periods, the proportion of fossil sources reduced by 6% and 9%, respectively. With the implementation of energy conservation and emission reduction policies, many non-clean fossil fuels have been transformed into clean energy. In 2019, the coal consumption in BJ was only 1.3 million tons, which was 91.5% lower than that in 2013 (BJMBS, 2020).

Different from the results in Beijing, the proportion of fossil sources in Xi'an has not decreased significantly for each season/period (Fig. 5). This difference might be

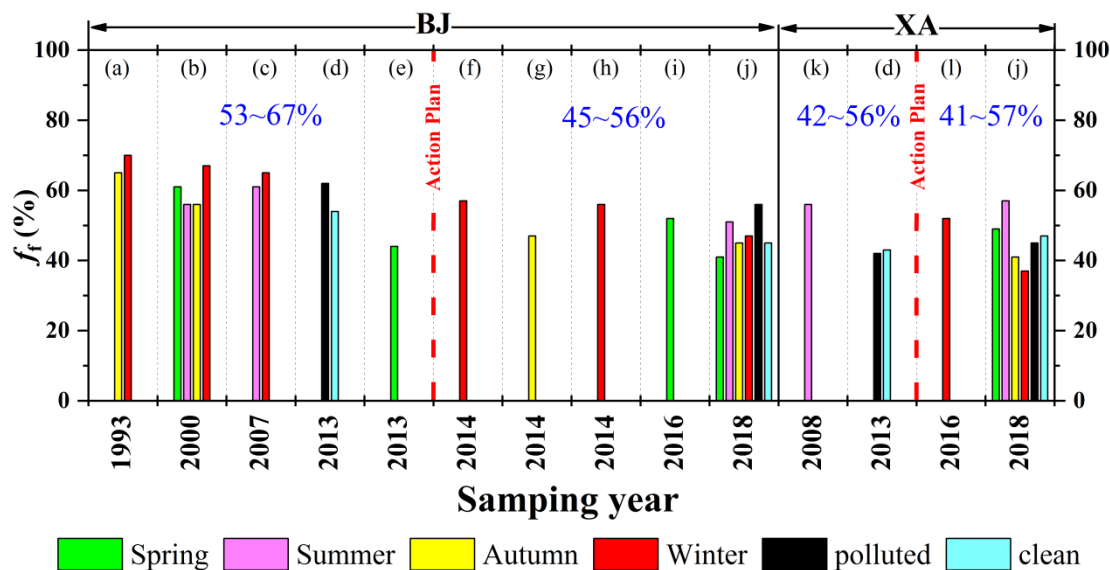related with a small decline (< 0.5%) in coal consumption in Xi'an during 2019 compared to 2013 (XAMBS, 2014, 2020)."

Fig. 5 Comparison of fossil proportion (f_f) of carbonaceous aerosol reported in different studies in Beijing (BJ) and Xi'an (XA), China for each season/period. The data has been converted to the ratio of total carbon. The ranges shown in the upper part of the figure are the average values of each season/period before and after the Action Plan. (a) Shao et al., 1996; (b) Yang et al., 2005; (c) Sun et al., 2012; (d) Zhang et al., 2015; (e) Liu et al., 2016a; (f) Zhang et al., 2017; (g) Liu et al., 2016b; (h) Fang et al, 2017; (i) Lim et al., 2020; (j)This study; (k) Ni et al., 2018; (l) Ni et al., 2021.

17 Line 537-538: The estimated contribution of biomass burning in EC for BJ is too low. I suggest the authors to compare this result with others using different methods (e.g., radiocarbon, bottom-up emission inventory, aethalometer). First, you need to consider the atmosphere degradation of Lev; Second, the ECbb/Lev end member used in this study probably is wrong.

Response:

Thank you for your comment! We have made a correction to the Lev after consulting the relevant references, and adjusted the parameter (Lev$_{bb}$/OC$_{bb}$, EC$_{bb}$/OC$_{bb}$) end member used in **Table 1** according to nearly research in northern China (Huang et al., 2014; Zhang et al., 2015). In the new source apportionment (**Fig.**

8), the contribution of EC_{bb} in Beijing was 1.9%, which is not much different from the previous result (1.4%).

