Response to reviewer 2

General comments This paper reports the results of source apportionment based on a 1-year campaign in China. Besides the specific results, the paper presents an interesting methodology, based on the synergic use of radioactive and stable carbon isotopes. The paper ends with an open question, but this may be the trigger to foster new research. Therefore, I think the paper is worth the publication.

Response: We appreciate the reviewer's thoughtful and valuable comments, which are very helpful for revising and improving our manuscript. We have carefully addressed the reviewer's comments. Below are point-to-point responses.

Specific comments

1) Introduction, page 2 lines 16-18: "The ^{14}C content of an aerosol sample is usually reported relative to an oxalic acid standard and expressed as fraction modern ($F^{14}C$). ^{14}C content of the standard is related to the unperturbed atmosphere in the reference year of 1950 (Mook and van der Plicht, 1999; Reimer et al., 2004)"; please change to "The ^{14}C content of an aerosol sample is usually reported relative to an oxalic acid standard and expressed as fraction modern ($F^{14}C$). ^{14}C content of the standard is related to the unperturbed atmosphere in the reference year of 1950 (Mook and van der Plicht, 1999; Reimer et al., 2004), and this is usually done with/by means of/similar a standard"; this sentence is more correct and actually describes much better the definition formula in the following line.

Response: We agree with the reviewer, that this formulation is not entirely clear. Text and Eq. (1) is revised to clarify the definition of $F^{14}C$ as shown in the revised manuscript (marked-up copy, page 2, line 26–31; page 3, line 1–3). We tried to be even more specific and refer now concretely to OxII standard multiplied by 0.7459.

2) Introduction, page 2 lines 21-22: the assumptions on $F^{14}C$ are reported in a far too simplistic in several points (later on they become simple "conversion factors"). I suggest the authors to better introduce these quantities to facilitate the reader and also the writing of the following sections. The same for $\delta^{13}C$: it is introduced in the following sections, it is true, but at this stage there are already some sentences maybe not clear to readers not too familiar with stable carbon isotopes (e.g. "signature depleted", but it is not clear with respect with which reference.)

Response: We introduce the $F^{14}C$ of contemporary sources in details as follows (page 3 line 5–14):

> "However, $F^{14}C$ values of the contemporary (or non-fossil) carbon sources are bigger than 1 due to the nuclear bomb tests that nearly doubled the $^{14}CO_2$ in the atmosphere in the 1960s and 1970s. Currently, $F^{14}C$ of the atmospheric CO_2 is approximately 1.04 (Levin et al., 2010).This value is decreasing every year, because the $^{14}CO_2$ produced by bomb testing is taken up by oceans and the biosphere and diluted by ^{14}C-free CO_2 produced by fossil fuel burning . For biogenic aerosols, aerosols emitted from cooking as well as annual crop, the $F^{14}C$ is close to the value of current atmospheric CO_2. $F^{14}C$ of

wood burning is higher than that, because a significant fraction of carbon in the wood burned today was fixed during times when atmospheric $^{14}C/^{12}C$ ratios were substantially higher than today. Estimates of $F^{14}C$ for wood burning are based on tree-growth models (e.g., Lewis et al., 2004; Mohn et al., 2008) and found to range from 1.08 to 1.30 (Szidat et al., 2006; Genberg et al., 2011; Gilardoni et al., 2011; Minguillón et al., 2011; Dusek et al., 2013)."

The new citations are included in the revised reference list.

Text in the method section is revised to clarify the "conversion factor" by adding their physical meanings, as follows (page 7 line 11–13; line 18–22):

"$F^{14}C$ of EC ($F^{14}C_{(EC)}$) was converted to the fraction of biomass burning (f_{bb}(EC)) by dividing with $F^{14}C$ of biomass burning ($F^{14}C_{bb}$= 1.10 ± 0.05; Lewis et al., 2004; Mohn et al., 2008; Palstra and Meijer, 2014) given that biomass burning is the only non-fossil source of EC, to eliminate the effect from nuclear bomb tests in the 1960s." (page 7 line 11–13)

