Response of the reviewers' comments on " How COVID-19 related policies reshaped organic aerosol source contributions in Central London" by Gang I. Chen et al.

We thank the two reviewers for all the constructive comments. The following response to the reviewers, details original review comments with normal italic font, green font for authors' responses, and *blue italic font for changes in the revised version.*

Reviewer #1

The authors addressed my comments well, and I now think this is acceptable for publication in Atmospheric Chemistry and Physics. I only have a few technical comments below. Line numbers refer to the cleaned, resubmitted version of the manuscript.

Thank you for the comments. We have addressed the comments as follows:

Line 199: Please revise "volatility of NH4NO3" to "semi-volatile nature of NH4NO3."

done

Line 201: Add "across Europe" after "… enhanced photochemistry."

done

Line 202: In addition to long-range transport, please note that airmasses originating from the sea can also affect SO4 enhancement, as mentioned in the author's response to comment #4.

Thank you for the comment, we have revised the sentence as shown below:

"During the lockdown in spring, SO_4 concentrations remained high, which was associated with long-range transport and marine aerosols (e.g., methanesulfonic acid, MSA) (Fig. S7)."

Lines 247–248: Still, seasonality is not clear as the effect of COVID-related policies from my perspective, considering the uncertainties of Q-ACSM. I suggest toning down this statement by removing "considerable."

done

Line 253-258: Please add appropriate references for this statement.

Two citations have been added as suggested.

"It's worth mentioning that the reduced POA concentrations in the warm season was not caused by reduced residential heating and energy consummation since Central London mainly uses natural gas and renewable energy rather than solid fuel combustion (Cliff et al., 2025). The OOA factor concentrations remain relatively consistent across seasons, while its contribution was larger during the warmer seasons (Fig. S4). This is because both high temperature and strong solar radiation will enhance the photochemistry and evaporation of POA sources, and increased

volatile organic compound (VOC) emissions lead to high OOA production despite the evaporation of semi-volatile OOA (Fig. S4) (Chen et al., 2022)."

Line 275: Briefly include the point you made in response to comment #13 to support the small diurnal variations observed for MO-OOA and LO-OOA.

Corrected as shown below:

"The MO-OOA showed a smaller diurnal variation compared to LO-OOA. This is because the LO-OOA is also known as semi-volatile OOA (SV-OOA), which evaporate during the day due to higher temperatures and accumulate in the evening due to the shallower boundary layer (Chen et al., 2022)."

Line 320: How about adding a dedicated section on pre-EOTHO? I assume the authors separated this period because the lockdown policy was suspended, right? Since this period is also distinguished in Figure 4, it may be beneficial to treat it as its own section, but I'll leave it to the authors to decide.

Agreed, we have made it as a dedicated section as suggested. It now reads:

"3.2.4 Pre-EOTHO

During pre-EOTHO (Jun 24th–Aug 2nd, 2020) after the lockdown policy was eased, HOA, COA, and BBOA all showed considerably increases of 16%, 30%, and 14%, respectively when compared to lockdown summer period. In which, MO-OOA increased by 45% and LO-OOA decreased by 13%, respectively as the relatively higher temperature and solar radiation favoured the evaporation of LO-OOA and production of MO-OOA from LO-OOA and POA. As shown in Fig. 5, the POA concentrations were much lower when compared to summer 2019 (Aug 1st–Aug 31st, 2019) as travel and economic activities did not return to pre-COVID levels (ONS, 2022; Transport for London, 2020). Specifically, lower HOA (-33%), BC (-37%) due to reduced vehicle mileage resulted, and COA (-59%) due to the reduced commercial cooking activity. As BBOA is co-emitted with COA during of cooking activities, BBOA also decreased slightly from 0.53 to 0.38 µg/m3 (-28%, Fig. 5)."

Reviewer #3

This manuscript presents a year-long ACSM dataset to investigate the influence of short-term anthropogenic emission perturbations on the chemical composition of urban aerosols in London, with a particular focus on the source-resolved organic aerosol (OA) components. The dataset itself is valuable and potentially informative. From the content of the manuscript, this is a revised version addressing previous reviewer comments. However, it seems that the authors have not made a concerted effort to improve the manuscript to the standard required for publication in ACP. There are still numerous instances of careless editing errors. For example, on page 3, line 53: "Click or tap here to enter text.", and on page 12, line 232: "tError!".

