# Point-by-Point Response to Reviewers' Comments

Manuscript Ref: acp-2020-1020

Title: Source Apportionment of Fine Aerosol at an Urban Site of Beijing using a Chemical Mass Balance Model

Journal: Atmospheric Chemistry and Physics

## Comments from Reviewer #1

### General Comments:

The manuscript presents a study conducted in 2016-2017 in Beijing to estimate the sources of OC in the fine PM fraction using the CMB source apportionment technique. The analytical procedures are described in detail and the manuscript presents a great deal of results which are compared with previous studies in the same or nearby areas. The study focuses mainly to the fine OC fraction. This should be better reflected in the title. In addition, the objectives of the study should be better specified. The apportionment of $PM_{2.5}$ is a secondary outcome of the study based on OM/$PM_{2.5}$ ratios from the literature, which introduces a considerable amount of uncertainty. This aspect of the methodology should be clarified to the reader in the presentation of the results. In addition, the reliability of the CMB in relation to other source apportionment techniques should be better documented. Despite the number of samples (31 in winter and 34 in summer) is coherent with the CMB technique, it is quite limited to represent the variability of meteorological and emission situations in a complex situation like a megacity. Some of the presented results need more discussion to explain what is their relevance for the purposes of the study. For instance: What is the added value of comparing Beijing with a rural location? What is the importance of distinguishing between haze and non-haze days? The readability of the text could be improved by removing the repetition of data already presented in figures and tables. The conclusions should explain what is the added value of this study with respect to the previous knowledge on $PM_{2.5}$/OC sources in Beijing. In addition, the authors should underline how the combination of two different approaches (CMB and PMF) contribute to obtain more complete/robust estimations.

**General response:** We thank the reviewer for very constructive suggestions and are pleased to respond.

(1) This study focuses mainly to the fine OC fraction. As suggested by the reviewer, to better reflect that in the title, the original title "Source Apportionment of Fine Aerosol at an Urban Site of Beijing using a Chemical Mass Balance Model" has been changed to "Source Apportionment of Fine Organic Carbon at an Urban Site of Beijing using a Chemical Mass Balance Model"

(2) The objectives of this study are specified and added in the revised manuscript, please see **lines 111-115** in the revised manuscript, which are also copied below:

“The objectives of this study are: 1) to quantify the contributions of pollution sources to OC by applying a CMB model and compare them with those at a rural site of Beijing; 2) to compare the source apportionment results by CMB with those from Aerosol Mass Spectrometer-PMF analysis (AMS-PMF), to improve our understanding of different sources of OC.”

(3) We agree with the reviewer that the calculation of $PM_{2.5}$ contributions using $OC/PM_{2.5}$ ratios from the applied source profiles introduces uncertainty. For example, due to the variability of the OC/ $PM_{2.5}$ ratio in the source profiles, the application using the average OC/ $PM_{2.5}$ ratio of each source to convert the OC to $PM_{2.5}$ in all samples may be subject to uncertainties, as both organic species and $PM_{2.5}$ mass measurements are subject to analytical imprecision. Unfortunately, insufficient data are available for a formal analysis of uncertainty, but errors of around +/- 10% seem very probable. In addition, instead of $OC/PM_{2.5}$, applying an OM/OC ratio to cooking and vegetative detritus sources for the calculation may result in an underestimation of $PM_{2.5}$ source contributions from these sources, because they can also emit inorganic pollutants. However, cooking emissions are mostly organic and the contribution from vegetative detritus to $PM_{2.5}$ is very small, so their effects on source contribution estimation here are considered negligible. Please also see response to Comment 26.

(4) The repetition of data that are already presented in figures and tables has been removed to improve the readability of the text. Please see the responses to specific comments below.

(5) It is valuable to separate haze and non-haze days, as if policy is directed at reducing the number of haze days, it is essential to know the contributing sources, which may differ from non-haze days. There is also value in studying both rural and urban sites, as both exceed health-based guidelines and require evidence-based mitigation policies which may differ depending on the source apportionment at each. Furthermore, urban air pollution may affect the pollution levels in rural areas (Chen et al., 2020), and domestic heating and cooking led to high emissions of particles and precursor gases, which may contribute to air pollution in the cities (Liu et al., 2021). Please also see responses to Comment 14 and 21.

(6) The added value of this study with respect to the previous knowledge on $PM_{2.5}$/OC sources in Beijing is provided in the conclusion and copied below:

“Although the annual average $PM_{2.5}$ levels in Beijing reduced from 88 µg $m^{-3}$ in year 2013 to 58 µg $m^{-3}$ in year 2017 (Vu et al., 2019), and the deweathered concentration of $PM_1$ decreased by -38% in 2017 comparing to 2007 (Zhang et al., 2020), our CMB modelling results indicate that the coal combustion and biomass burning still remained the dominant primary OC sources in winter 2016 and summer2017, with road traffic ranked as the third highest. Cooking was a more significant source of OC than biomass burning at the urban site during summer. Compared to other CMB studies in Beijing, our study revealed an increase of the contributions from coal combustion, biomass burning and traffic to $PM_{2.5}$ in winter 2016 compared to winter 2000, while those in this study remained similar compared to winter 2013."

(7) Receptor modelling techniques are still an active area of research. None is infallible, and although they are rarely compared, they may not give consistent findings when they are. Such comparisons are important as they highlight the areas of uncertainty. In this study, most factor contributions were in good agreement, but some of the factor contributions from CMB and AMS-PMF varied significantly; for example, cooking OC was 2 times different between the two methods, and it is important to highlight such uncertainties.

(8) Variability of meteorological and emission situations - see response to comment 8. We recognize that our study did not cover all seasons, but the winter and summer study periods represent the typical emission situations in respective seasons.

**Specific Comments:**

Comment 1 - Abstract: the abbreviation SNA is not explained

**Response:** In abstract line 29-30, the original phrase "Secondary inorganic ions (sulfate, nitrate, ammonium; SNA)" has been changed to "Secondary inorganic ions (sulfate, nitrate, ammonium- SNA)" to avoid confusion. Please see **line 30** in the revised manuscript.

Comment 2 - Abstract Line 46: this sentence is misleading as the CMB did not apportion any source of secondary OA.

**Response:** The original sentence "The CMB was found to resolve more primary OA sources than AMS-PMF but the latter apportioned more secondary OA sources." has been modified as "The CMB was found to resolve more primary OA sources than AMS-PMF but the latter could apportion secondary OA sources." to avoid any misunderstanding. Please see **lines 45-46** in the revised manuscript.

Comment 3 - Line 73: This statement is not fully correct. At present, there are many numerical tests to support the decisions about the best solution in PMF. The citation here is out of date for this topic.

**Response:** The original sentence has been modified, and new citations have been added. The original sentence "This approach has some underlying issues: firstly, PMF requires a relatively large sample size; and a "best" solution of achieved factors is subjective (Ulbrich et al., 2009);" has been revised as "This approach has some underlying challenges. For example, PMF requires a relatively large sample size and a "best" solution of achieved factors requires critical assessment of its mathematical parameters and evaluation of the physical reasonability of the factor profiles (de Miranda et al., 2018; Ikemori et al., 2021; Oduber et al., 2021);" Please see **lines 73-77** in the revised manuscript.

Added references:

de Miranda, R. M., de Fatima Andrade, M., Dutra Ribeiro, F. N., Mendonça Francisco, K. J., and Pérez-Martínez, P. J.: Source apportionment of fine particulate matter by positive matrix factorization in the metropolitan area of São Paulo, Brazil, Journal of Cleaner Production, 202, 253-263, https://doi.org/10.1016/j.jclepro.2018.08.100, 2018.

Ikemori, F., Uranishi, K., Asakawa, D., Nakatsubo, R., Makino, M., Kido, M., Mitamura, N., Asano, K., Nonaka, S., Nishimura, R., and Sugata, S.: Source apportionment in PM2.5 in central Japan using positive matrix factorization focusing on small-scale local biomass burning, Atmospheric Pollution Research, https://doi.org/10.1016/j.apr.2021.01.006, 2021.

Oduber, F., Calvo, A. I., Castro, A., Blanco-Alegre, C., Alves, C., Calzolai, G., Nava, S., Lucarelli, F., Nunes, T., Barata, J., and Fraile, R.: Characterization of aerosol sources in León (Spain) using Positive Matrix Factorization and weather types, Science of The Total Environment, 754, 142045, https://doi.org/10.1016/j.scitotenv.2020.142045, 2021.

Comment 4 - Line 78: "composed" instead of "comprised".

**Response:** This is revised. Please see **line 80** in the revised manuscript.

Comment 5 - Line 80: Explain where.

**Response:** The original sentence "OM was the largest contributor to $PM_{2.5}$ mass, which accounted for 30%-60% of $PM_{2.5}$ (Song et al., 2007; He et al., 2001; Huang et al., 2014), and can contribute up to 90% of submicron PM mass (Zhou et al., 2018)." has been modified to "OM was the largest contributor to $PM_{2.5}$ mass, which was reported to account for 30%-50% of $PM_{2.5}$ in some Chinese cities such as Beijing, Guangzhou, Xi'an and Shanghai (Song et al., 2007; He et al., 2001; Huang et al., 2014), and can contribute up to 90% of submicron PM mass in Beijing (Zhou et al., 2018)." Please see **lines 83-86** in the revised manuscript.