"$F^{14}C$ of OC ($F^{14}C_{(OC)}$) was converted to the fraction of non-fossil (f_{nf}(OC)) by dividing the $F^{14}C$ of non-fossil sources including both biogenic and biomass burning ($F^{14}C_{nf}$ =1.09 ± 0.05; Lewis et al., 2004; Levin et al., 2010; Y. Zhang et al., 2014a). The lower limit of 1.04 corresponds to current biospheric sources as the source of OC, the upper limit corresponds to burning of wood as the main source of OC, with only little input from annual crops." (page 7 line 18–22)

$F^{14}C_{bb}$ (1.10 ± 0.05) for EC is slightly smaller than $F^{14}C_{nf}$ (1.09 ± 0.05) for OC, because except biomass burning, biogenic emissions also contribute to OC, but have a smaller $F^{14}C$ than that of biomass burning.

For stable carbon isotopes, we change "the stable carbon isotope (^{13}C, expressed as $\delta^{13}C$)" to "the stable carbon isotope composition (namely the $^{13}C/^{12}C$ ratio, expressed as $\delta^{13}C$ in Eq. (2))" in the revised text (page 3 line 19–20). $\delta^{13}C$ is useful to distinguish sources of aerosols. The distinction is possible because for example, $\delta^{13}C$ values of carbon from coal combustion are less depleted (or enriched, i.e., enrichment in $^{13}C/^{12}C$ ratio) compared to the aerosol carbon emitted by other sources, for example, liquid fossil fuel combustion (-28 ‰ to -24 ‰) and C3 plants burning (-35 ‰ to -24 ‰) (Andersson et al. (2015) and references therein). The "signature depleted" is based on the comparison between different emission sources. To clarify, we explain the term "enriched" and "depleted" $\delta^{13}C$ as follows:

"Different emission sources have their own source signature: carbonaceous aerosol from coal combustion is enriched in ^{13}C (i.e., has higher $\delta^{13}C$ values of ~ -25 ‰ to -21 ‰) compared to aerosol from liquid fossil fuel combustion ($\delta^{13}C$ ~ -28 ‰ to -24 ‰) and from burning of C3 plants ($\delta^{13}C$ ~ -35 ‰ to -24 ‰) (Andersson et al. (2015) and references therein)." (page 3 line 21–25)

3) Introduction, page 3 line 7 and table S1: actually, source signatures are not that well distinct, as they have overlaps: may the authors discuss deeper this point?

Response: We appreciate this point. If it is ok with the reviewer, we will put the deeper discussion into the methods section 2.6, where we introduce the use of ^{13}C data for source apportionment and can further elaborate on the consequences of the overlapping signatures.

Though with some overlaps, different emission sources have their own source signature. To avoid misunderstanding, we change "Different emission sources have their ***distinct*** source signature" in the introduction section to "Different emission sources have their ***own*** source signature" (page 3 line 22).

Source signatures of $\delta^{13}C$ presented in Table S1 (shown as mean ± standard deviation) are established for the purpose of the source apportionment for EC (see revised Sect. 2.6). For burning C3 plants, coal and liquid fossil fuel, their $\delta^{13}C$ source signatures for EC are fully complied and established in Andersson et al. (2015) by a thorough literature search. In brief, the mean and standard deviation for $\delta^{13}C$ endmembers for the different sources are estimated as the average and standard deviation of the different data sets, respectively. For burning corn stalk residues in the study area, our $\delta^{13}C$ values of aerosols from corn stalk burning were complied in Fig. S6 and its source signatures are established as -16.4 ± 1.4 ‰ (mean ± standard deviation), as described in Sect 4.3.1 (Sect. 3.4.1 in the draft manuscript) and as addressed in Question 14. To clarify, we add ranges of source signatures of $\delta^{13}C$ into Supplemental Table S1, besides the established mean ± standard deviation for source apportionment of EC.