We agree with the reviewer that this work is valuable and will be informative, especially for policymakers to improve air quality in urban environments. We apologise for the editorial errors, which have been all addressed in the revised version.

The writing also requires substantial improvement in terms of logic, coherence, rigour, and clarity. For instance, the Introduction is particularly difficult to follow. The narrative does not clearly articulate the main scientific question or its necessity in the context of prior studies. The figures also need improvement. Some of them, such as Figures 3 and 5, contain labels and text that are too small to be legible, and Figure 5 appears to be missing axis lines altogether.

We apologise the small mistakes we made in the last revision, which have been dealt with in this version.

Figure 3 and Figure 5 have been updated according to reviewer's suggestions.

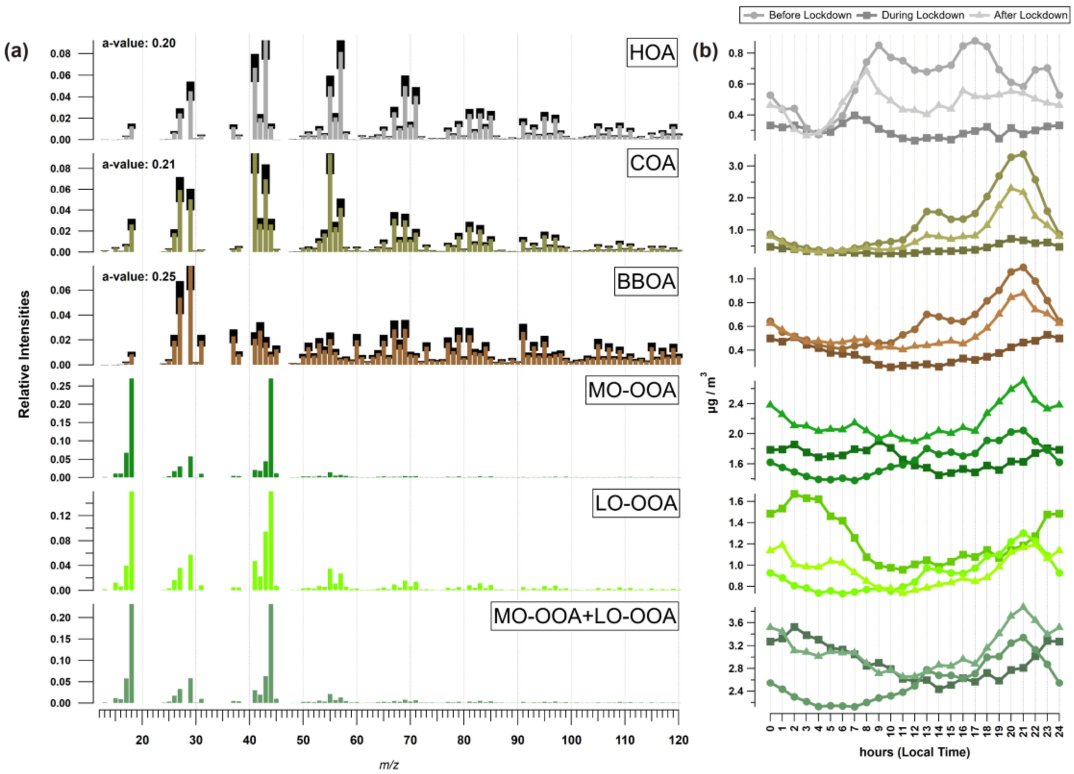

Figure 1 Yearly averaged profiles (a) and diurnal cycles (b) of resolved factors from the rolling PMF analysis at the MY site. Time is expressed in local time.