Comment 6 - Line 86: Provide references about the reliability of CMB compared to other source apportionment methods.

**Response:** Regarding the reliability of the CMB, more discussion has been included and references have been added.

The original sentence "A few studies have also applied a Chemical Mass balance (CMB) model for source apportionment of PM in Beijing." has been modified to "Chemical Mass balance (CMB) model has been used for source apportionment of PM worldwide, including in the US (Antony Chen et al., 2010), UK (Yin et al., 2015), and China (Chen et al., 2015b). The CMB model assumes that source profiles remain unchanged between the emitter and receptor (Sarnat et al., 2008; Viana et al., 2008). Xu et al. (2021)

compared the source apportionment results of fine particles by multiple receptor modelling approaches, and found that CMB can provide the most complete and representative source apportionment of Beijing aerosols. A few studies have applied a CMB model for source apportionment of PM in Beijing (Zheng et al., 2005; Liu et al., 2016; Guo et al., 2013; Wang et al., 2009).” Please see **lines 90-98** in the revised manuscript.

References:

Antony Chen, L. W., Watson, J. G., Chow, J. C., DuBois, D. W., and Herschberger, L.: Chemical mass balance source apportionment for combined PM2.5 measurements from U.S. non-urban and urban long-term networks, Atmospheric Environment, 44, 4908-4918, https://doi.org/10.1016/j.atmosenv.2010.08.030, 2010.

Chen, P., Wang, T., Hu, X., and Xie, M.: Chemical Mass Balance Source Apportionment of Size-Fractionated Particulate Matter in Nanjing, China, Aerosol Air Qual. Res., 15, 1855-1867, 10.4209/aaqr.2015.03.0172, 2015.

Guo, S., Hu, M., Guo, Q., Zhang, X., Schauer, J. J., and Zhang, R.: Quantitative evaluation of emission controls on primary and secondary organic aerosol sources during Beijing 2008 Olympics, Atmos. Chem. Phys., 13, 8303-8314, 10.5194/acp-13-8303-2013, 2013.

Liu, Q., Baumgartner, J., Zhang, Y., and Schauer, J. J.: Source apportionment of Beijing air pollution during a severe winter haze event and associated pro-inflammatory responses in lung epithelial cells, Atmospheric Environment, 126, 28-35, https://doi.org/10.1016/j.atmosenv.2015.11.031, 2016.

Sarnat, J. A., Marmur, A., Klein, M., Kim, E., Russell, A. G., Sarnat, S. E., Mulholland, J. A., Hopke, P. K., and Tolbert, P. E.: Fine Particle Sources and Cardiorespiratory Morbidity: An Application of Chemical Mass Balance and Factor Analytical Source-Apportionment Methods, Environmental Health Perspectives, 116, 459-466, doi:10.1289/ehp.10873, 2008.

Viana, M., Kuhlbusch, T. A. J., Querol, X., Alastuey, A., Harrison, R. M., Hopke, P. K., Winiwarter, W., Vallius, M., Szidat, S., Prévôt, A. S. H., Hueglin, C., Bloemen, H., Wåhlin, P., Vecchi, R., Miranda, A. I., Kasper-Giebl, A., Maenhaut, W., and Hitzenberger, R.: Source apportionment of particulate matter in Europe: A review of methods and results, Journal of Aerosol Science, 39, 827-849, https://doi.org/10.1016/j.jaerosci.2008.05.007, 2008.

Wang, Q., Shao, M., Zhang, Y., Wei, Y., Hu, M., and Guo, S.: Source apportionment of fine organic aerosols in Beijing, Atmos. Chem. Phys., 9, 8573-8585, 10.5194/acp-9-8573-2009, 2009.

Xu, J., Srivastava, D., Wu, X., Hou, S., Vu, Tuan V., Liu, D., Sun, Y., Vlachou, A., Moschos, V., Salazar, G., Szidat, S., Prévôt, A. S. H., Fu, P., Harrison, R. M., and Shi, Z.: An evaluation of source apportionment of fine OC and PM2.5 by multiple methods: APHH-Beijing campaigns as a case study, Faraday Discussions, 10.1039/D0FD00095G, 2021.

Yin, J., Cumberland, S. A., Harrison, R. M., Allan, J., Young, D. E., Williams, P. I., and Coe, H.: Receptor modelling of fine particles in southern England using CMB including comparison with AMS-PMF factors, Atmos. Chem. Phys., 15, 2139-2158, 10.5194/acp-15-2139-2015, 2015.

Zheng, M., Salmon, L. G., Schauer, J. J., Zeng, L., Kiang, C. S., Zhang, Y., and Cass, G. R.: Seasonal trends in PM2.5 source contributions in Beijing, China, Atmospheric Environment, 39, 3967-3976, https://doi.org/10.1016/j.atmosenv.2005.03.036, 2005.

Comment 7 - Line 99 and foll.: The list of source profiles is redundant here as it is mentioned in the methodological section.

**Response:** The list of source profiles has been deleted here. The original sentence “To ensure that the source profiles used in the CMB model are representative, we mainly selected those studies which had been based in China: straw burning (Zhang et al., 2007b), wood burning (Wang et al., 2009), gasoline and diesel vehicles (Cai et al., 2017), industrial and residential coal combustion (Zhang et al., 2008), and cooking (Zhao et al., 2015).” has been changed to “To ensure that the source profiles used in the CMB model are representative, we mainly selected those studies which had been based in China.”. Please see **lines 109-111** in the revised manuscript.

Comment 8 - Line 111: Did you check if the number of samples is enough to describe the average conditions in the studied area?

**Response:** Yes, we have investigated how representative the campaign periods were of the selected seasons in Beijing by comparing pollutant levels with those from the same period each year over the 2013–2017 period. The NAQPMS model was run for the full 5-year period driven by NCEP meteorology and using temporally varying emissions for a single year that is broadly representative of 2013 conditions. The same emissions were used each year so that the meteorological contribution to pollutant levels could be assessed. This provides important information that cannot be obtained from the monitoring data (as emission varies year by year). The results from NAQPMS model showed that winter 2016 was broadly typical of the 5-year period, with similar characteristics to winter 2014. In contrast, the summer period in 2017 was cleaner than average, with $PM_{2.5}$ levels very similar to 2015, and about 25 % less than in 2013, 2014 or 2016.

We also assessed if the IAP air quality was broadly representative of the wider Beijing megacity, air quality parameters at the 12 national air quality stations around Beijing were correlated with each other. More details can be found in Shi et al. (2019).

Comment 9 - Line 137: Federal Reference Method of which country? Please, provide a reference.

**Response:** It is the Federal Reference Method of the United States, and references are provided as suggested.

The original sentence “Online $PM_{2.5}$ were determined by the TEOM FDMS 1405-DF instrument at IAP with filter equilibrating and weighing conditions comparable with the Federal Reference Method (RH: 30-40%; temperature; 20-23°C).” has been modified as “Online $PM_{2.5}$ was determined by the TEOM FDMS 1405-DF instrument at IAP with filter equilibrating and weighing conditions comparable with the United States Federal Reference Method (RH: 30-40%; temperature; 20-23°C) (Le et al., 2020; U.S.EPA, 2016).” Please see **lines 148-151** in the revised manuscript.

Added references:

Le, T.-C., Shukla, K. K., Chen, Y.-T., Chang, S.-C., Lin, T.-Y., Li, Z., Pui, D. Y. H., and Tsai, C.-J.: On the concentration differences between PM2.5 FEM monitors and FRM samplers, Atmospheric Environment, 222, 117138, https://doi.org/10.1016/j.atmosenv.2019.117138, 2020.

U.S.EPA: Monitoring PM2.5 in ambient air using designated reference or class I equivalent methods, in: Quality Assurance Handbook for Air Pollution Measurement Systems, 2-12, https://www3.epa.gov/ttnamti1/files/ambient/pm25/qa/m212.pdf, 2016.

Comment 10 - Line 142: EUSAAR2 method, a reference is needed.

**Response:** Please see the revised sentence below for added references:

“A 1.5 $cm^2$ punch from each large quartz filter sample was taken for organic carbon (OC) and elemental carbon (EC) measurements by a thermal/optical carbon analyzer (model RT-4, Sunset Laboratory Inc., USA) based on the EUSAAR2 (European Supersites for Atmospheric Aerosol Research) transmittance protocol (Cavalli et al., 2010; Chen et al., 2015).”

Please see **lines 154-158** in the revised manuscript.

Added reference:

Cavalli, F., Viana, M., Yttri, K. E., Genberg, J., and Putaud, J. P.: Toward a standardised thermal-optical protocol for measuring atmospheric organic and elemental carbon: the EUSAAR protocol, Atmos. Meas. Tech., 3, 79-89, 10.5194/amt-3-79-2010, 2010.

Chen, L. W. A., Chow, J. C., Wang, X. L., Robles, J. A., Sumlin, B. J., Lowenthal, D. H., Zimmermann, R., and Watson, J. G.: Multi-wavelength optical measurement to enhance thermal/optical analysis for carbonaceous aerosol, Atmos. Meas. Tech., 8, 451-461, 10.5194/amt-8-451-2015, 2015.