The source endmembers for $\delta^{13}C$ are less well-constrained than for $F^{14}C$, as $\delta^{13}C$ varies with fuel types and combustion conditions. For example, $\delta^{13}C$ values for liquid fossil fuel combustion overlaps with $\delta^{13}C$ values for both coal and C3 plant combustion. In this study, to account for the variability of the isotope signatures of $\delta^{13}C$ and $F^{14}C$ from the different sources, the Bayesian Markov chain Monte Carlo techniques (MCMC) were used as explained in the Method section. Uncertainties of both source endmembers for each source and the measured ambient $\delta^{13}C_{EC}$ and $F^{14}C_{(EC)}$ are propagated. These serve as input of MCMC to estimate the source contributions to EC. The MCMC results are the posterior probability density functions (PDF) for the relative contributions from the sources (Fig. S7, Fig. S8). The PDF of the relative source contributions of liquid fossil fuel combustion (vehicle) and coal combustion is skewed. By contrast, the PDF of the relative source contributions of biomass burning is symmetric as it is well-constrained by $F^{14}C$ (Fig. S8(a)). In this study, the median with its interquartile range was used to give the estimates of the contribution of any particular source to EC throughout the manuscript (e.g., Table 1, Sect. 4.3.2).

This point is clarified in the Method Sect. 2.6 by adding the following (page 8 line 14-22):

> "$\delta^{13}C_{bb}$, $\delta^{13}C_{liq.fossil}$ and $\delta^{13}C_{coal}$ are the $\delta^{13}C$ signature emitted from biomass burning, liquid fossil fuel combustion and coal combustion, respectively. The means and the standard deviations for $\delta^{13}C_{bb}$ (-26.7 ± 1.8 ‰ for C3 plants, and -16.4 ± 1.4 ‰ for corn stalk), $\delta^{13}C_{liq.fossil}$ (-25.5 ± 1.3 ‰) and $\delta^{13}C_{coal}$ (-23.4 ± 1.3 ‰) are presented in Table S1

(Andersson et al., 2015 and reference therein; Sect. 4.3.1), and serves as input of MCMC. The source endmembers for $\delta^{13}C$ are less well-constrained than for $F^{14}C$, as $\delta^{13}C$ varies with fuel types and combustion conditions. For example, the range of possible $\delta^{13}C$ values for liquid fossil fuel combustion overlaps to a small extent with the range for coal combustion, although liquid fossil fuels are usually more depleted than coal. The MCMC technique takes into account the variability in the source-signatures of $F^{14}C$ and $\delta^{13}C$ (Table S1), where $\delta^{13}C$ introduces a larger uncertainty than $F^{14}C$. Uncertainties of both source endmembers for each source and the measured ambient $\delta^{13}C_{EC}$ and $F^{14}C_{(EC)}$ are propagated."

Further, to give the readers an idea of this point, we also changed the sentence in the Introduction section "Further, $\delta^{13}C$ of EC allows separation of fossil sources into coal and liquid fossil fuel burning (Andersson et al., 2015; Winiger et al., 2015, 2016), as they have their distinct source signatures" to:

"Further, $\delta^{13}C$ of EC allows separation of fossil sources into coal and liquid fossil fuel burning (Andersson et al., 2015; Winiger et al., 2015, 2016), due to their different source signatures. Typical $\delta^{13}C$ values for EC from previous studies are summarized in Table S1." (page 3 line 30–32).

4) Sampling, page 3 line 28: why was the sampling time chosen to be 10 am to 10 am next day? Due to manual change? How long were the samples kept inside the sampler after sampling?

Response: The sampling time was long chosen to be 10:00 a.m. to 10:00 a.m. the next day (e.g., Cao et al., 2009; Han et al., 2016) due to manual change and safety reasons in accessing the site at midnight.

Only one filter can be loaded into the sampler, so we took the filter out of the sampler after 24 hr sampling and did not keep it in the sampler. The revised text (page 5 line 1) shows that:

"After sampling, we immediately removed the filter from the sampler. All filters were packed in pre-baked aluminum foils, sealed in polyethylene bags and stored at -18 °C in a freezer"

5) Sampling, page 4 line 4: citations missing ("previous studies" are not cited)

Response: Citations are added on page 5 line 2–3.

