Response to the reviewers' comments point-by-point

Reviewer 3:

General comments. This manuscript describes measurements of polar organic species in Nanchang, 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:

Overall comments 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.

Response: We thank the reviewer for the careful and constructive comment on the selection and representativeness of source profiles and tracers. In response, we have made targeted revisions. Both source-apportionment methods applied in this study—the CMB model and the tracer-based approach—depend critically on the choice and interpretation of source profiles and representative chemical species. Recognizing that the original manuscript provided limited detail on this point, we have substantially expanded and clarified the relevant material in the revised manuscript

First, we expanded Section 2.3 (Source apportionment methods) to provide a detailed description of the source profiles and representative tracers used in both the CBM model and tracer-based approaches. Where available, we prioritized regional and local profiles that best match the study area and observation period. Specific revisions are as follows:

"To quantify the contributions of various primary sources to OC and $PM_{2.5}$, i.e., the POC and POA, we utilized the CMB model (version 8.2), a widely accepted source apportionment method developed by the United States Environmental Protection Agency (Lewandowski et al., 2008;

Stone et al., 2009; Guo et al., 2012; Wu et al., 2020; Xu et al., 2021). The model assumes that the chemical composition of pollutants remains unchanged during transport, allowing measured chemical species at the receptor to be expressed as the linear sum of contributions from individual source categories. Accurate CMB results depend critically on the selection of representative emission sources and chemical tracers. These must include all major contributors and feature chemical markers that are stable during transport and distinct among sources (Stone et al., 2009). To meet these requirements, we first identified four major primary OC sources: biomass burning, coal combustion, vehicular exhaust, and cooking emissions, based on China's atmospheric PM and gaseous pollutant emission inventories (Huang et al., 2015; Li et al., 2019; Tong et al., 2020), We also considered the contributions of plant debris and fungal spores to OC, base on the regional studies on OC sources (Fan et al., 2020; Xu et al., 2020; Wu et al., 2020). Representative source profiles and tracers of these sources were compiled from recent local and regional reports. For example, biomass burning releases substantial amounts of levoglucosan, accounting for ~8.3% of OC in local profiles (Zhang et al., 2007), and was therefore chosen as its marker compound. For coal combustion and vehicular exhaust, both characterized by complex mixtures include n-alkanes, polycyclic aromatic hydrocarbons (PAHs), and hopanes (Rogge et al., 1993a; Oros and Simoneit, 2000; Zhang et al., 2008; Cai et al, 2017). Local emission profiles indicated coal combustion tends to emit a higher proportion of 3- to 4-ring PAHs (Zhang et al., 2008), whereas vehicular exhaust is characterized by higher contributions from C20–C22 n-alkanes (Cai et al, 2017). Consequently, we selected 3- to 4-ring PAHs and C20–C22 n-alkanes as the representative organic markers for local coal combustion and vehicle emissions, respectively. Cooking emissions are dominated by fatty acids, particularly saturated palmitic acid (C16:0) and stearic acid (C18:0), as well as unsaturated fatty acids like palmitoleic acid (C16:1) and oleic acid (C18:1). Given the instability of unsaturated fatty acids in ambient air (Kawamura and Gagosian, 1987; Rudich et al., 2007), only the more stable saturated fatty acids—palmitic and stearic acids—were used as characteristic markers for local cooking emissions (He et al., 2004; Zhao et al., 2007 and 2015). For plant debris, emission profiles from Los Angeles area indicate that plants release considerable quantities of long-chain odd-carbon-number n-alkanes (e.g., C25, C27, C29) (Rogge et al., 1993b); however, many local studies indicate that biomass burning, coal combustion, and vehicle exhaust also emit long-chain n-alkanes to some extent (Zhang et al., 2007, 2008; Cai et al., 2017). Overlap among sources diminishes the diagnostic value of long-chain n-alkanes, some investigations have consequently proposed glucose as a more selective tracer for plant debris (Fan et al., 2020; Xu et al., 2021). Puxbaum and Tenze-Kunit (2003) report that glucose comprises approximately 5.2% of plant-derived organic carbon, whereas other sources other than plants rarely emit glucose. On this basis, glucose was adopted here as the plant emission biomarker. Additionally, certain fungal spores contribute to OC in ambient air; literature reports indicate that particles originating from fungal spores contain abundant mannitol and arabitol, accounting for roughly 13% and 19% of OC, respectively (Bauer et al., 2002, 2008). Accordingly, mannitol and arabitol were chosen as the marker compounds representing fungal spores in this study. Detailed source profile information used in CMB model is provided in Fig. S3 of the supplementary material. It should be noted that the local source profile dataset subdivides major sources—such as coal combustion, vehicle exhaust, and cooking—into multiple categories (for example, industrial versus residential coal burning, gasoline versus diesel exhaust, and regional cooking styles such as Guangdong versus Sichuan). For each major source category, we represented typical source characteristics by using