Comment 11 - Line 215: How did you check that no relevant sources are missing?

**Response:** The missing sources have been checked and discussed in the manuscript.

Firstly, as mentioned in section 3.3.6 “Other OC”, there are four major source categories of OC in Beijing based on the Multi-resolution Emission Inventory for China (MEIC), which include power, industry, residential and transportation. In our study, the OC sources applied in the CMB model were biomass (straw and wood) burning, gasoline and diesel vehicles, industrial and residential coal combustion, cooking and vegetative detritus, which included the four major source categories of OC in Beijing as described above.

Secondly, we also mentioned in section 3.3.6 that industrial coal combustion has been resolved by the CMB model, but there are industrial processes related emissions of OC which have not been resolved by the CMB model. The annual average OC emissions from these sources in Beijing were 1161 and 1083 tonnes in 2016 and 2017 respectively, which accounted for 7.7% and 9.0% of the total OC emissions (POC). Therefore, the contribution from industrial processes to the total OC in the atmosphere (POC+SOC) was considered relatively small.

Thirdly, because the CMB model explained the majority of fine OC in winter (75.7%) and summer (56.1%). In addition, the Other (unexplained) OC was found correlated with the secondary OC (SOC) estimated by the EC-tracer method, with correlation coefficients ($R^2$) of 0.58 and 0.73, and slopes of 1.16 and 0.80 in winter and summer, respectively. Hence, we believe the Other OC in this study is likely to be a mixture of predominantly SOC and a small portion of POC from missing sources such as industrial processes.

Comment 12 - Line 215: The used source profiles should be provided as table (e.g. suppl. info). Did you check that the sources do not vary considerably within the sampling interval (e.g. winter, summer).

**Response:** We didn't provide the source profiles in this study because the same source profiles have already been provided in Table S1 of Wu et al. (2020). To be more informative as suggested by the reviewer, we have added the following sentence in section 2.4:

"The source profiles with EC and organic tracers used in the CMB model were provided in Table S1 of Wu et al. (2020)." Please see **lines 234-235** in the revised manuscript.

Regarding the sources in Beijing during the sampling interval, as mentioned above, we have included the major sources of aerosols in Beijing, the CMB model explained the majority of fine OC in winter (75.7%) and summer (56.1%). Zheng et al. (2005) studied the seasonal trends in $PM_{2.5}$ source contributions in Beijing, and the major sources for organics in different seasons were the same as in this study. If the source contributions varied significantly between winter and summer, this would be reflected in the source apportionment results. If the sources types varied significantly, then the missing sources will cause less explained OC, and the Other OC would be poorly correlated with SOC by other methods. Hence, regarding source types changing within a season, we believe that we have included all relevant major source types in our CMB model. Source contributions changed significantly from day-to-day, largely due to variations in meteorology, as seen in the outputs of the CMB, but our study focuses on seasonal campaign averages. The representativeness of the meteorological conditions during the campaigns are given in Shi et al., (2019).

Reference:

Wu, X., Chen, C., Vu, T. V., Liu, D., Baldo, C., Shen, X., Zhang, Q., Cen, K., Zheng, M., He, K., Shi, Z., and Harrison, R. M.: Source apportionment of fine organic carbon (OC) using receptor modelling at a rural site of Beijing: Insight into seasonal and diurnal variation of source contributions, Environmental Pollution, 266, 115078, https://doi.org/10.1016/j.envpol.2020.115078, 2020.

Comment 13 - Line 256 -257: The period does not match the one described in section

2.5 to compare the results of the two studies you have to align the two time windows.

**Response:** As mentioned in section 2.5, the AMS data were missing during the period 09th - 15th November 2016 due to the malfunction of the AMS. Hence, the period of the CMB study did not match exactly the one described in section 2.5 for AMS-PMF analysis. The AMS data were collected from 16th November to 11th December 2016 and 22nd May to 24th June 2017, while the CMB results were from 9th November to 11th December 2016 and 22nd May to 24th June 2017. The summer periods for both studies were the same. In order to compare the results of the two studies, we have added the following sentence in section 3.5 (**lines 633-635** in the revised manuscript):

"As the AMS data were missing during the period 09th - 15th November 2016, the comparison of the AMS-PMF and CMB results for this period has been excluded."

In addition, the source contributions in winter CMB results have been modified in section 3.5 as well. Because the changes were small, they did not affect the general discussion. Please see **lines 643, 657-658, 664-669, 682** in the revised manuscript for revised CMB source contributions.

Comment 14 - Line 260: What's the importance of distinguishing haze and non-haze and how was the threshold between the two defined?

**Response:**

Distinguishing haze and non-haze days in this study is important, because the comparison of source contributions between the two can indicate possible major contributors for the haze formation, shown as the elevated sources (for example, industrial and residential coal combustion) during haze period in Table 2. The haze and non-haze days were defined based on the $PM_{2.5}$ threshold of 75 µg $m^{-3}$, which is the National Ambient Air Quality Standard Grade II of the limit for 24-hour average $PM_{2.5}$ concentration.

The original sentence "The winter sampling period was divided as haze (daily $PM_{2.5}$ > 75 µg $m^{-3}$) and non-haze days (<75 µg $m^{-3}$)." has been modified as "The winter sampling period was divided into haze (daily $PM_{2.5}$ > 75 µg $m^{-3}$) and non-haze days (<75 µg $m^{-3}$), based on the National Ambient Air Quality Standard Grade II of the limit for 24-hour average $PM_{2.5}$ concentration. The differentiation between haze and non-haze days enabled us to study the major sources contributing to the haze formation.". Please see **lines 270-274** in the revised manuscript.

Comment 15 - Line 280: Correlation could be also caused by dispersion- dilution mechanisms.

**Response:** The original sentence "Additionally, OC and EC were well-correlated in this study, with $R^2$ values of 0.85 and 0.63 during winter and summer, respectively, suggesting similar sources of carbonaceous aerosols, especially in winter." has been revised as "Additionally, OC and EC were well-correlated in this study, with $R^2$ values of 0.85 and 0.63 during winter and summer, respectively, suggesting similar paths of OC and EC dispersion and dilution, and/or similar sources of carbonaceous aerosols, especially in winter.”. Please see **lines 294-297** in the revised manuscript.

Comment 16 - Line 303: This statement is not supported by the data. The agreement is only good in winter for offline and in summer for online.

**Response:** In order to be more precise, we changed the original sentence “As shown in Fig. 3, measured offline/online $PM_{2.5}$ generally agrees well with the reconstructed $PM_{2.5}$.” to “As shown in Fig. 3, measured offline/online $PM_{2.5}$ were moderately well correlated with the reconstructed $PM_{2.5}$ with slopes of 0.77~1.26 and $R^2$ of 0.67~0.96.”. Please see **lines 317-318** in the revised manuscript.

Comment 17 - Line 312 and foll.: This behavior could also be explained by the loss of semivolatiles from PTFE filters

**Response:**

We accept this point, and hence the original sentence “This could be due to the positive artefacts of quartz filters for chemical analyses, which can absorb more organics than PTFE filters that are used for PM weighing.” has been modified as “This could be due to the loss of semi-volatile compounds from PTFE filters or the positive artifacts of quartz filters for chemical analyses, which can absorb more organics than PTFE filters that are used for PM weighing. To avoid loss of semi-volatiles, all collected samples were stored in cold conditions, including during shipment.”. Please see **lines 327-331** in the revised manuscript.

Comment 18 - Line 326 and foll.: Section 3.3 seems not to be in line with the objective of the study. In the introduction you claim that the study focuses on sources of $PM_{2.5}$ with particular reference to SOA. However, the focus of the study is on OC sources.

**Response:** To be consistent, we have modified the original sentence in the introduction “The results of this study are discussed and compared with the results from Aerosol Mass Spectrometer-PMF analysis (AMS-PMF) (Ulbrich et al., 2009; Elser et al., 2016) to improve our understanding of different sources of $PM_{2.5}$, especially for secondary organic aerosols.” as “The objectives of this study are: 1) to quantify the contributions of pollution sources to OC by applying a CMB model and compare them with those at a rural site of Beijing; 2) to compare the source apportionment results by CMB with those from Aerosol Mass Spectrometer-PMF analysis (AMS-PMF), to improve our understanding of different sources of OC.”. Please see **lines 111-115** in the revised manuscript.

Comment 19 - Line 357: Coal cannot be considered a single contributor but as a single fuel type.

**Response:** The original sentence “In this study, coal combustion is the single largest contributor to primary OC in both winter and summer.” has been revised as “In this study, coal combustion is the single largest source that contributed to primary OC in both winter and summer.”. Please see **lines 377-379** in the revised manuscript.

Comment 20 - Line 370: $Cl^-$ has many sources, not a good candidate for tracer. Line 372: In Fig 5 there is no clear relationship between $Cl^-$ and coal combustion in summer.

**Response:** We agree with the reviewer that $Cl^-$ has many sources such as sea-salt, coal combustion and biomass burning, and is not a good candidate as a tracer for coal combustion. The reasons for no clear relationship between $Cl^-$ and coal combustion in summer are discussed in the revised manuscript. Please see details below.