6) Stable carbon isotope ($\delta^{13}C$) analysis of OC and EC, page 4: some more details on the analysis would be welcome. Further, the title is maybe misleading, as it suggests that only ^{13}C is measured, while I guess that also ^{12}C is assessed for determining $^{13}C/^{12}C$ ratios.

Response: More details on the $\delta^{13}C$ analysis are added as shown in the revised manuscript (page 5 line 21–30; page 6 line 1–7).

The title is changed to "2.3 Stable carbon isotopic composition of OC and EC", to indicate that $^{13}C/^{12}C$ ratios are determined, not only ^{13}C. (page 5 line 20)

7) Radiocarbon (^{14}C) measurement of OC and EC, page 5, line 9: "Two standards with known ^{14}C content are analyzed as quality control: an oxalic acid standard and a graphite standard.": maybe I did not understand, but I believe these standards are respectively for normalization and blank evaluation; if this is correct, they cannot be defined as "for quality control". In case further standards are measured as unknown, these can be defined "for quality control".

Response: Two standards with known ^{14}C content that extracted using our aerosol combustion system (ACS) are analyzed to assess the contamination introduced by the combustion process, and they are treated exactly like the samples (e.g., normalized to the oxalic acid OXII calibration material). The measured deviation in $F^{14}C$ from the nominal values is caused by contamination introduced by the combustion process. The contamination is assessed but not used for further data correction, because the contamination is relatively small (typically below 1.5 µgC per combustion) compared the sample sizes (ranging between 50 and 270 µgC). To clarify, the revised texts show:

> "Two standards with known ^{14}C content are combusted as quality control for the combustion process: an oxalic acid standard and a graphite standard. Small amounts of solid standard material are directly put on the filter holder of the combustion tube and heated at 650 °C for 10 min. In the further ^{14}C analysis, the CO_2 derived from combustion of the standards is treated exactly like the samples. Therefore, the contamination introduced by the combustion process can be estimated from the deviation of measured values (Table S9) from the nominal values of the standards. The contamination is below 1.5 µgC per combustion, which is relatively small compared the samples ranging between 50 and 270 µgC in this study." (page 6, line 19–24)

8) Radiocarbon (^{14}C) measurement of OC and EC, page 5, lines 11-13 and 24-25: there is a repetition of the information, and actually not completely in the same way: please correct it. Further, is this contamination modern or fossil?

Response: ^{14}C measurements of aerosol samples are subject to contaminations, which can be introduced during the combustion process using ACS, or during the graphitization and AMS measurements. For contamination caused by the combustion process, it is already explained in the response to Question 7. Here we addressed the contamination during the graphitization and AMS measurement, thus it is not a repetition of the information.

$F^{14}C$ of aerosols samples was corrected for contamination that occurred during graphitization and AMS measurement. For AMS measurements, samples are usually analyzed together with varying amounts of reference material covering the range of sample mass. Two such materials with known ^{14}C content are used: the oxalic acid OXII calibration material ($F^{14}C = 1.3406$) and a ^{14}C-free CO_2 gas ($F^{14}C = 0$). Contamination during the graphitization and AMS measurement results into the differences between measured and nominal $F^{14}C$ values. The magnitude of these deviations can be used to quantify the contamination with fossil carbon ($F^{14}C_F = 0$) and modern carbon ($F^{14}C_M = 1$), which in turn are used for correcting the sample values (de Rooij et al., 2010; Dusek et al., 2014).

The contamination with fossil carbon and modern carbon is quantified using isotope mass balance (Dusek et al., 2014):

$$F^{14}C_m \cdot M_m = F^{14}C_{st} \cdot M_{st} + F^{14}C_F \cdot M_F + F^{14}C_M \cdot M_M. \quad (S1)$$

M_m and M_{st} stand for the experimentally determined mass and the mass of reference materials either the oxalic acid OXII calibration material ($F^{14}C$ = 1.3406) or a ^{14}C-free CO_2 gas ($F^{14}C$ = 0) with a unit of µgC, respectively. $F^{14}C_m$ and $F^{14}C_{st}$ represent the experimentally determined $F^{14}C$ measured by AMS and nominal $F^{14}C$ of reference materials (Table S9).