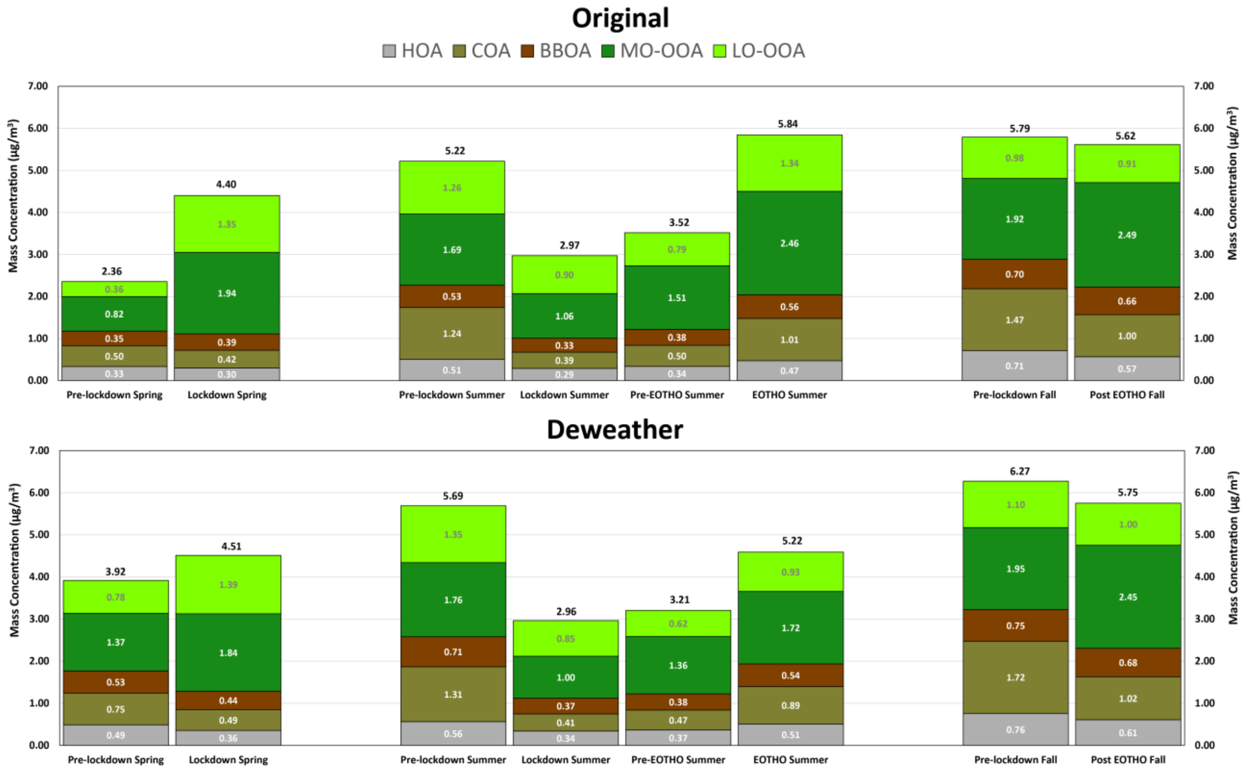

Figure 2 The impacts on OA sources during different periods compared with business-as-usual cases with and without deweathering analysis.

Also, the whole introduction has been rewritten as follows to more coherently outline the context of the study, previous research and the impact of the study:

"1 Introduction

Atmospheric particulate matter (PM) are tiny particles suspended in the air, which impact the climate directly and indirectly (IPCC, 2021; Seinfeld et al., 2006), and cause adverse human health effects (Kelly and Fussell, 2012; World Health Organization, 2021). The PM present in urban areas, such as London, is emitted directly or indirectly from a wide range of natural and anthropogenic sources, can be changed through atmospheric reactions and remain airborne for many days. It is consequently a complex mixture including inorganic species (metals, minerals, black carbon, nitrate, sulphate, etc.) and thousands of organic compounds whose origins remain too complicated to fully quantify. $PM_{2.5}$ (PM with aerodynamic diameter smaller than 2.5 μm) is strongly associated with increased risks of cardiovascular and respiratory related mortalities and hospital admissions (Dominici et al., 2006; Joo et al., 2024; Pye et al., 2021; Wei et al., 2022, 2024). Some studies (Lippmann et al., 2013; UK Health Security Agency (UKHSA), 2022) have begun to demonstrate that some PM constituents and sources have stronger associations with a range of health metrics, including mortality, morbidity, and toxicities although the evidence remains inconsistent (Kelly and Fussell, 2012; Liu et al., 2023; Vasilakopoulou et al., 2023). With 99% of the urban population in Europe exposed to PM2.5 concentrations exceeding the WHO air quality guideline (European Environment Agency, 2024; World Health Organization, 2021), delivering clean air is a target for European and international governments according to the EU air quality directive (European Union, 2024). However, delivering publicly acceptable policies to improve air quality *remains challenging (Mebrahtu et al., 2023; Oltra et al., 2021). Targeting the sources of PM that are most health-relevant could be a more cost-effective (Wu et al., 2023), more easily communicated and more publicly acceptable approaches (Pinakidou, 2025) to improve public health. It is therefore important to better quantify the sources of PM and understand how they respond to policy interventions.*