Beijing is an inland city, so the contribution of marine aerosols to particulate $Cl^-$ is considered minor. This is also supported by the higher $Cl^-/Na^+$ mass ratios in winter (10.1±4.8) and summer (2.7±1.8) than sea water (1.81), indicative of significant contributions from anthropogenic sources (Bondy et al., 2017). Yang et al. (2018) also reported that the contribution of sea-salt aerosol to fine particulate chloride was negligible in inland areas even during summer. Hence, particulate $Cl^-$ in this study was mainly from anthropogenic sources. The moderately good correlation of $Cl^-$ and coal combustion OC in winter could be due to enhanced coal combustion activities in this season. No correlation of $Cl^-$ and coal combustion OC in summer was a result of less coal combustion, and the abundance of $Cl^-$ in summer was more influenced by other sources. Therefore, we have modified the discussion accordingly.

The original discussion:

"Chloride has been considered as a tracer for coal combustion (Chen et al., 2014). The time series of OC from coal combustion (OC-CC) and $Cl^-$ during winter and summer of Beijing are shown in Fig. 5. OC-CC and $Cl^-$ exihibited similar trends in both seasons. The correlation coefficient ($R^2$) between OC-CC and $Cl^-$ during winter was 0.62 but there is no significant correlation between the two during the summer campaign. This is probably related to the semi-volatility of ammonium chloride, which is liable to evaporate in summer (Pio and Harrison, 1987). A similar phenomenon has been observed in Delhi (Pant et al., 2015). "

The revised discussions:

"Coal combustion is also a major source for particulate chloride (Chen et al., 2014). Because Beijing is an inland city, the contribution of marine aerosols to particulate $Cl^-$ is considered minor, which is also supported by the higher $Cl^-/Na^+$ mass ratios in winter (10.1±4.8) and summer (2.7±1.8) than sea water (1.81), indicative of significant contributions from anthropogenic sources (Bondy et al., 2017). Yang et al. (2018) also reported that the contribution of sea-salt aerosol to fine particulate chloride was negligible in China inland areas even during summer. Hence, $Cl^-$ in this study was mainly from anthropogenic sources. The time series of OC from coal combustion (OC-CC) and $Cl^-$ during winter and summer of Beijing are shown in Fig. 5. OC-CC and $Cl^-$ exhibited similar trends in both seasons. The correlation coefficient ($R^2$) between OC-CC and $Cl^-$ during winter was 0.62, which could be attributed to enhanced coal combustion activities in this season. No significant correlation between the two was found during the summer campaign, indicating the abundance of $Cl^-$ in summer was more influenced by other sources, probably including biomass burning. In addition, due to the semi-volatility of ammonium chloride, it is liable to evaporate in summer (Pio and Harrison, 1987). A similar phenomenon has been observed in Delhi (Pant et al., 2015). ”

Please see **lines 391-407** in the revised manuscript.

Added reference:

Bondy, A. L., Wang, B., Laskin, A., Craig, R. L., Nhliziyo, M. V., Bertman, S. B., Pratt, K. A., Shepson, P. B., and Ault, A. P.: Inland Sea Spray Aerosol Transport and Incomplete Chloride Depletion: Varying Degrees of Reactive Processing Observed during SOAS, Environ. Sci. Technol., 51, 9533-9542, 10.1021/acs.est.7b02085, 2017.

Yang, X., Wang, T., Xia, M., Gao, X., Li, Q., Zhang, N., Gao, Y., Lee, S., Wang, X., Xue, L., Yang, L., and Wang, W.: Abundance and origin of fine particulate chloride in continental China, Science of The Total Environment, 624, 1041-1051, https://doi.org/10.1016/j.scitotenv.2017.12.205, 2018.

Comment 21 - Line 529: Provide more information about this site. Why is it important for this study?

**Response:**

More information about this site has been provided in section 2.1 “Aerosol sampling”. The original discussion “The location of a rural site in Beijing - Pinggu during the APHH-China campaigns is also shown in Fig. 1. Other information regarding the sampling site is described elsewhere (Shi et al., 2019).” has been modified as “The location of a rural site in Beijing - Pinggu during the APHH-China campaigns is also shown in Fig. 1. The rural site in Xibaidian village in Pinggu is about 60 km away from IAP and 4 km north-west of the Pinggu town centre. It is surrounded by trees and farmland with several similar small villages nearby. A provincial highway is approximately 500 m away on its eastside running north-south. This site is far from industrial sources and located in a residential area. Other information regarding the sampling site is described elsewhere (Shi et al., 2019).” Please see **lines 125-131** in the revised manuscript.

Both IAP and Pinggu sites were chosen in the APHH-Beijing programme for the study of emission sources, atmospheric processes and health effects of air pollution in Beijing.

It is valuable to study both rural and urban sites, as both exceed health-based guidelines and require evidence-based mitigation policies which may differ depending on the source apportionment at each. Furthermore, urban air pollution may affect the pollution levels in rural areas (Chen et al., 2020), and domestic heating and cooking led to high emissions of particles and precursor gases, which may contribute to air pollution in the cities (Liu et al., 2021). Hence, to address the importance of including the rural site, the original discussion in section 3.4 “The OC source apportionment results in this study are also compared with those in another study conducted at a rural site of Beijing - Pinggu during APHH-Beijing campaigns (Wu et al., 2020). CMB was run based on the results from high-time resolution $PM_{2.5}$ samples that were collected in Pinggu during the same sampling period, but not on identical days. The comparison results are presented in Table 3.” has been revised as

"The OC source apportionment results in this study are also compared with those in another study conducted at a rural site of Beijing - Pinggu during APHH-Beijing campaigns (Wu et al., 2020). CMB was run based on the results from high-time resolution $PM_{2.5}$ samples that were collected in Pinggu during the same sampling period, but not on identical days. It is valuable to study both rural and urban sites, as both exceed health-based guidelines and require evidence-based mitigation policies which may differ depending on the source apportionment at each. Furthermore, urban air pollution may affect the pollution levels in rural areas (Chen et al., 2020), and domestic heating and cooking led to high emissions of particles and precursor gases, which may contribute to air pollution in the cities (Liu et al., 2021). The comparison of results is presented in Fig. 7 and Table S3." Please see **lines 558-568** in the revised manuscript.

Reference:

Chen, Y., Shi, G., Cai, J., Shi, Z., Wang, Z., Yao, X., Tian, M., Peng, C., Han, Y., Zhu, T., Liu, Y., Yang, X., Zheng, M., Yang, F., Zhang, Q., and He, K.: Simultaneous measurements of urban and rural particles in Beijing – Part 2: Case studies of haze events and regional transport, Atmospheric Chemistry and Physics, 20, 9249-9263, 10.5194/acp-20-9249-2020, 2020.

Liu, L., Zhang, J., Zhang, Y., Wang, Y., Xu, L., Yuan, Q., Liu, D., Sun, Y., Fu, P., Shi, Z., and Li, W.: Persistent residential burning-related primary organic particles during wintertime hazes in North China: insights into their aging and optical changes, Atmos. Chem. Phys., 21, 2251-2265, 10.5194/acp-21-2251-2021, 2021.

Shi, Z., Vu, T., Kotthaus, S., Harrison, R. M., Grimmond, S., Yue, S., Zhu, T., Lee, J., Han, Y., Demuzere, M., Dunmore, R. E., Ren, L., Liu, D., Wang, Y., Wild, O., Allan, J., Acton, W. J., Barlow, J., Barratt, B., Beddows, D., Bloss, W. J., Calzolai, G., Carruthers, D., Carslaw, D. C., Chan, Q., Chatzidiakou, L., Chen, Y., Crilley, L., Coe, H., Dai, T., Doherty, R., Duan, F., Fu, P., Ge, B., Ge, M., Guan, D., Hamilton, J. F., He, K., Heal, M., Heard, D., Hewitt, C. N., Hollaway, M., Hu, M., Ji, D., Jiang, X., Jones, R., Kalberer, M., Kelly, F. J., Kramer, L., Langford, B., Lin, C., Lewis, A. C., Li, J., Li, W., Liu, H., Liu, J., Loh, M., Lu, K., Lucarelli, F., Mann, G., McFiggans, G., Miller, M. R., Mills, G., Monk, P., Nemitz, E., O'Connor, F., Ouyang, B., Palmer, P. I., Percival, C., Popoola, O., Reeves, C., Rickard, A. R., Shao, L., Shi, G., Spracklen, D., Stevenson, D., Sun, Y., Sun, Z., Tao, S., Tong, S., Wang, Q., Wang, W., Wang, X., Wang, X., Wang, Z., Wei, L., Whalley, L., Wu, X., Wu, Z., Xie, P., Yang, F., Zhang, Q., Zhang, Y., Zhang, Y., and Zheng, M.: Introduction to the special issue "In-depth study of air pollution sources and processes within Beijing and its surrounding region (APHH-Beijing)", Atmos. Chem. Phys., 19, 7519-7546, 10.5194/acp-19-7519-2019, 2019.

Comment 22 - Table 3: It is too complicated to read, I suggest to convert it into a bar plot with error bars

**Response:** The original Table 3 has been moved to Table S3 in the revised supplemental file, and the data is converted into bar plots with error bars. Please see **lines 588-590** in the revised manuscript for added new Fig. 7.

Comment 23 - Line 556: More discussion is needed about the different size fractions used in the two methods and how this affects the single sources.