The relationships among all masses are described as Eq. (S2):

$$M_m = M_{st} + M_F + M_M, \quad (S2)$$

where M_M is calculated using Eq. (S1) by substituting $F^{14}C_{st} = 0$ for a ^{14}C-free CO_2 gas as:

$$M_M = F^{14}C_m \cdot M_m. \quad (S3)$$

Substitute $F^{14}C_{st} = 1.3406$ for OXII and the derived M_M from Eq. (S3), M_F is derived by combining Eq. (S1) and Eq. (S2) as:

$$M_F = ((1.3406 - F^{14}C_m) \cdot M_m - (1.3406 - 1) \cdot M_M)/1.3406. \quad (S4)$$

M_M and M_F is calculated by applying Eq. (S3) and Eq. (S4), and they are mass dependent. The modern carbon contamination (M_M) is between 0.35 and 0.50 µg C, and the fossil carbon contamination (M_F) is around 2 µg C for sample bigger than 100 µgC.

In the revised manuscript, we add the detailed calculation of modern and fossil contamination in the supplemental material (Supplemental S3). The revised manuscript adds:

> "The differences between measured and nominal $F^{14}C$ values are used to correct the sample values (de Rooij et al., 2010; Dusek et al., 2014) for contamination during graphitization and AMS measurement (Supplemental S3). The modern carbon contamination is between 0.35 and 0.50 µg C, and the fossil carbon contamination is around 2 µg C for sample bigger than 100 µgC." (page 7 line 5–9)

9) Source apportionment methodology using ^{14}C: as already aforementioned, the use of the definition "conversion factors" is misleading, as they have a physical meaning (as it is clear at page 6, line 24). Authors should introduce this concept earlier in the text, so that they can also explain the use of different values for their "conversion factors". This would definitely make the paper easier to read.

Response: As addressed in the response to Question 2, the revised manuscript uses the physical meanings (i.e., $F^{14}C$ of biomass burning ($F^{14}C_{bb}$) for EC, $F^{14}C$ of non-fossil sources ($F^{14}C_{nf}$) for OC) instead of "conversion factors" (page 7 line 11–13, line 18–22). This concept is added in the revised Introduction section (page 3 line 5–13).

10) Temporal variation of fossil and non-fossil fractions of OC and EC, page 7: levoglucosan, hopanes and picene are cited for the first time, with no reference to S2, where the measurements

are described. The existence of ancillary/additional measurements deserves to be introduced as part of the methodology.

Response: We add briefly the measurement of source markers in the Sect. 2.2 as part of the methodology and change the title to "2.2 Organic carbon (OC), elemental carbon (EC) and source markers measurement". S2 is kept in the supplemental material for details on the measurements.

Further, S2 is mentioned (page 9 line 17) when organic markers (levoglucosan, hopanes and picene) are cited in the main text for the first time.

11) ^{13}C signature of OC and EC, page 9, line 12: "$\delta^{13}C_{OC}$ was in general similar to $\delta^{13}C_{EC}$": this means that the biogenic source is roughly negligible: can the author comment with finding also in relation to the radiocarbon measurement results?

Response: $\delta^{13}C_{OC}$ was in general similar to $\delta^{13}C_{EC}$: it suggests that biogenic OC is probably not very important, as could be expected from the high TC concentrations and strong anthropogenic sources. This can be true, as we would expect a bit different $\delta^{13}C_{OC}$ from $\delta^{13}C_{EC}$ if biogenic sources play an important role on OC.

In light of ^{14}C, we still measure a considerable fraction of non-fossil OC, and it would seem that this is more related to the biomass burning. Or, if there is biogenic OC, but by chance their $\delta^{13}C$ signatures are relatively similar with those for the source mixture of EC, which is not very likely.

The following statements were added to the revised manuscript:

> "$\delta^{13}C_{OC}$ was in general similar to $\delta^{13}C_{EC}$. This suggests that biogenic OC is probably not very important, as could be expected from the high TC concentrations. ^{14}C analysis indicates a considerable fraction of non-fossil OC than non-fossil EC, and it would seem that this is mainly related to the biomass burning, which has higher OC/EC ratios than fossil fuel burning. If the contribution of biogenic OC plays an important role, then the biogenic $\delta^{13}C$ signatures should be relatively similar to the source mixture of EC, which is rather unlikely, especially as this source mixture is not constant" (page 11 line 5–9)

12) I suggest moving section 4.4 straight after 4.2, as this discussion follows directly from the last sentences of 4.2.