either the mean profile of its subcategories or the average profile derived from multiple observational studies. Similarly, using the proportions of characteristic species in $PM_{2.5}$ from these emission source profiles, the CMB model subsequently estimated each source's contribution to $PM_{2.5}$. In the primary emission profiles applied here, the PM-to-OC ratio ranged from 1.42 to 2.15." **(Lines 126–172, pages 6–7)**.

"To assess the contributions of various secondary sources to OC and $PM_{2.5}$, i.e., SOC and SOA concentrations, we employed tracer-based method, a well-established method for SOC and SOA source apportionment. The tracer-based method operates on principles analogous to those involved in the CMB model, relying on the mass fractions of characteristic tracers in SOC (f_{SOC}) and SOAs (f_{SOA}) from emission sources to ascertain the contributions of different sources. The SOC and SOA tracer-based method was first proposed and utilized by Kleindienst et al. (2007 and 2012). Its specific calculation procedure is as follows:

$$[SOC]=\frac{\sum_i[tr_i]}{f_{SOC}}, \quad (1)$$

$$[SOA]=\frac{\sum_i[tr_i]}{f_{SOA}}, \quad (2)$$

where $\sum i[tri]$ is the total concentration of the selected tracers in the sample, denoting representative compounds from specific emission categories; f_{SOC} and f_{SOA} are the mass fractions of the tracers in OC and $PM_{2.5}$ from secondary emissions, respectively. The values of f_{SOC} and f_{SOA} are determined according to the chamber experiments conducted, in which gaseous precursors are transformed into SOC and SOAs under oxidative and illuminative conditions. Herein, we employed six compounds associated with hemiterpenes, four compounds related to monoterpenes, as well as β-caryophyllinic acid, 2,3-dihydroxy-4-oxopentanoic acid, and phthalic acid as marker compounds to indicate the contributions of different biogenic (hemiterpenes, monoterpenes, and sesquiterpenes) and anthropogenic (toluene and naphthalene) sources to SOC and SOA. We adopted the f_{SOC} and f_{SOA} values used by Kleindienst et al. (2007 and 2012). to calculate the SOC and SOA concentrations originating from various sources. All marker selections, f-values, and the conversion coefficients between f_{SOA} and f_{SOC} derived from chamber studies are listed in Table S2, and the calculations assume that the chamber-derived SOC and SOA relationships are representative of the relevant atmospheric oxidation regimes, acknowledging associated uncertainties." **(Lines 173–193, pages 7–8)**

Second, we have added a detailed explanation and discussion of the errors and uncertainties introduced by the choice of source profiles and representative tracers in Section 3.5 (Source apportionment of OC and aerosol). The specific content is as follows:

"The calculation of POC and SOC contributions based on the CMB model and tracer-based method inherently involves uncertainties. For the CMB model, a primary source of uncertainty is the variability of tracer mass fractions within OC (f_{OC}) from the same primary emission sources across various observational studies. For example, reported levoglucosan/OC fractions range from 12% (Andreae, et al., 2001, 2019) to 13% (Zheng et al., 2002) and 16% (Fine et al., 2004), whereas the local source profile we reference suggests 8.3% (Zhang et al., 2007). Since recent, site-specific emission data are often unavailable, the choice of source profile and corresponding f_{OC} values can strongly influence apportionment outcomes. Secondly, a fundamental assumption

of the CMB model is that the selected markers representing various sources remain stable during atmospheric transport and do not undergo significant chemical transformation. However, truly conservative species are rare. For example, levoglucosan, commonly used as a biomass-burning marker and generally considered relatively stable (Wan et al., 2021), has nonetheless been shown to decrease during long-range transport through wet and dry deposition (Fu et al., 2011) and by photodegradation (Holmes and Petrucci, 2006; Stohl et al., 2007; Hoffmann et al., 2010). Analogous loss or alteration of other source tracers can bias CMB results, tending to underestimate contributions from sources whose markers are degraded or removed during transport. For tracer-based methods, the appropriateness of the selected f_{SOC} and f_{SOA} values is also crucial to the accuracy of the results. Compared with primary source profiles, direct observational information on secondary source profiles is sparse. This paucity reflects the experimental difficulty of reproducing atmospheric oxidation and photochemical ageing under representative light and precursor conditions in laboratory or chamber systems. Most studies employing tracer-based methods to estimate SOC and SOA contributions have relied on f_{SOC} and f_{SOA} values reported by Kleindienst et al. (2007, 2012), which are now relatively dated. SOC and SOA formation are influenced not only by oxidants and sunlight but also by factors such as relative humidity, precursor concentrations, NO_x levels, and other ambient variables. The chamber conditions cannot fully reproduce the complexity of real atmospheres, leading to inevitable discrepancies in f_{SOC} and f_{SOA} values between chamber-derived results and ambient air (Fu et al., 2009; Guo et al., 2012; Ding et al., 2012 and 2014; Haque et al., 2023). Additionally, the f_{SOC} and f_{SOA} values used were average, and their inherent standard deviations could result in deviations of 21% to 48% in the calculated SOC contributions from different sources (Kleindienst et al. 2007 and 2012). Despite the inherent uncertainties associated with both the CMB model and tracer-based method, these approaches remain convenient tools for estimating the contributions of various POC and SOC, yielding relatively reasonable results (Stone et al., 2009; Ding et al., 2012 and 2014; Al-Naiema et al., 2017; Ren et al., 2021; Xu et al., 2021; Haque et al., 2023). However, further efforts are needed to reduce these uncertainties. One critical step will be to conduct more extensive, site-specific observational studies of different types of primary and secondary emission sources. Such data are essential for identifying representative tracers within different sources and accurately determining their mass fractions in OC and OAs. Additionally, when f_{SOC} and f_{SOA} values are applied in both CMB model and tracer-based calculations, using the most recent, locally relevant emission source profile to minimize uncertainties is advisable." **(Lines 599–634, pages 23–24)**

Overall comments 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.

Response: We thank the reviewer for this important comment. As noted in our response to Comment 1, we have expanded Section 2.3 (Source apportionment methods) to describe in detail the source profiles used in the CMB analysis, the rationale for their selection, and the representative tracers chosen. We have also expanded Section 3.5 (Source apportionment of OC and Aerosol) to provide a thorough discussion of the uncertainties and potential errors introduced by differences in source profiles and tracer representativeness. Please refer to our response to Comment 1 and the revised manuscript (Sections 2.3 and 3.5) for the full details.

Overall comments 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.

Response: Thank you for this comment. In the original manuscript we estimated POC and SOC using the tracer-based method and compared those estimates with CMB results. In the revised manuscript we treat the two approaches independently: the CMB is used to apportion POC sources, while the tracer-based method is applied separately to estimate SOC fractions. Because the methods are now applied to different OC fractions, the previous mutual-validation comparisons between the tracer-based method and the CMB have been removed. As noted in our responses to the comment 1 and 2, we have expanded Section 2.3, "Source apportionment methods," to provide a detailed description of the source profiles employed in the CMB analysis and the representative chemical species selected, together with justification for these choices.

