The manuscript describes a novel approach of radiocarbon source apportionment, which investigates the contributions of fossil vs. non-fossil emissions to the more volatile organic carbon (mvOC) fraction that evaporates in helium at 200°C. This approach shows for particulate matter samples from different cities in China that the fossil impact on mvOC is larger than for total OC and secondary OC. This new insight has implications on a better understanding of sources of carbonaceous aerosols, which is currently a hot topic in atmospheric chemistry and physics. Therefore, I recommend accepting the manuscript after minor revisions.

Main comments:

1. P4, L1-2, P12, L18-19, P18, L10-11: In previous work (Agrios et al., 2017; https://doi.org/10.1017/RDC.2016.88), we established a continuous-flow coupling of the Sunset OC/EC analyzer with the MICADAS and measured ^{14}C online for low-temperature OC steps and even monitored the change of the ^{14}C signal during the temperature ramp. We also found a more fossil signal for the 200°C step than for the following OC steps using higher temperature. We furthermore observed that even a shift from fossil to non-fossil emissions occurred within the 200°C peak for some samples, which indicates that the fossil character of mvOC would probably have been even more pronounced, if Ni et al. had chosen a temperature lower than 200°C.
2. P10, L22-24: Fig. 1b reveals that low recoveries are associated with a bias in $F^{14}C$(mvOC). Therefore, the correction of sample winter-L should address this bias accordingly by subtraction. A simple increase of the uncertainty is not appropriate.
3. P11, L1-2 and P12, L8-11: In order to identify reasons for differences between cities and individual samples, meteorological data should be shown in the Supplement.
4. P13, L16-18: The fact that the correlation between f_{nf}(mvOC) and f_{nf}(OC) is better than the correlation between f_{nf}(mvOC) and f_{bb}(EC) is mainly caused by the comparison of different fractions of the carbonaceous aerosol: in the former case, two OC fractions are compared (i.e. mvOC and OC), whereas in the latter case, OC and EC are compared. EC_{bb} may be transferred into POC_{bb} (see Eq. 8), but one has to take into account that the large variability of rbb contributes to the r2 value of the correlation between f_{nf}(mvOC) and f_{bb}(EC) in Fig. 5. (The uncertainty of r_{bb} was estimated to be 25%; see P8, L12.) Therefore, the suggestion of the importance of secondary formation of mvOC and/or other non-fossil contribution to mvOC besides primary biomass burning is not valid. This sentence should be removed. Consequently, the corresponding sentence P18, L30 to P19, L2 should also be deleted.
5. P15, L1-14: The authors try to draw conclusions from different r^2 values of correlations between $mvOC_{nf}$ with POC_{bb} and with SOC_{nf} (Fig. 7b). However, the statistical difference of both r^2 values was not proven by a proper test (e.g. an F-test). Furthermore, the high uncertainties of POC_{bb} (see my comment to P13, L16-18) and SOC_{nf} (which are indicated in Fig. 6) are also not considered for this discussion. As these important factors were not taken into account, the whole passage (P15, L1-14) should be removed.

Technical comments:

6. P1, L20: Better use the following phrasing: (range: 7 %–25 %)
7. P2, L4-7: The focus of this sentence should be changed, because a) PAHs are only a minor fraction of OC and b) EC is carcinogenic as well. I suggest characterizing OC and EC very broadly without mentioning health effects or special substance classes.
8. P2, L32: A comma is missing before “can”
9. P3, L1-3: Examples of high-volatility BBOA components should be given, as these may be relevant for the mvOC fraction.
10. P4, L10-11: pre-baked

11. P5, L29-30: Even though the blank is small compared to the sample amount, a blank correction has to be performed for both mvOC concentrations and their $F^{14}C$ values. If a direct analysis of the $F^{14}C$ of the blank hasn't been performed, a value of 0.50±0.29 should be applied to cover the full $F^{14}C$ range from 0 to 1 based on the assumption that a continuous uniform distribution (i.e. a rectangular distribution) is valid.
12. P6, L7-8: Here, the same applies as for P5, L29-30.
13. P6, L22: "and 1970s" should be deleted.
14. P7-9, Chapter 2.4: In the explanation of the calculation "can be" should be substituted by "was" several times.
15. P7, L26-27: A reference should be shown for the statement that the contributions from plant detritus, bioaerosols and spores to $PM_{2.5}$ are likely small.
16. P10, L14: "the" was erroneously repeated at the beginning of the line.
17. P10, L20: "Taken together" should be removed.
18. P13, L7: I suggest to begin the sentence with "As f_{nf}(mvOC) is smaller"
19. P13, L14: The citation "(Fig. 4b)" should be moved to the end of the sentence in L10.
20. P15, L19: we conclude (remove "can")
21. P15, L20-21: In Fig. 7c there are two outlier data points from sample
22. P15, L30: In other words
23. P18, L3: Consequently, our conclusion
24. P18, L30: References to the literature (Masalaite et al., 2017, 2018) and figures (Fig. 5) from the paper should be removed from the Conclusions.
25. Fig. 2: An uncertainty of the average $F^{14}C$(mvOC) should be given in line 6 using the standard deviation of the three replicates.
26. Fig. 3: The following sentence should be added to the caption: "For details see Tab. S4."
27. Supplement PS4, second to last line: $OC_{280°C}$ (instead of $OC_{2800°C}$)
28. Fig. S5, last line of the caption: The panels (a) and (b) have
29. Table S4: Uncertainties are missing for $d^{13}C_{EC}$ (last column)