Response: The order of the two sections has been changed. The order of Fig. 7 and Fig. 8 is also changed accordingly.

13) Changes in emission sources in Xi'an, China (2008/2009 vs. 2012/2013), pages 15-16: the cited papers taken for comparison focus, respectively, on a big haze episode and on an intensive campaign (2 winter months), and not on a campaign aiming at being representative for a year, therefore I think this comparison is not very useful. Further, contributions are roughly the same within the uncertainties.

Response: We did the whole year measurement in Xi'an for the year 2008/2009. In this study, we selected samples with varying $PM_{2.5}$ mass and carbonaceous aerosols for ^{14}C analysis. The

selected samples cover periods of low, medium and high $PM_{2.5}$ concentrations to get samples representative of the various pollution conditions that did occur in each season. Here we only compare the winter season due to the limited source apportionment results for EC in Xi'an. For Xi'an, we see from this study (see Fig. 1(a), except the Chinese New Year eve, which is not included in the comparison) but also some studies in preparation that the ^{14}C values do not change very much between polluted days and clean days. In addition, two months (the intensive campaign by Wang et al. (2016)) almost cover the whole winter (in total, 3 months). Thus, we think that it makes sense to compare the results from this study with the two cited paper.

For EC source apportionment, it is noted that the quartile ranges for 2008/2009 values (this study) overlaps ranges for 2012/2013 values (average ± SD). Compared to the uncertainties of radiocarbon measurements, the uncertainties of PMF results are always larger, making the overlapped ranges very likely. However, comparing the probability distribution functions for both cases give a more complete picture. Figure S14 and Figure S15 shows the probability density functions (PDF) of the relative source contributions to EC from coal combustion and vehicle emissions, respectively. Results from this study for the year 2008/2009 are shown in grey (this is also shown in the original Fig. S8), and from Wang et al. (2016) for the year 2012/2014 shown in yellow. For the PDF by Wang et al. (2016), we assume normal distribution as their source apportionment results are not known and given in the form of average ± SD. As shown in Fig. S14 and Fig. S15, though with some overlaps, the PDF of the relative source contribution of coal combustion (vehicle emissions) does clearly shift to the lower side (higher side) from the year 2008/2009 to 2012/2013. With the current inherent uncertainties in these two states of the art source apportionment methods it will not be possible to draw more firm conclusions than that these probability distributions show a certain trend, despite some possible overlap.

We also have some additional observation data to support the conclusion as discussed in Sect. 4.6. The decreased contributions of coal combustion are also evidenced from the declined enrichment factor of As and Pb, indicators of coal combustion. Increasing vehicular emissions is supported by the increasing level of NO_2, an indicator for the contribution of vehicular emissions.

In the revised manuscript, the above discussion is added in the section of Changes in emission sources in Xi'an, China (2008/2009 vs. 2012/2013) (page 18 line 25–27; page 19, line 7–14). The Fig. S14 and Fig. S15 is added to the supplemental material.