Overall comments 4. Improve readability to integrate results from multiple tracers, rather than treating them one by one (i.e. pages X to Y).

Response: Thank you for this valuable suggestion. We have reorganized the Results to present tracer information in a more integrated and synthetic way. Specifically, primary-emission tracers are discussed in grouped categories (e.g., sugar compounds are considered together; lignin, resin-derived products and sterols are discussed as a unit; glycerol, hydroxy acids and aromatic acids are treated collectively). Secondary-emission tracers are likewise grouped into anthropogenic and biogenic classes and interpreted together to highlight coherent source and formation patterns. These changes improve readability and emphasize cross-tracer consistency; please see the revised manuscript Sections 3.2–3.4 (Major polar components, Minor polar components, and SOA tracers in $PM_{2.5}$) for the updated presentation.

Overall comments 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 theselected 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.

Response: We thank the reviewer for highlighting the portion of OC (other OC) that remains unattributed. We have added detail analysis and discussion of other OC in the revised manuscript. The relevant discussion is expressed in the updated manuscript as follows:

"The fraction of OC that remains unexplained by the CMB model and tracer-based methods and is therefore classified as "other OC". This component is defined as the difference between measured OC and the summed contributions of POC and SOC from all apportioned sources estimated by the CMB model and tracer approaches. On average, other OC accounts for ~34% of measured OC (Figures. 5c, 5f). The presence of other OC likely reflects limitations in source identification:

some OC forms (e.g., liquid-phase OC or aged primary OC) are difficult to capture with either the CMB model or tracer techniques (Ding et al., 2014; Fan et al., 2019; Wu et al., 2020; Xu et al., 2021). Although absolute concentrations of other OC are lower in spring and summer than in autumn and winter, its fractional contribution shows the opposite seasonal pattern—about 45%–50% in spring and summer versus 12%–29% in autumn and winter (Figures. 5c, 5f). Similar patterns have been reported elsewhere in other citie of China, e.g., Beijing, where estimated concentrations of other OC are higher in winter but its proportion is greater in summer (~44% vs. 22–25% in winter) (Wang et al., 2009; Wu et al., 2020; Xu et al., 2021). Previous studies indicate that this "other OC" is likely dominated by SOC (Guo et al., 2012; Wu et al., 2020; Xu et al., 2021). To assess whether other OC is predominantly primary or secondary, we used an EC-based method to estimate total POC and SOC; unlike tracer methods, the EC approach simply partitions OC into POC and SOC and therefore can include unapportioned components. Accordingly, the POC calculated via EC-based method minus the POC calculated via CMB model yields the "other POC" (unapportioned POC), and similarly, SOC calculated via EC-based method minus the tracer-based SOC yields the "other SOC" (Figure. S8). Our results show that, across all four seasons, the concentration of other SOC exceeds that of other POC (Figures. S8a, S8b), indicating that these unidentified other OC components are likely predominantly SOC. The concentration of other SOC is substantially higher in autumn and winter—especially during pollution periods in winter—highlighting the critical role of SOC in aerosol pollution during these seasons (Figures. S8c, S8d). Conversely, the proportion of other SOC during summer is the highest among all seasons (around 32%) (Figure. S8b), suggesting that elevated temperatures and intense radiation during summer enhance SOC formation efficiency (Wu et al., 2020; Xu et al., 2021). Although the other SOC fraction exceeds other POC, EC-based estimates of total SOC remain smaller than total POC in all seasons. Similarly, SOC estimated by the tracer method is markedly lower than POC apportioned by the CMB model. These results together indicate that primary emissions play a significant role in urban OC pollution, with substantial POC contributions forming the basis for elevated OC concentrations. However, the influence of SOC should not be overlooked. Numerous studies have shown that even minor increases in SOC during winter pollution episodes can exacerbate OA pollution (Li et al., 2019; Xu et al., 2021; Haque et al., 2023)." **(Lines 560–590, pages 21–22)**

Overall comments 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.