**Response:** More discussion is added in the revised manuscript.

Original discussion:

"Results from AMS-PMF were compared with the CMB source apportionment results to investigate the consistency and potential uncertainties of both methods, and also to provide supplemental source apportionment results."

Revised discussion:

"Results from AMS-PMF were compared with the CMB source apportionment results to investigate the consistency and potential uncertainties of both methods, and also to provide supplemental source apportionment results. Similar comparisons have yielded valuable insights in earlier studies (Aiken et al., 2009; Yin et al., 2015). It is noteworthy that the CMB model was applied to $PM_{2.5}$ samples, while AMS-PMF was applied for NR-$PM_1$ species. This may consequently cause differences in the chemical composition and source attribution between the two methods, as larger particles were not captured by the AMS. However, as mentioned in the study of Aiken et al. (2009), the mass concentration between $PM_1$ and $PM_{2.5}$ was small with a reduced fraction of OA and increased fraction of dust. In addition, OC fractions in fine particles were found mostly concentrated in particles <1 μm (Chen et al., 2020; Zhang et al., 2018; Tian et al., 2021). Hence, the bias was expected to be relatively small." Please see **lines 592-604** in the revised manuscript.

Added references:

Aiken, A. C., Salcedo, D., Cubison, M. J., Huffman, J. A., DeCarlo, P. F., Ulbrich, I. M., Docherty, K. S., Sueper, D., Kimmel, J. R., Worsnop, D. R., Trimborn, A., Northway, M., Stone, E. A., Schauer, J. J., Volkamer, R. M., Fortner, E., de Foy, B., Wang, J., Laskin, A., Shutthanandan, V., Zheng, J., Zhang, R., Gaffney, J., Marley, N. A., Paredes-Miranda, G., Arnott, W. P., Molina, L. T., Sosa, G., and Jimenez, J. L.: Mexico City aerosol analysis during MILAGRO using high resolution aerosol mass spectrometry at the urban supersite (T0) – Part 1: Fine particle composition and organic source apportionment, Atmos. Chem. Phys., 9, 6633-6653, 10.5194/acp-9-6633-2009, 2009.

Chen, C., Zhang, H., Li, H., Wu, N., and Zhang, Q.: Chemical characteristics and source apportionment of ambient PM1.0 and PM2.5 in a polluted city in North China plain, Atmospheric Environment, 242, 117867, https://doi.org/10.1016/j.atmosenv.2020.117867, 2020.

Tian, Y., Feng, Y., Liang, Y., Li, Y., Xue, Q., Shi, Z., Xu, J., and Harrison, R. M.: Size distributions of inorganic and organic components in particulate matter from a megacity in northern China: dependence upon seasons and pollution levels (under review), 2020, Environmental Pollution.

Yin, J., Cumberland, S. A., Harrison, R. M., Allan, J., Young, D. E., Williams, P. I., and Coe, H.: Receptor modelling of fine particles in southern England using CMB including comparison with AMS-PMF factors, Atmos. Chem. Phys., 15, 2139-2158, 10.5194/acp-15-2139-2015, 2015.

Zhang, Y., Lang, J., Cheng, S., Li, S., Zhou, Y., Chen, D., Zhang, H., and Wang, H.: Chemical composition and sources of PM1 and PM2.5 in Beijing in autumn, Science of The Total Environment, 630, 72-82, https://doi.org/10.1016/j.scitotenv.2018.02.151, 2018.

Comment 24 - Line 564: What is the meaning of OOA1, 2 and 3? The profiles of the AMS sources should be shown in a figure

**Response:** Based on diurnal cycles, mass spectra and the correlations between OA factors and other measured species, three SOA factors were identified by PMF. These SOA factors showed significantly elevated O/C ratios (0.67-1.48), and correlated well with SIA (R=0.52-0.69). Hence, OOA1, OOA2 and OOA3 represent three types of SOA.

Please added discussions in **lines 611-625** in the revised manuscript, which are also copied below:

"These OOA factors were identified by PMF based on diurnal cycles, mass spectra and the correlations between OA factors and other measured species. Three OOA factors showed significantly elevated O/C ratios (0.67-1.48), and correlated well with SIA (R=0.52-0.69). Hence, OOA1, OOA2 and OOA3 represent three types of SOA. Compared to OOA2 and OOA3, OOA1 showed relatively higher f43 (fraction of m/z 43 in OA). In addition, the concentrations of OOA1 and OOA3 were higher in daytime, implying the effect of photochemical processing. The variations of OOA2 tracked well with $C_2H_2O_2^+$ (R=0.89), an aqueous-processing related fragment ion (Sun et al., 2016), indicating that OOA2 was an OA factor associated with aqueous-phase processing. Previous studies suggested that aqueous-phase processing plays an important role in the formation of nitrogen-containing compounds (Xu et al., 2017). The fact that OOA2, with relatively high N/C ratios (0.046), was correlated with several N-containing ions (e.g. $CH_4N^+$, $C_2H_6N^+$, R=0.71-0.77) further supports the above argument. The factor profiles of AMS-PMF in winter and summer are provided in Figs. S5 and S6, respectively.".

Comment 25 - Table 4: Please, add the names to the single columns.

**Response:** The names of the single columns have been added in the revised Table 4.

Comment 26 - Line 631 and foll.: This calculation of $PM_{2.5}$ contributions using $OC/PM_{2.5}$ ratios from the literature introduces a considerable amount of uncertainty because of the variability of such factors. The uncertainty of these results should be clearly illustrated to the reader.

**Response:** We agree with the reviewer that the calculation of $PM_{2.5}$ contributions using $OC/PM_{2.5}$ ratios from the applied source profiles introduces a considerable amount of uncertainty. Hence, apart from the discussion in the original manuscript, we also added more discussion regarding the uncertainties of the results.

Original discussion:

"Instead of OC/$PM_{2.5}$, applying an OM/OC ratio for the calculation may result in an underestimation of $PM_{2.5}$ source contributions, because sources like cooking and vegetative detritus can also emit inorganic pollutants. However, cooking emissions are mostly organic and the contribution from vegetative detritus to $PM_{2.5}$ is very small, their effects on source contribution estimation here are considered negligible."

Modified discussion:

"Due to the variability of the OC/ $PM_{2.5}$ ratio in the source profiles, the application using the average OC/ $PM_{2.5}$ ratio of each source to convert the OC to $PM_{2.5}$ in all samples may be subject to uncertainties, as both organic species and $PM_{2.5}$ mass measurements are subject to analytical imprecision. Unfortunately, insufficient data are available for a formal analysis of uncertainty, but errors of around +/- 10% seem very probable. In addition, instead of OC/$PM_{2.5}$, applying an OM/OC ratio to cooking and vegetative detritus sources for the calculation may result in an underestimation of $PM_{2.5}$ source contributions from these sources, because they can also emit inorganic pollutants. However, cooking emissions are mostly organic and the contribution from vegetative detritus to $PM_{2.5}$ is very small, so their effects on source contribution estimation here are considered negligible."

Please see **lines 711-722** in the revised manuscript.

Comment 27 - Lines 633-638: It is not necessary to repeat the information that is already available in Table S3.

**Response:** Please see the modification below:

Original text:

"For cooking, an OM/OC ratio of 1.4 was applied (Zhao et al., 2007). For vegetative detritus, OM/OC ratio of 2.1 was applied (Bae et al., 2006b). The OM/OC ratios for oxygenated OA were in the range of 1.85-2.3 (Zhang et al., 2005; Aiken et al., 2008), and the OM/OC ratio was 2.17 in secondary organic aerosols of $PM_{2.5}$ (Bae et al., 2006a). Therefore, an OM/OC ratio of 2.2 is applied in this study to convert the Other OC to OM."

Modified text:

"For cooking, vegetative detritus and secondary organic aerosols, OM/OC ratios were applied considering the low contribution of inorganic species to $PM_{2.5}$ mass from these sources (Zhao et al., 2007; Bae et al., 2006b). The OM/OC ratios for oxygenated OA were in the range of 1.85-2.3 (Zhang et al., 2005; Aiken et al., 2008), and the OM/OC ratio was 2.17 in secondary organic aerosols of $PM_{2.5}$ (Bae et al., 2006a). Therefore, an OM/OC ratio of 2.2 is applied in this study to convert the Other OC to OM."

Please see **lines 705-711** in the revised manuscript.

Comment 28 - Line 649. Specify what does the range after the value means. Is it a confidence interval, a standard deviation or something else? Does it include the uncertainty of the OC/$PM_{2.5}$ ratios?

**Response:** The value in the sentence "As shown in Table S4, $PM_{2.5}$ mass was well explained by those sources which accounted for 91.9±24.1% and 99.0±19.1% of online $PM_{2.5}$ in winter and summer, respectively." represents "mean±SD of the daily values", which is added in Table S5 in the revised manuscript to avoid any confusion. Because the uncertainties of $OC/PM_{2.5}$ ratios were not known, they were not considered in the calculation. The possible uncertainties of using $OC/PM_{2.5}$ ratios have been discussed in the revised manuscript, which are also illustrated above.

Comment 29 - Line 652 and foll.: Since these results are in a table it is not necessary to list them here.