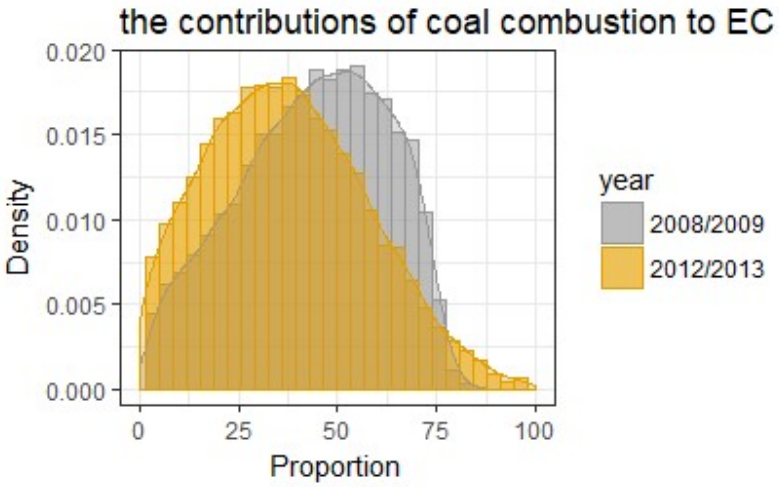

Figure S14. Probability density functions (PDF) of the relative source contributions of coal combustion to EC in winter in the year 2008/2009 (this study, shown in grey; this is also shown in Fig. S8) and 2012/2013 by Wang et al. (2016), shown in yellow.

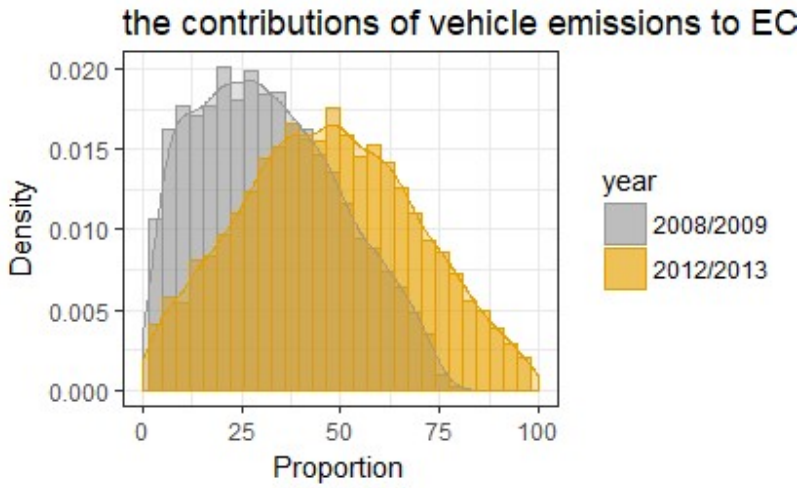

Figure S15. Probability density functions (PDF) of the relative source contributions of vehicle emissions to EC in winter in the year 2008/2009 (this study, shown in grey; this is also shown in Fig. S8.) and 2012/2013 by Wang et al. (2016), shown in yellow

14) Supplement, table S1: far as I get, the reported interval for C4 plants is wide as different plants (corn, sugar cane, grass and maybe more) have different signatures: why do the authors "decrease" this range to -16.4 ± 1.4 per mil? (Further, please pay attention to number of digits, e.g. -23.4 ±1.3 and not -23.38 ±1.3)

Response: In this study, $\delta^{13}C$ for corn stalk is used as it is the dominant C4 plant in Xi'an and its surrounding areas (Sun et al., 2017; Zhu et al., 2017), with little sugarcane and other C4 plants as explained in Sect. 4.3.1 where details on selection of $\delta^{13}C$ endmembers for C4 plants in the study area are described. $\delta^{13}C$ values of aerosols from corn stalk burning were compiled from literature, ranging from -19.3 ‰ to -13.6 ‰ (Fig. S6). $\delta^{13}C$ source signatures for emissions from burning corn stalk were determined as -16.4 ± 1.4 ‰ (mean ± standard deviation): the mean (-16.4 ‰) was computed as the average of the different data sets, and standard deviation (1.4 ‰) analogously calculated.

In the revised manuscript, we change the title of Sect. 4.3.1 to "4.3.1 Selection of $\delta^{13}C$ endmembers for aerosols from corn stalk burning in the study area", to clarify that the $\delta^{13}C$=-16.4 ± 1.4 ‰ is specific for burning corn stalk, which is a subtype of C4 plant. Further, in the notation of Table S1, we add the following as a reminder:

> "In this study, $\delta^{13}C$ for corn stalk is used as it is the dominant C4 plant in Xi'an and its surrounding areas (Sun et al., 2017; Zhu et al., 2017), with little sugarcane and other C4 plants."

Number of digits are all corrected for $\delta^{13}C$ values throughout the manuscript, according the reviewer's comments.

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