Response: Thank you very much for your valuable comment. In the revised manuscript, all data used for linear fitting were first tested for normality. Specifically, the Shapiro-Wilk test was applied to assess the normality of the data. Relevant explanations and discussions can be found in Section 3.6 (Characteristics of OAs during winter pollution) **(Lines 662–664, page 25)**. Additionally, figures that include linear fits (Figures 7 and S12) now include the following note: "Before conducting linear correlation analyses, the Shapiro-Wilk test was used to assess the normality of the data, and linear correlations were conducted only for datasets significantly conforming to a normal distribution ($p > 0.05$)."

Overall comments 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.

Response: Thank you for this helpful suggestion. In the revised manuscript, concentration values are reported with two to three significant figures, consistent with the estimated uncertainties. This unified format has been applied consistently throughout the text, tables, figures, and Supplementary Information.

Overall comments 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.

Response: Thank you very much for your valuable comment. Considering that unsaturated fatty acids are prone to degradation during the transfer process, we excluded unsaturated fatty acids when selecting fatty acids as characteristic compounds for cooking emission sources. The relevant revisions are as follows:

"Cooking emissions are dominated by fatty acids, particularly saturated palmitic acid (C16:0) and stearic acid (C18:0), as well as unsaturated fatty acids like palmitoleic acid (C16:1) and oleic acid (C18:1). Given the instability of unsaturated fatty acids in ambient air (Kawamura and Gagosian, 1987; Rudich et al., 2007), only the more stable saturated fatty acids—palmitic and stearic acids—were used as characteristic markers for local cooking emissions (He et al., 2004; Zhao et al., 2007 and 2015)." **(lines 148–152, page 6)**

Overall comments 9. A more thorough discussion of the limitations of the current work are needed.

Response: Thank you very much for this important suggestion. We have expanded the Conclusion to include an in-depth consideration of the study's limitations (please see Conclusion, new paragraph). The added text is given below:

"The study findings underscore the necessity for targeted management strategies that consider primary and secondary anthropogenic emission sources across different seasons and pollution periods. Although the CMB model and tracer-based method provided preliminary insights into the OC and OA from diverse sources, the study encountered several inherent limitations. First, the primary emission source profiles employed in the CMB model exhibit variability across different studies and regions, making it challenging to establish standardized source characteristic parameters and potentially affecting the accuracy of source apportionment. Second, the proportions of SOC tracers obtained from laboratory chamber experiments are influenced by various factors, and incorporating these proportions into tracer-based methods may introduce potential biases or uncertainties in the estimates. To address these issues, future research should prioritize comprehensive observational studies of primary emission sources to obtain

high-resolution, region-specific emission data, thereby improving the applicability of source profiles. Combining field observations with laboratory simulations can also provide a more accurate characterization of secondary emissions, ultimately reducing the uncertainties associated with tracer-based estimates of SOC contributions." **(lines 694–706, pages 26–27)**

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.

Response: Thank you for these valuable comments. We have added a description of the relevant analytical methods in the abstract. Specific biomarkers and diagnostic ratios were applied to characterize OA sources and distribution patterns, while chemical mass balance (CMB) models and tracer-based approaches were used to estimate source contributions. Statistical analyses were conducted to investigate OA characteristics and drivers during winter pollution episodes. The revised abstract is as follows:

"Due to the complex composition of organic aerosols (OAs), identifying their sources and understanding their dynamics remain challenging, particularly in urban environments of China where natural and anthropogenic influences to OAs intersect. This study aimed to clarify the relative contributions of primary emissions and secondary formation to urban OAs and confirm the sources and influencing factors of OA pollution. We analyzed major polar organic compounds in fine particulate matter ($PM_{2.5}$) collected over one year in Nanchang, Central China. Specific biomarkers and diagnostic ratios were applied to characterize OA sources and distribution patterns, while chemical mass balance (CMB) models and tracer-based approaches were used to estimate source contributions. Statistical analyses were conducted to investigate OA characteristics and drivers during winter pollution episodes. Notably, fatty acids, fatty alcohols, and saccharides predominated over lignin, resin products, sterols, glycerol, hydroxy acids, and aromatic acids, with molecular profiles indicating both anthropogenic and biogenic origins. Source apportionment results showed that primary organic carbon (POC) and primary OAs (POAs) contributed 58% of total organic carbon and 23% of $PM_{2.5}$ mass, respectively, compared with 8% and 4% from secondary organic carbon (SOC) and secondary OAs (SOAs). Anthropogenic sources dominated, accounting for approximately 90% of POC and POAs as well as 60% of SOC and SOAs. Seasonal patterns revealed stronger biogenic influences in spring–summer, whereas anthropogenic emissions dominated in autumn–winter. Short-term winter episodes were characterized by rapid secondary formation, facilitated by elevated primary emissions and favorable oxidation conditions, including enhanced light intensity and nitrogen oxides." **(lines 11–28, pages 1–2)**

Specific comments 11. At lines 92-94, indicate the specific locations of the meteorological and gas sensors and their relation to the PM sampling site.

Response: Thank you for this helpful suggestion. We have added a precise description of the sensor locations and their spatial relationship to the PM sampling site. The specific content is as follows:

"In particular, the meteorological data were retrieved from the ground meteorological observation

station at Nanchang Changbei Airport, and gaseous pollutant data were obtained from the air quality monitoring station operated by the Jiangxi Academy of Forestry. These stations are the closest to the sampling site, located approximately 10 km and 2 km away, respectively. Both stations are ground-based, equipped with online monitoring instruments, and situated on open, flat terrain without surrounding buildings or obstructions. A detailed summary of the prevailing meteorological conditions and air quality during the sampling period is provided in Text S1 and Figure S2." **(Lines 96–102, page 4)**.

Specific comments 12. Lines 99-100, include a reference to the IMPROVE protocol used for EC and OC analysis.

Response: Thank you — we have added a reference to the IMPROVE thermal/optical reflectance protocol and included a concise methods description.

Specifically: "The OC and EC concentrations in the $PM_{2.5}$ samples were quantified using the Desert Research Institute Model 2001 Carbon Analyzer, following the thermal/optical reflectance protocol established by the Interagency Monitoring of Protected Visual Environments (IMPROVE) (Chow et al., 2007). A 1.0-cm^2 filter sample was placed in a quartz boat in the analyzer and subjected to incremental heating at predetermined temperatures: 140 °C for OC1, 280 °C for OC2, 480 °C for OC3, and 580 °C for OC4 in a non-oxidizing helium atmosphere; and 580 °C for EC1, 740 °C for EC2, and 840 °C for EC3 in an oxidizing atmosphere containing 2% oxygen in helium." **(Lines 104–110, pages 4–5)**.

Specific comments 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..

Response: Thank you for these valuable comments. We included the n-alkane standards mainly because the extracted species consist of polar and non-polar organic compounds, which are detected simultaneously by the instrument. Therefore, we added non-polar C13 n-alkanes as internal standards for quantitative analysis. The relevant explanation has been added in Section 2.2 "Chemical analyses" the to the manuscript. **(Lines 115–117, page 5)**

Specific comments 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.

Response: Thank you for these valuable comments. The relevant content has been removed from the manuscript.

Specific comments 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.

