Summary: This manuscript describes measurements of polar organic species in Nanchange, China and source apportionment of organic carbon (OC) using tracer based and chemical mass balance (CMB) approaches. The approaches taken by the authors are relatively standard, and the integration of multiple approaches strengthens the paper. The paper reports insights to annual variations in primary and secondary sources. Further insight is provided into the characteristics of OC during winter pollution episodes, when coal burning and secondary aerosol had relatively larger impacts. The figures are very detailed and contain a lot of information. There are numerous aspects of the manuscript, detailed below, that should be addressed prior to publication.

Overall comments that should be addressed throughout the manuscript:

- 1. In applying the "tracer-based" method to source apportionment (page 5), the results are highly dependent on the source profiles utilized. It is best practice to use locally-sourced profiles, when available, and those that are representative of the relevant sources at the time of the study. There is no justification provided for the selection of profiles beyond that they were utilized by Kleindienst et al. in their 2007 and 2012 publications. The authors must provide justification for the selected profiles, discuss their representativeness, and the potential errors introduced by these selections. Additionally, the authors should make a diligent effort to utilize the most relevant, regionally-specific, and up-to-date information when available, noting that the profiles used in these studies can be 20 years old. For example, regional profiles for straw burning should be used, given the conclusions of the authors of the importance of this source (10.1016/s1001-0742(07)60027-8). Regional profiles for other relevant sources are available and should be considered for robust results.
- 2. In applying the CMB approach to source apportionment, there is likewise a need for discussion of the selected profiles, their representativeness, and uncertainties introduced by differences in these profiles and local and/or regional sources.
- 3. The authors should also specifically state which chemical species were used in the CMB model and provide justification for these choices and discussion of what sources are and are not represented. The extraordinarily good agreement between the tracer-based and CMB results implies that only a few fitting species may have been used, which means that the CMB model may not be well-constrained.
- 4. Improve readability to integrate results from multiple tracers, rather than treating them one by one (i.e. pages X to Y). For example, integrating results from
- 5. There is a sizable portion of OC that is not attributed to the primary and secondary sources considered. This requires further discussion is it due to a mismatch of the selected tracer-to-OC fractions (or source profiles) to the ambient data? Are

- important sources in the region not included or considered? If a major secondary organic aerosol source has not been considered, then statements regarding the dominance of primary over secondary sources are not accurate.
- 6. The authors use linear correlations as a tool for data analysis, which assumes that date are normally distributed. Are the data, in fact, normally distributed? Please perform a statistical test for normality and include that result in the discussion.
- 7. In reporting organic species concentrations (i.e. lines 191, 192, and elsewhere), consider the appropriate number of significant figures. Likely 2-3 digits are statistically significant (considering uncertainties in the range of 10%) and should be reported, rather than 5.
- 8. Application of the "tracer-based" method and CMB approaches to source apportionment assume conservation of mass between source and receptor. However, the fatty acid results indicate that "unsaturated FAs underwent significant photochemical degradation" (line 220). Significant chemical transformations would lead to errors in source attribution. The influence of chemical transformations on source apportionment must be discussed.
- 9. A more thorough discussion of the limitations of the current work are needed.

## Specific comments:

- 10. It would be helpful if the authors could clarify in the abstract (lines 15-16) their approach to "comprehensive analysis" of polar compounds and source attribution to primary/secondary and anthropogenic/natural sources.
- 11. At lines 92-94, indicate the specific locations of the meteorological and gas sensors and their relation to the PM sampling site.
- 12. Lines 99-100, include a reference to the IMPROVE protocol used for EC and OC analysis.
- 13. Line 106, justification is needed for the use of n-alkanes as internal standards for polar compounds, especially because the alkanes do not undergo silylation derivitazation.
- 14. The statements about OC/EC values from 172-176 do not seem to consider secondary sources, or mixtures of sources. This seems contradictory with the other results in the study and should be removed.
- 15. Line 268, the authors should also consider reports of chemical degradation of levoglucosan in the atmosphere, for example, and how this may influence their source apportionment results.
- 16. In section 3.4.1, the authors report concentrations of C5 alkene triols. The authors should consider more up to date information available in the literature regarding this group of compounds (i.e. Frauenheim, et al. doi/10.1021/acs.estlett.2c00548).

- The majority of these "triols" have been demonstrated to be artifacts, and a structure with a ring rather than a double bond is major isomer.
- 17. Figure 5, S6, F7. The text in the legends is very small and difficult to read. The important information that distinguishes the various sources is sub-scripted and difficult to see. Please enlarge the text in the legend to improve readability.
- 18. Lines 527-528, please explain how OC was converted into OA.