**Response:** Please see modifications below:

The original text:

"On average, the source contributions in winter ranked as SNA (30.5 μg $m^{-3}$, 34.1% of reconstructed $PM_{2.5}$ and hereafter), coal combustion (industrial & residential CC; 17.7 μg $m^{-3}$, 21.4%), Other OM (14.6 μg $m^{-3}$, 14.8%), biomass burning (8.9 μg $m^{-3}$, 11.0%), gasoline & diesel (5.6 μg $m^{-3}$, 7.5%), geological minerals (5.3 μg $m^{-3}$, 7.0%), cooking (3.1 μg $m^{-3}$, 3.9%) and vegetative detritus (0.2 μg $m^{-3}$, 0.3%); in summer these ranked as SNA (17.7 μg $m^{-3}$, 48.5%), other OM (8.0 μg $m^{-3}$, 18.3%), coal combustion (4.7 μg $m^{-3}$, 14.6%), geological minerals (3.5 μg $m^{-3}$, 10.4%), cooking (0.9 μg $m^{-3}$, 2.8%), gasoline & diesel (0.8 μg $m^{-3}$, 2.6%), biomass burning (0.8 μg $m^{-3}$, 2.1%) and vegetative detritus (0.2 μg $m^{-3}$, 0.7%)."

The revised text:

"On average, the source contributions in winter ranked as SNA > coal combustion > Other OM > biomass burning > gasoline & diesel > geological minerals > cooking > vegetative detritus; in summer these ranked as SNA > other OM > coal combustion > geological minerals > cooking > gasoline & diesel > biomass burning > vegetative detritus."

Please see **lines 729-733** in the revised manuscript.

Comment 30 - Figure 9: I suggest to replace the daily data with the overall averages (now in table S4).

**Response:** The daily data in original Fig. 9 was replaced with the overall averages (now in Fig. 10). The text is also modified as below:

"The daily $PM_{2.5}$ contribution estimates and seasonal average source contributions are provided in Fig. S7 and Fig. 10, respectively. Detailed data and their relative abundance in the reconstructed $PM_{2.5}$ are summarized in Table S5."

Please see **lines 758-760** in the revised manuscript for revised Fig. 10 and **lines 722-724** for revised text.

Comment 31 - Conclusions: this section is mandatory according to the instructions for authors of this journal

**Response:** We have changed the "Summary" to "Conclusion" and the added value of this study with respect to the previous knowledge on $PM_{2.5}$/OC sources in Beijing are provided in **lines 778-788** in the revised manuscript, which is also copied below:

"Although the annual average $PM_{2.5}$ levels in Beijing reduced from 88 µg $m^{-3}$ in year 2013 to 58 µg $m^{-3}$ in year 2017 (Vu et al., 2019), and the deweathered concentration of $PM_1$ decreased by -38% in 2017 comparing to 2007 (Zhang et al., 2020), our CMB modelling results indicate that the coal combustion and biomass burning still remained the dominant primary OC sources in winter 2016 and summer2017, with road traffic ranked as the third highest. Cooking was a more significant source of OC than biomass burning at the urban site during summer. Compared to other CMB studies in Beijing, our study revealed an increase of the contributions from coal combustion, biomass burning and traffic to $PM_{2.5}$ in winter 2016 compared to winter 2000, while those in this study remained similar compared to winter 2013."

**Comments from Reviewer #2**

In the manuscript by Xu et al., the atmospheric fine particle samples from winter and summer at an urban site in Beijing were collected and analyzed. The source apportionment analysis from the Chemical mass balance (CMB) model shows comparable results with that of Positive Matrix Factorization (PMF) analysis of co-located Aerosol Mass Spectrometer (AMS). The paper provides useful scientific evidence to the source contributions in Beijing. However, there are still a few important issues that need to be clarified.

**Response:** Thanks for the reviewer's comments. Please see our point-by-point responses below.

Comment 1 - Major issues: One of the major issues in the paper is that the AMS analyzed the non-refractory fraction of $PM_1$, but the CMB method was applied to $PM_{2.5}$ samples. The difference in particle size segments is an important factor that could contribute significantly to the difference between the results from these two source apportionment methods. However, the authors didn't give any discussion about it. Especially, as shown in Figure 8, for the AMS-PMF derived OOA concentrations (in $PM_1$), which generally show slightly higher concentrations than other OC estimated by CMB methods (in $PM_{2.5}$). The authors have to give some reasonable explanations for this.

**Response:** We have provided more discussion regarding the size fractions of two methods, please see the modification below.

Original discussion:

"Results from AMS-PMF were compared with the CMB source apportionment results to investigate the consistency and potential uncertainties of both methods, and also to provide supplemental source apportionment results."

Revised discussion:

"Results from AMS-PMF were compared with the CMB source apportionment results to investigate the consistency and potential uncertainties of both methods, and also to provide supplemental source apportionment results (Ulbrich et al., 2009; Elser et al., 2016). Similar comparisons have yielded valuable insights in earlier studies (Aiken et al., 2009; Yin et al., 2015). It is noteworthy that the CMB model was applied to $PM_{2.5}$ samples, while AMS-PMF was applied for NR-$PM_1$ species. This may consequently cause differences in the chemical composition and source attribution between the two methods, as larger particles were not captured by the AMS. However, as mentioned in the study of Aiken et al. (2009), the mass concentration between $PM_1$ and $PM_{2.5}$ was small with a reduced fraction of OA and increased fraction of dust. In addition, OC fractions in fine particles were found mostly concentrated in particles <1 µm (Chen et al., 2020; Zhang et al., 2018; Tian et al., 2020). Hence, the bias was expected to be relatively small." Please see **lines 592-604** in the revised manuscript.

For the AMS-PMF derived OOA concentrations (in $PM_1$), which generally show slightly higher concentrations than other OC estimated by CMB methods (in $PM_{2.5}$), we have also provided more explanations in the revised manuscript, please see the modification below:

Original discussions:

"The derived OOC concentrations in winter and summer were 10.1±7.4 and 4.3±2.1 µg $m^{-3}$ in winter and summer, respectively, higher than the Other OC estimated by CMB in winter (5.3±4.9 µg $m^{-3}$) and summer (2.9±1.5 µg $m^{-3}$) in this study."

Revised discussions:

"The derived OOC-AMS concentrations in winter and summer were 10.1±7.4 and 4.3±2.1 µg $m^{-3}$ in winter and summer, respectively, higher than the Other OC estimated by CMB (Other OC-CMB) in winter (6.1±5.5 µg $m^{-3}$) and summer (2.9±1.5 µg $m^{-3}$) in this study. This could be because AMS-PMF did not resolve HOC in winter and CCOC in summer, which may be mixed with the OOA factors and lead to overestimation of OOC concentrations." Please see **lines 680-685** in the revised manuscript.

Added references:

Aiken, A. C., Salcedo, D., Cubison, M. J., Huffman, J. A., DeCarlo, P. F., Ulbrich, I. M., Docherty, K. S., Sueper, D., Kimmel, J. R., Worsnop, D. R., Trimborn, A., Northway, M., Stone, E. A., Schauer, J. J., Volkamer, R. M., Fortner, E., de Foy, B., Wang, J., Laskin, A., Shutthanandan, V., Zheng, J., Zhang, R., Gaffney, J., Marley, N. A., Paredes-Miranda, G., Arnott, W. P., Molina, L. T., Sosa, G., and Jimenez, J. L.: Mexico City aerosol analysis during MILAGRO using high resolution aerosol mass spectrometry at the urban supersite (T0) – Part 1: Fine particle composition and organic source apportionment, Atmos. Chem. Phys., 9, 6633-6653, 10.5194/acp-9-6633-2009, 2009.

Chen, C., Zhang, H., Li, H., Wu, N., and Zhang, Q.: Chemical characteristics and source apportionment of ambient PM1.0 and PM2.5 in a polluted city in North China plain, Atmospheric Environment, 242, 117867, https://doi.org/10.1016/j.atmosenv.2020.117867, 2020.

Tian, Y., Feng, Y., Liang, Y., Li, Y., Xue, Q., Shi, Z., Xu, J., and Harrison, R. M.: Size distributions of inorganic and organic components in particulate matter from a megacity in northern China: dependence upon seasons and pollution levels (under review), Environmental Pollution, 2020.

Yin, J., Cumberland, S. A., Harrison, R. M., Allan, J., Young, D. E., Williams, P. I., and Coe, H.: Receptor modelling of fine particles in southern England using CMB including comparison with AMS-PMF factors, Atmos. Chem. Phys., 15, 2139-2158, 10.5194/acp-15-2139-2015, 2015.

Zhang, Y., Lang, J., Cheng, S., Li, S., Zhou, Y., Chen, D., Zhang, H., and Wang, H.: Chemical composition and sources of PM1 and PM2.5 in Beijing in autumn, Science of The Total Environment, 630, 72-82, https://doi.org/10.1016/j.scitotenv.2018.02.151, 2018.

Comment 2 - P10, line 307-313: It is biased to attribute all deviations to the uncertainty of experimental measurements for heavily polluted periods or the sampling artifacts. Are the reconstructed $PM_{2.5}$ concentrations absolutely right?

