The review for egusphere-2023-2593: Non-negligible secondary contribution to brown carbon in autumn and winter: inspiration from particulate nitrated and oxygenated aromatic compounds in urban Beijing

In view of its unique light absorption properties and photochemical reactivity, brown carbon is believed to have potential impacts on global climate and atmospheric environmental quality. Therefore, brown carbon has become a hot topic in the field of environmental research in recent years. This paper uses offline methods and GC/MS to investigate the concentration variation and source characteristics of various brown carbon molecules before and during heating period in Beijing. It is found that vehicle emissions, biomass burning and coat combustion are important sources of brown carbon before and during heating period. And secondary source was important for brown carbon. However, the information of the dozen or so molecules that serve as BrC are lacking and the source apportionment are needed more logically accurate and in-depth analysis. Here are the comments.

Major comment:

1.1. The title of this article suggests that the research object is brown carbon, and the author has surfaced in the text that NAC and OPAH are two types of substances that have been confirmed as BrC molecules in previous reports. The problem is that the objects studied by the authors are actually more than a dozen confirmed molecules, some of which have been studied and confirmed as BrC molecules, determining the light absorption characteristics of these molecules, such as xie et al., 2017 and Huang, R.-J., 2018. In order to make the conclusions of the paper more rigorous, the author should supplement relevant information on the light absorption parameters of these molecules, otherwise BrC and these more than a dozen molecules are not equivalent.

1.2. What were the reasons behind the authors' selection of CO over levo as a tracer? In my interpretation, the long-lived and inert nature of the tracer (as per the principle and assumptions of the EC-tracer method) ensures that its mass concentration closely approximates that at the start of emission during atmospheric

transport. Consequently, the initial emission value can be derived through an emission ratio calculation, as illustrated by Eq. 1. Both levo and CO satisfy this condition, so it is reasonable to use both as tracers in the calculation. However, the authors state otherwise, citing the literature. This prompted the reviewers to question. The authors should provide a more comprehensive explanation of the fundamental assumptions and principles underlying this method, ensuring that readers can comprehend it thoroughly. The correlation analysis reveals a strong association between NAC and OPAH with levo, suggesting a preference towards biomass combustion sources. This finding contradicts the author's initial conclusion. Would the use of levo as a tracer yield a different outcome?

Xie, M., Chen, X., Hays, M. D., Lewandowski, M., Offenberg, J., Kleindienst, T. E., and Holder, A. L.: Light Absorption of Secondary Organic Aerosol: Composition and Contribution of Nitroaromatic Compounds, Environ. Sci. Technol., 51, 11607-11616, 10.1021/acs.est.7b03263, 2017.

Huang, R.-J., Yang, L., Cao, J., Chen, Y., Chen, Q., Li, Y., Duan, J., Zhu, C., Dai, W., Wang, K., Lin, C., Ni, H., Corbin, J. C., Wu, Y., Zhang, R., Tie, X., Hoffmann, T., O'Dowd, C., and Dusek, U.: Brown Carbon Aerosol in Urban Xi'an, Northwest China: The Composition and Light Absorption Properties, Environ. Sci. Technol., 52, 6825-6833, 10.1021/acs.est.8b02386, 2018.

Other comment:

1.1. Line 25: From the content of the article abstract, it seems to be expressed here during heating rather than after heating.

1.2. Line 68-70: 67% is a specific number, is it determined for all or for a certain site at a certain time?

1.3. Line 79 Please re-write citation formats correctly.

1.4. Line 96: Cai et al., 2022 and Yi Chen et al., 2022 did not study the importance of NAC as brown carbon but studied the NAC itself, please quote the literature correctly.

1.5. Line 164: Does this period indicate the whole study period or the 15% point selected by the authors? The assumption that the lowest 15% of the ratio of

NAC/CO is used to calculate emission ratios, is there any or some direct data or reports on emissions that can be used to justify that this proportion is correct to some extent?

1.6. Line 209: It is not clear here. The authors suggest that the increase of OM during heating was contributed by POA and the secondary formation of emitted VOC? Also, it is interesting to know what could be the reason for the increase of un-resolved (others) during heating? Is it because of the combustion of coal and others during heating which leads to an increase in the proportion of components such as heavy metals? In addition, the large temperature and humidity difference before and after heating may be one of the reasons for the SIA difference, since temperature and humidity affect the partitioning of NO3 and SO4, which can be analyzed more detailedly by the authors in a discussion combined with previous reports.

1.7. Line 294: The author thinks that there are only two sources for NAC and OPAH, combustion sources and secondary formation sources, have they been reported before, will there be other sources? According to Cai et al., 2022 and the text above, has biomass burning been included in the combustion source here?