

The authors thank the referees to review our manuscript and particularly for the valuable comments and suggestions that have significantly improved the manuscript. We provide below point-by-point responses (in blue) to the referees' comments and have made changes accordingly in the revised manuscript.

#### Referee #2

The paper reports the contribution of PM<sub>2.5</sub>-bound nitrated aromatic compounds to the optical properties of brown carbon. Seasonal variations of concerned species were discussed as well. The topic is interesting and suitable for the journal, and the paper is well organized and understandable. However, some problems need further discussion.

In conclusion, I suggest it for publication after the authors addressing the following specific points:

1. Line 63-65, considering that this paper mainly focuses on the optical properties of NACs, the introduction should include more previous research findings regarding the light absorption ability of NACs rather than just one sentence.

Response: Thanks for pointing this out. We have added more previous research findings in line 66-73, it now reads "...; Teich et al., 2017; Li et al., 2020). For example, Zhang et al. (2013) estimated the contribution of NACs to BrC light absorption of ~4% in the Los Angeles Basin. Mohr et al. (2013) calculated the contribution of NACs to BrC light absorption of about 4% in Detling, United Kingdom. Teich et al. (2017) investigated the contribution of NACs to BrC light absorption during six campaigns of 0.02-4.41% for acidic conditions and 0.02-9.86% for alkaline conditions. Li et al. (2020) estimated the contribution of NACs to BrC light absorption in Beijing of 0.28-3.44% in fall and 1.03-6.49% in winter."

2. Line 66-67, could the author add a few more sentences on why NACs are harmful to human health?

Response: We have added a few more sentences in line 75-77, it now reads "...For

example, NACs can interact with DNA and cause mutagenesis (Purohit and Basu, 2000; Ju and Parales, 2010). NACs can also damage cells, resulting in cell degeneration and canceration (Kovacic and Somanathan, 2014).”

3. Line 153, what are the uncertainties of the input species?

Response: We re-checked the uncertainties (RSDs) and have now added these values in the revised manuscript. In line 160, it now reads “..., with uncertainties (RSD) < 10%.”

4. Line 154, The constrain of specific species in different sources will influence the Q value of the solution, thus the setting should be extremely cautious. After this kind of setting, is the %dQ value acceptable? Is the PMF solution still robust?

Response: The constrain of specific species in different sources do affect the Q value of the solution. In this study, the  $Q/Q_{exp}$  value was 1-5 after setting, which is acceptable, and the PMF was ran in the robust model.

5. Line 160, any reason to choose a 72-h backward trajectory instead of 24-h or 48h?

Response: We have added the reason in Line 171-172, it now reads “According to the lifetimes of different secondary species (Wojcik and Chang, 1997; Chow et al., 2015), ...”

6. Line 298-300, besides the high emissions of NACs in winter, are any other PM2.5 components that may contribute to the enhanced light absorption between 300-500 nm during winter?

Response: In addition to NACs, some PAHs also have strong light absorption capacity in wavelength of 300-500 nm (Huang et al., 2018; Lin et al., 2018). In winter, the emissions of PAHs increase because of heating activities, which may contribute to the enhanced light absorption.

7. Section 3.4, based on the PMF results, the author may consider using multilinear

regression analysis to investigate which source contributes most to the light absorption ability of NACs.

Response: Multilinear regression analysis and PMF receptor model can both be used to investigate the sources of BrC. In this study, to get non-negative result, we used PMF rather than multilinear regression analysis to analyse the sources of NACs, and the results are discussed in Section 3.2. "Sources of NACs".

## References

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Referee #3

This work provides a comprehensive report to the variation, sources, origins, and light absorption of nitrated aromatic compounds (NACs) in four seasons in a mega city in Northwest China. The results highlight the elevated concentrations and enhanced light absorption of NACs in winter in East Asia and confirm the dominant contributions from combustions sources including coal combustion, biomass burning, and vehicle exhausts. The manuscript is generally well written with clear logic, full discussion, and fluent language. It can be accepted after addressing a few minor comments.

Specific comments:

1. Line 105-107, are any blank samples collected or obtained in this study?

Response: In this study, at least one blank filter sample was measured for light absorption and organic compounds for every ten ambient samples. In line 126-127, it now reads, "At least one blank filter sample was measured for every ten ambient samples."

2. Line 118-119, is any inner standard used when determining the concentrations of NACs?

Response: In our study, 4-nitrophenol-2,3,5,6-d<sub>4</sub> was used as an internal standard to correct for potential loss for NAC quantification (Chow et al., 2015).

3. What is the wavelength range of the UV-Vis spectrophotometer used in this study?

Response: 300-700 nm.

4. Line 262-263, it's better to derive some implications to pollution control here.

Response: Thanks for pointing this out. In line 279-280, it now reads "..., suggesting that control of anthropogenic emissions (biomass burning and coal burning) is important for mitigating pollution of NACs in this region."

5. Line 271-275, were the relatively high concentrations of NACs in the air masses from Gansu and Xinjiang mainly caused by the intensive emissions from urban areas along the trajectories?

Response: As far as we known, there was no studies reporting the concentration of NACs in Gansu and Xinjiang. However, the annual average concentration of PM<sub>2.5</sub> was about 40 µg/m<sup>3</sup> in 14 cities in Gansu in 2016, especially, the PM<sub>2.5</sub> concentration in Lanzhou in 2016 was over 50 µg/m<sup>3</sup> (Liao et al., 2020). The annual average concentration of PM<sub>2.5</sub> was about 55 µg/m<sup>3</sup> in 16 cities in Xinjiang in 2016 (Rupakheti et al., 2021). Therefore, it is possible that NACs were transported/formed along the trajectories from areas with strong emissions.

6. Line 332-333, the difference in the light absorption ability among different NACs is of course the major cause. Suggest elaborating on the differences here, e.g., 4NC has high light absorption ability.

Response: Thanks for pointing this out. In line 353-354, it now reads “...For example, 4-nitrocatechol has lower mass concentration, but higher light absorption contribution, compared to 4-nitrophenol.”

## References

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