**Response:** Please see the modifications below.

The original discussion:

"After excluding the outliers (2 outliers of offline-$PM_{2.5}$ > 200 $\mu g\ m^{-3}$ and 4 outliers of online-$PM_{2.5}$ > 170 $\mu g\ m^{-3}$), the regression results improved with both slopes and $R^2$ approaching unity (Fig. S3). This could indicate some uncertainties in offline and online $PM_{2.5}$ measurement for heavily polluted samples. During the summer campaign, the slope of the reconstructed $PM_{2.5}$ and online-$PM_{2.5}$ was close to 1, but that of reconstructed $PM_{2.5}$ and offline-$PM_{2.5}$ was 1.26. This could be due to the positive artifacts of quartz filters for chemical analyses, which can absorb more organics than PTFE filters that are used for PM weighing. The datapoints were more scattered in summer, which could result from the large difference in OM-OC relationships from day to day. The reconstructed inorganics (reconstructed $PM_{2.5}$ excluding OM) correlated well with offline-$PM_{2.5}$, but OM did not (Fig. S4)."

The revised discussion:

"After excluding the outliers (2 outliers of offline-$PM_{2.5}$ > 200 $\mu g\ m^{-3}$ and 4 outliers of online-$PM_{2.5}$ > 170 $\mu g\ m^{-3}$), the regression results improved with both slopes and $R^2$ approaching unity (Fig. S3). This could indicate some uncertainties in offline and/or online $PM_{2.5}$ measurement for heavily polluted samples, or the applied OM/OC ratio in winter was not suitable for converting OC to OM in heavily polluted samples. During the summer campaign, the slope of the reconstructed $PM_{2.5}$ and online-$PM_{2.5}$ was close to 1, but that of reconstructed $PM_{2.5}$ and offline-$PM_{2.5}$ was 1.26. This could be due to the loss of semi-volatile compounds from PTFE filters or the positive artifacts of quartz filters for chemical analyses, which can absorb more organics than PTFE filters that are used for PM weighing. To avoid loss of semi-volatiles, all collected samples were stored in cold conditions, including during shipment. The datapoints were more scattered in summer, which could result from the large difference in OM-OC relationships from day to day. The reconstructed inorganics (reconstructed $PM_{2.5}$ excluding OM) correlated well with offline-$PM_{2.5}$, but OM did not (Fig. S4). Hence, the discrepancies of between reconstructed $PM_{2.5}$ and offline/online $PM_{2.5}$ in summer may be mainly attributable to variable OM/OC ratios." Please see **lines 321-336** in the revised manuscript.

Comment 3 - Minor suggestions: Page 3, line 63-64: "A better understanding of the sources of $PM_{2.5}$ in Beijing is essential to provide scientific evidence to control the $PM_{2.5}$ pollution." Such an expression is inaccurate.

**Response:**

The original sentence "A better understanding of the sources of $PM_{2.5}$ in Beijing is essential to provide scientific evidence to control the $PM_{2.5}$ pollution." has been revised as "A better understanding of $PM_{2.5}$ sources in Beijing is essential, as it can provide important scientific evidence to develop measures to control $PM_{2.5}$ pollution.". Please see **lines 63-65** in the revised manuscript.

Comment 4 - Page 3, the last paragraph: Please pay attention to the logical order of these sentences. For example, you have to introduce the CMB method was used to do the source apportionment first, then state how the source profiles were determined. Similar mistakes were also found in P5, line 143-146.

**Response:** We have modified the paragraph to follow the logic, please see our revision below.

The original paragraph:

"In this study, $PM_{2.5}$ samples were collected at an urban site of Beijing during winter and summer 2016-2017. OC, EC, PAHs, alkanes, hopanes, fatty acids and monosaccharide anhydrides were determined. To ensure that the source profiles used in the CMB model are representative, we mainly selected those studies which had been based in China: straw burning (Zhang et al., 2007b), wood burning (Wang et al., 2009), gasoline and diesel vehicles (Cai et al., 2017), industrial and residential coal combustion (Zhang et al., 2008), and cooking (Zhao et al., 2015). Source contributions of organic carbon were examined and quantified applying the CMB model based on the source profiles mentioned above. The results of this study are discussed and compared with the results from Aerosol Mass Spectrometer-PMF analysis (AMS-PMF) (Ulbrich et al., 2009; Elser et al., 2016) to improve our understanding of different sources of $PM_{2.5}$, especially for secondary organic aerosols."

The revised paragraph:

"In this study, $PM_{2.5}$ samples were collected at an urban site of Beijing in winter 2016 and summer 2017. OC, EC, PAHs, alkanes, hopanes, fatty acids and monosaccharide anhydrides in the $PM_{2.5}$ samples were determined, and applied in the CMB model for apportioning the organic carbon sources. To ensure that the source profiles used in the CMB model are representative, we mainly selected data which had been determined in China. The objectives of this study are: 1) to quantify the contributions of pollution sources to OC by applying a CMB model and compare them with those at a rural site of Beijing; 2) to compare the source apportionment results by CMB with those from Aerosol Mass Spectrometer-PMF analysis (AMS-PMF), to improve our understanding of different sources of OC.” Please see **lines 106-115** in the revised manuscript.

P5, line 143-146: “The uncertainties from duplicate analyses of filters were <10%. Replicate analyses were conducted once every ten samples. All sample results were corrected by the values obtained from field blanks, which were 0.40 and 0.01 μg $m^{-3}$ for OC and EC, respectively.” has been revised as “Replicate analyses of OC and EC were conducted once every ten samples. The uncertainties from duplicate analyses of filters were <10%. All sample results were corrected by the values obtained from field blanks, which were 0.40 and 0.01 μg $m^{-3}$ for OC and EC, respectively.” to follow the logic order of the sentences. Please see **lines 158-161** in the revised manuscript.

Comment 5 - P5, line 159-160: Please describe exactly how much is “A portion of the filters.” So did the other analyses, such as inorganic components using Ion Chromatograph and ICP-MS in Section 2.2.3 (P6, second paragraph).

**Response:** Please see added information below.

Line 159-160: “A portion of the filters was extracted 3 times with dichloromethane/methanol (HPLC grade, v/v: 2:1) under ultrasonication for 10 minutes.” has been change to “9 $cm^2$ of the quartz filters were extracted 3 times with dichloromethane/methanol (HPLC grade, v/v: 2:1) under ultrasonication for 10 minutes.”. Please see **lines 172-174** in the revised manuscript.

“Half of the PTFE filter was extracted with 10 mL ultrapure water for the analysis of inorganic ions.” has been added in the beginning of the second paragraph on P6. Please see **lines 191-192** in the revised manuscript.

“Other elements including V, Cr, Co, Mn, Ni, Cu, Zn, As, Sr, Cd, Sb, Ba and Pb were analyzed by Inductively-coupled plasma-mass spectrometer (ICP-MS), the detection limits of them were 1.32, 0.25, 0.04, 0.06, 2.05, 1.25, 1.22, 1.74, 0.02, 0.03, 0.11, 0.06 and 0.04 ng $m^{-3}$, respectively.” has been revised as “Other elements including V, Cr, Co, Mn, Ni, Cu, Zn, As, Sr, Cd, Sb, Ba and Pb were analyzed by Inductively-coupled plasma-mass spectrometer (ICP-MS) after extraction of 1/2 PTFE filter by diluted acid mixture ($HNO_3$/HCl), and the detection limits of them were 1.32, 0.25, 0.04, 0.06, 2.05, 1.25, 1.22, 1.74, 0.02, 0.03, 0.11, 0.06 and 0.04 ng $m^{-3}$, respectively.” Please see **lines 199-203** in the revised manuscript.

Comment 6 - P7, First paragraph: Please rephrase and reorganized these sentences.

**Response:** Please find the modifications below.

The original paragraph:

“A receptor model, namely the chemical mass balance model (US EPA CMB8.2), was applied in this study to apportion the sources of OC. It utilizes a linear least squares solution and both uncertainties in source profiles and ambient measurements were taken into consideration in this model. The essential criteria in this model were met to ensure reliable fitting results. For instance, in all samples, $R^2$ were >0.80 (mostly >0.9), $Chi^2$ were <2, Tstat values were mostly greater than 2 except the source of vegetative detritus, and C/M ratios (ratio of calculated to measured concentration) for all fitting species were in range of 0.8-1.2 in this study. The source profiles applied here were mostly from local studies in China to better represent the source characteristics, including straw burning (wheat, corn, rice straw burning) (Zhang et al., 2007b), wood burning (Wang et al., 2009), gasoline and diesel vehicles (including motorcycles, light- and heavy-duty gasoline and diesel vehicles) (Cai et al., 2017), industrial and residential coal combustion (including anthracite, sub-bituminite, bituminite, and brown coal) (Zhang et al., 2008), and cooking (Zhao et al., 2015), except vegetative detritus (Rogge et al., 1993; Wang et al., 2009). Fitting species should be stable during the transport from sources to receptor site and can represent the chemical characteristics of the sources (Wang et al., 2009). The selected fitting species were EC, levoglucosan, palmitic acid, stearic acid, fluoranthene, phenanthrene, retene, benz(a)anthracene, chrysene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo[ghi]perylene, picene, 17a (H) -22, 29, 30-trisnorhopane, 17b (H), 21a (H) -norhopane and n-alkanes (C24-C33), the concentrations of which are provided in Table 1."