Other related studies were combined with our results for comparison. In the research of Zhang et al. (2015) using radiocarbon and lev, the EC_{bb} contribution in Beijing was 3.5%. The study of Fang et al. (2019) using radiocarbon showed a contribution of EC_{bb} about 4.1%. Zhang et al. (2017a) used the AMS-PMF model along with radiocarbon and molecular tracer to analyze the source of particular matter in Beijing, and the results showed that EC_{bb} accounted for 4.9%. In the source apportionment by Zhang et al. (2017b) and Tao et al. (2016) using a combination of different molecular tracers and PMF model, the contribution of biomass burning to $PM_{2.5}$ in Beijing was around 7-16.5%, and we estimated that the contribution of EC_{bb} to TC was about 1.6-4.4%. Thus, our EC_{bb} result was slightly smaller than those in previous studies in Beijing, but the differences were not obvious. The low EC_{bb} contribution in Beijing in this study was mainly due to the low Lev concentration (Fig. S3) (0.15 ± 0.17 μg m^{-3}), while the Lev concentration in previous studies in Beijing were 0.20~1.26 μg m^{-3} (Tao et al., 2016; Zhang et al., 2015; Zhang et al., 2017b).

The relevant data and results have been changed throughout the manuscript.

Table 1. Values with limits of input parameters for source apportionment using Latin hypercube sampling (LHS).

Parameters	Low	Probable value	High
Lev_{bb}/OC_{bb}	0.01	0.11	0.20
EC_{bb}/OC_{bb}	0.13	0.22	0.31
$\delta^{13}C_{liq.fossil}$ (‰)	−28.00	−27.00	−25.00
$\delta^{13}C_{Coal}$ (‰)	−25.00	−22.95	−21.00
$\delta^{13}C_{nf}$ [a] (‰)	−26.00	−25.25	−24.00
$\delta^{13}C_{nf}$ [b] (‰)	−27.00	−26.50	−25.00

Agnihotri et al., 2011; Engling et al., 2006, 2009; Gelencsér et al., 2007; Huang et al., 2006; Lopez-Veneroni, 2009; Martinelli et al., 2002; Moura et al., 2008; Oros et al., 2001a, b; Puxbaumet et al., 2007; Smith & Epstein, 1971; Szidat et al., 2006; Turekian et al., 1998; Wang et al., 2009; Widory, 2006; Zhang et al., 2007.

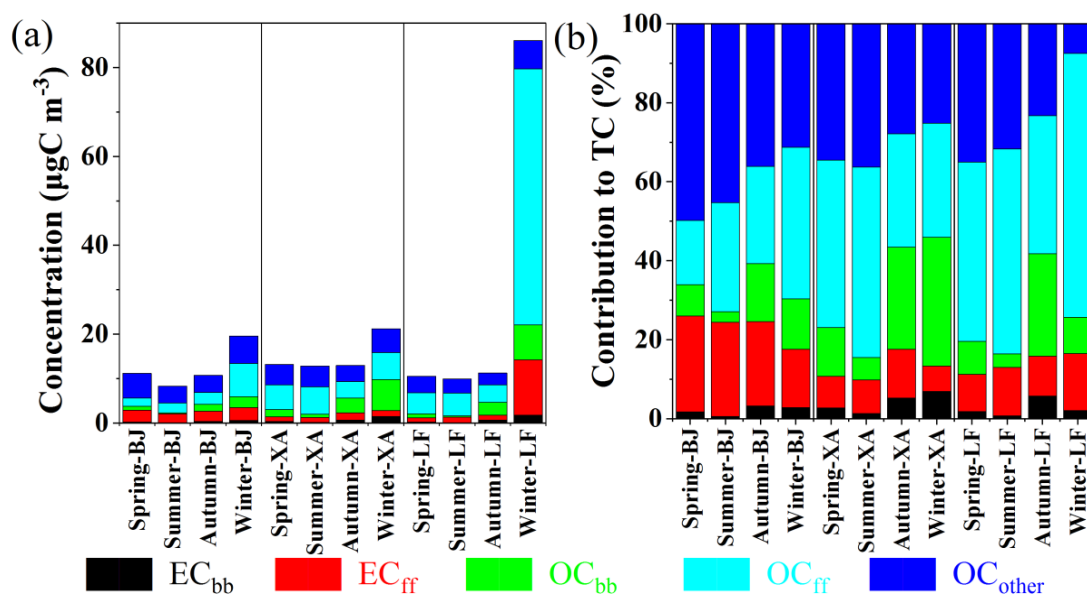[a] Values used in BJ/LF

[b] Values used in XA

Fig. 8 Mass concentrations ($\mu gC\ m^{-3}$) (a) and percentage (b) of elemental carbon from biomass burning (EC_{bb}) and fossil-fuel combustion (EC_{ff}), organic carbon from biomass burning (OC_{bb}), fossil-fuel combustion (OC_{ff}), and other sources (OC_{other}) for carbonaceous aerosols samples in Beijing (BJ), Xi'an (XA), and Linfen (LF) during different seasons.

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