Response: Thank you for this important comment. We have added a relevant discussion in Section

3.5 (Source apportionment of OC and aerosols) regarding the potential bias in source estimation caused by the wet and dry deposition as well as photochemical degradation of levoglucosan during transport. The specific details are as follows:

"Secondly, a fundamental assumption of the CMB model is that the selected markers representing various sources remain stable during atmospheric transport and do not undergo significant chemical transformation. However, truly conservative species are rare. For example, levoglucosan, commonly used as a biomass-burning marker and generally considered relatively stable (Wan et al., 2021), has nonetheless been shown to decrease during long-range transport through wet and dry deposition (Fu et al., 2011) and by photodegradation (Holmes and Petrucci, 2006; Stohl et al., 2007; Hoffmann et al., 2010). Analogous loss or alteration of other source tracers can bias CMB results, tending to underestimate contributions from sources whose markers are degraded or removed during transport." **(lines 605–613, page 23)**

Specific comments 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.

Response: Thank you for bringing this important literature to our attention. We have incorporated the Frauenheim et al. (2022) findings that the C5-alkene triols with double bond structures are likely artifacts resulting from thermal decomposition during the GC–MS analysis. Therefore, in the revised manuscript, we have provided a detailed discussion on the impact of these artifacts on our source apportionment results. The specific discussion is as follows:

"Notably, the C5-alkene triols detected in our study predominantly exist as double-bonded "triol" compounds, including cis-2-methyl-1,3,4-trihydroxy-1-butene, 3-methyl-2,3,4-trihydroxy-1-butene, and trans-2-methyl-1,3,4-trihydroxy-1-butene. In fact, acid-catalyzed ring-opening reactions and isomerization of particle-phase IEPOX can also produce various "diol" compounds, which typically exist as cyclic structures, such as trans-3-methyltetrahydrofuran-3,4-diol and cis-3-methyltetrahydrofuran-3,4-diol. These "diols" are also important components of C5-alkene triols (Li et al., 2013; Frauenheim et al., 2022). Recent work, however, indicates that C5-alkene triols detected via GC–MS are unlikely to originate solely from the acid-catalyzed ring-opening reactions and isomerization of IEPOX; instead, they may largely be artifacts produced by thermal decomposition during GC–MS analysis, with roughly 90% of the detected "triol" signal attributable to such artifacts (Frauenheim et al., 2022). If such artifacts indeed exist in the GC–MS measurements, it implies that the C5-alkene triol levels reported in our results are overestimated, potentially leading to an overall overestimation of isoprene-derived SOA tracers by approximately 30%. Despite the potential artifact issue in GC–MS detection, the contribution of C5-alkene triols to SOA should not be underestimated. These compounds are semi-volatile compounds that can volatilize back into the atmosphere from the particle phase, where they may undergo further oxidation by OH radicals. This oxidation process can significantly influence both the mass and composition of SOAs (Frauenheim et al., 2022 and 2024)." **(lines 455–471, pages 17–18)**

Specific comments 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.

Response: Thank you for this helpful suggestion. We have revised Figures 5, S6 and F7 to improve readability by enlarging the legend text and increasing the size/clarity of subscripts that distinguish the source labels. High-resolution versions of the updated figures are included in the revised manuscript and Supplementary Information.

Specific comments 18. Lines 527-528, please explain how OC was converted into OA.

Response: Thank you for pointing this out. In Section 2.3 (Source apportionment methods), we have provided an explanation of how the conversion between OC and OA was handled when calculating the contributions of OC and OA using the CMB model and tracer-based methods.

Specifically: "Similarly, using the proportions of characteristic species in $PM_{2.5}$ from these emission source profiles, the CMB model subsequently estimated each source's contribution to $PM_{2.5}$. In the primary emission profiles applied here, the PM-to-OC ratio ranged from 1.42 to 2.15." **(lines 169–172, page 7)**

"All marker selections, f-values, and the conversion coefficients between f_{SOA} and f_{SOC} derived from chamber studies are listed in Table S2, and the calculations assume that the chamber-derived SOC and SOA relationships are representative of the relevant atmospheric oxidation regimes, acknowledging associated uncertainties." **(lines 190–193, pages 8)**

The authors would like to express their sincere gratitude to the editor and reviewers for their valuable suggestions and comments, which have greatly improved the quality of this manuscript.