The modified paragraph:

"The chemical mass balance model (US EPA CMB8.2) was applied in this study to apportion the sources of OC by utilizing a linear least squares solution. Both uncertainties in source profiles and ambient measurements were taken into consideration in this model. The source profiles applied here were from local studies in China to better represent the source characteristics, including straw burning (wheat, corn, rice straw burning) (Zhang et al., 2007b), wood burning (Wang et al., 2009), gasoline and diesel vehicles (including motorcycles, light- and heavy-duty gasoline and diesel vehicles) (Cai et al., 2017), industrial and residential coal combustion (including anthracite, sub-bituminite, bituminite, and brown coal) (Zhang et al., 2008), and cooking (Zhao et al., 2015), except vegetative detritus (Rogge et al., 1993; Wang et al., 2009). The source profiles with EC and organic tracers used in the CMB model were provided in Table S1 of Wu et al. (2020). The selected fitting species were EC, levoglucosan, palmitic acid, stearic acid, fluoranthene, phenanthrene, retene, benz(a)anthracene, chrysene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo[ghi]perylene, picene, 17a (H) -22, 29, 30-trisnorhopane, 17b (H), 21a (H) -norhopane and n-alkanes (C24-C33), the concentrations of which are provided in Table 1. The essential criteria in this model were met to ensure reliable fitting results. For instance, in all samples, $R^2$ were >0.80 (mostly >0.9), $Chi^2$ were <2, Tstat values were mostly greater than 2 except the source of vegetative detritus, and C/M ratios (ratio of calculated to measured concentration) for all fitting species were in range of 0.8-1.2 in this study." Please see **line 224-244** in the revised manuscript.

Comment 7 - P7, line 241-244: Rewording, please!

**Response:**

The sentences in line 241-244 of the original manuscript "While the source apportionment of fine OC in this study was conducted by using an offline chemical speciation dataset and source profiles, the source apportionment of organics in NR-$PM_1$ was carried out by applying PMF to the high-resolution mass spectra of OA." has been revised as “The source apportionment of organics in NR-$PM_1$ was carried out by applying PMF to the high-resolution mass spectra of OA, while that of fine OC in this study was conducted by applying source profiles along with an offline chemical speciation dataset.” Please see **lines 257-260** in the revised manuscript.

Comment 8 - P18, line 560-565, and P7 line 245-251: Unnecessary to repeat this.

**Response:** To avoid repetition the text in P7 line 245-251 in the original manuscript “The PMF analysis resulted in an optimal solution of 2 primary factors in summer: traffic-related hydrocarbon-like OA (HOA) and cooking OA (COA) and 3 secondary factors of oxygenated OA (OOA): OOA1, OOA2, OOA3. In winter, 3 primary factors were identified: coal combustion OA (CCOA), COA, biomass burning OA (BBOA), and 3 secondary factors: oxidized primary OA (OPOA), less-oxidized OA (LOOOA), and more-oxidized OA (MOOOA).” has been deleted.

Comment 9 - P19-P20: when describing the comparisons between the results from CMB and AMSPMF, the language and symbols sometimes are quite confusing; please reorganize them.

**Response:** The phrases of AMS-PMF and CMB resolved OC sources are revised as suggested to avoid any confusion:

“Six factors in non-refractory (NR)-$PM_1$ from the AMS were identified based on the mass spectra measured in winter at IAP by applying a PMF model, including coal combustion OA (CCOA-AMS), cooking OA (COA-AMS), biomass burning OA (BBOA-AMS) and 3 secondary factors of oxidized primary OA (OPOA-AMS), less-oxidized OA (LOOOA-AMS), and more-oxidized OA (MOOOA-AMS). In summer, the PMF analysis resulted in 5 factors including 2 primary factors of hydrocarbon-like OA (HOA-AMS), cooking OA (COA-AMS) and 3 secondary factors of oxygenated OA (OOA-AMS): OOA1, OOA2, OOA3.” Please see **lines 604-611** in the revised manuscript.

“In order to compare with the source apportionment results of OC in this study from the CMB model, the OA concentrations from the AMS-PMF were converted to OC based on various OA/OC ratios measured in Beijing: 1.35 for CCOA/CCOC (coal combustion organic carbon), 1.31 for HOA/HOC (hydrocarbon-like organic carbon) (Sun et al., 2016), 1.38 for COA/COC (cooking organic carbon), 1.58 for BBOA/BBOC (biomass burning organic carbon) (Xu et al., 2019b), and 1.78 for OOA/OOC (Huang et al., 2010). The concentrations of OA and corresponding OC from AMS-PMF analysis are presented in Table 3. As the AMS data were missing during the period 09th - 15th November 2016, the comparison of the AMS-PMF and CMB results for this period has been excluded.

The CCOA-AMS factor was mainly characterized by m/z of 44, 73 and 115 (Sun et al., 2016). In winter, CCOA-AMS was 6.2±4.4 $\mu g\ m^{-3}$, contributing 16.9% of OM. CCOC-AMS was 4.6±3.3 $\mu g\ m^{-3}$, which was much lower than the estimated coal combustion OC (7.5±5.0 $\mu g\ m^{-3}$, industrial and residential coal combustion OC) by CMB (CCOC-CMB). The time series of CCOC-CMB and CCOC-AMS analysis in Fig. 8 showed a similar trend with relatively good correlation of $R^2$ = 0.71, but coal combustion estimated by CMB was consistently higher than by AMS-PMF, probably because AMS-PMF only resolved the sources of NR-$PM_1$, and some coal combustion particles are larger (Xu et al., 2011). The correlation coefficients ($R^2$) of CCOC-AMS with $Cl^-$ and NR-$Cl^-$ were 0.49 and 0.65, respectively in the winter data.

BBOA-AMS in winter was 6.5±5.8 µg $m^{-3}$, contributing 17.7% of OM. This BBOA-AMS factor included a high proportion of m/z 60 and 73, which are typical fragments of anhydrous sugars like levoglucosan (Srivastava et al., 2019). BBOC-AMS was 4.1±3.7 µg $m^{-3}$, which was very close to BBOC-CMB (3.72±2.79 µg $m^{-3}$, 16.4% of OC) in this study.

COA-AMS is as a common factor identified in both winter and summer results. It is characterized by high m/z of 55 and 57 in the mass spectrum (Sun et al., 2016). COA-AMS was 5.9±4.1 and 1.8±1.0 µg $m^{-3}$ in winter and summer, respectively, contributing 16.1% and 17.8% of OM. COC-AMS was 4.3±3.0 and 1.3±0.7 µg $m^{-3}$ in winter and summer, respectively, which were almost 2 times of the COC-CMB results for winter (2.20±1.97 µg $m^{-3}$) and summer (0.66±0.43 µg $m^{-3}$). Yin et al. (2015) also reported that COC-AMS was about 2 times of COC-CMB. The overestimation of COC-AMS could be due to a low relative ionization efficiency (RIE) for cooking OAs (1.4) in AMS while the actual RIE could be higher, such as 1.56-3.06 (Reyes-Villegas et al., 2018), and/or the use of a relatively low OA/OC ratio for cooking (Xu et al., 2021).

HOA-AMS was 0.7±0.4 µg $m^{-3}$ in summer, accounting for 6.9% of OM. HOA-AMS is usually identified based on the high contribution of aliphatic hydrocarbons in this factor, particularly m/z of 27, 41, 55, 57, 69 and 71 (Aiken et al., 2009). This result is lower than that (17% of OM) in rural Beijing during summer 2015 (Hua et al., 2018). HOC-AMS was 0.5±0.3 µg $m^{-3}$ in summer, which is higher than the traffic (gasoline+diesel) emitted OC (0.4±0.2 µg $m^{-3}$) from the CMB model. No obvious correlation was observed between HOC with nitrate and traffic emitted OC from the CMB model during summer.

OOA-AMS concentrations (the sum of all oxidized OA) were 18.0±13.2 and 7.6±3.7 µg $m^{-3}$ in winter and summer, respectively, accounting for 49.0% and 75.2% of OM. The derived OOC-AMS concentrations in winter and summer were 10.1±7.4 and 4.3±2.1 µg $m^{-3}$ in winter and summer, respectively, higher than the Other OC estimated by CMB (Other OC-CMB) in winter (6.1±5.5 µg $m^{-3}$) and summer (2.9±1.5 µg $m^{-3}$) in this study. This could be because AMS-PMF did not resolve HOC in winter and CCOC in summer, which may be mixed with the OOA factors and lead to overestimation of OOC concentrations. The time series and correlation of Other OC-CMB and OOC-AMS is plotted in Fig. 9. A similar temporal trend was found between them, especially in summer, which was also observed with a better correlation ($R^2$=0.73).

In summary, CMB is able to resolve almost all major known primary OA sources, but AMS-PMF can resolve more secondary OA sources. The AMS-PMF results for major components, such as CCOC-AMS and OOC-AMS agreed well with the results from CMB in the winter. However, discrepancies or poor agreement was found for other sources, such as BBOA-AMS and COA-AMS, although the temporal features were very similar. Furthermore, AMS-PMF did not identify certain sources, probably due to their relatively small contribution to particle mass. Overall, CMB and AMS-PMF offered complementary data to resolve both primary and secondary sources.”. Please see **lines 626-700** in the revised manuscript.