The COVID-19 pandemic is a natural experiment to assess the impact of policies which, while not aiming to reduce $PM_{2.5}$ concentrations, significantly restricted social and economic activities and consequently reduced emissions. During the UK national lockdown, people were ordered to stay at home, and all non-essential businesses were closed, including pubs, cafes and restaurants from the end of Mar 26th, 2020. Non-essential shops were allowed to open from Jun 15th, and the first national lockdown came to an end on Jun 23rd, 2020. However, pubs, restaurants, and cafes were only allowed to open from July 4th, 2020. The UK recorded a 2.5% drop in Gross Domestic Product (GDP) in the first quarter of 2020, partly as people reduced their own activity prior to national lockdown. This accelerated to a 19.8% fall in GDP in April to June 2020 and household spending fell by over 20% over this period, the largest quarterly contraction on record, which was driven by falls in spending on restaurants, hotels, transport, and recreation (ONS, 2022). The UK Government Eat Out to Help Out (EOTHO) Scheme is examined specifically in this study as it influenced emissions from the commercial cooking sector. It was designed to help the hospitality industry recover from the financial impact of the national lockdown, offering a 50% meal discount up to a maximum of £10, which operated Mon to Wed, Aug 3rd to Aug 31st, 2020.

While the impact of these lockdown policies on some air quality metrics was smaller than expected given the large change in emissions (Shi et al., 2021), the abrupt nature of the intervention ensures it is easier to detect than other air quality policies that are more incremental in nature (Mudway et al., 2019). Some studies have investigated the lockdown impacts on chemical composition and sources of PM, which mainly focused on cities in China (Hu et al., 2022; Tian et al., 2021; Xu et al., 2020), a kerbside site in Toronto, Canada (Jeong et al., 2022), and an urban background site in Paris, France (Petit et al., 2021). These studies all resolved primary sources including traffic related emissions, biomass burning emissions from residential heating, cooking emissions (except Paris), and secondary sources from PMF analysis on organic aerosol (OA). Traffic and cooking emissions appeared to decrease during the lockdown in all sites, while biomass burning predominately from residential heating sources in Chinese cities increased as result of remote work and rather early lockdown measures (Jan-Feb 2020) compared to France. Secondary organic aerosol (SOA) showed a more complex phenomenon given its abundance in organic components and dynamic spatiotemporal conditions. Overall, the lockdowns resulted in decreased SOA in both northwest cities in China (Tian et al., 2021; Xu et al., 2020) and Paris (Petit et al., 2021) due to lower primary emissions, and therefore fewer SOA formation products. However, Beijing experienced a large increase in SOA concentrations due to increased fossil fuel and biomass burning emissions, long-range transport influences as well as favourable meteorological conditions (high RH, low wind speed and low boundary layer height) for SOA formation during the lockdown period (Hu et al., 2022). Therefore, the lockdown effects on the SOA were dependent on the abundance of primary emissions, long-range transported air masses, and meteorological conditions. To date, there are few studies that investigate how COVID-related policies could have impacted PM chemical composition and sources. Petit et al. (2021) and Gamelas et al. (2023) are the only two studies in Europe. The unique

COVID-related policies in the UK therefore provide a rare opportunity to investigate the impacts these policies had on chemical composition and OA. We used highly time resolved measurements from an air quality supersite located in the Central London from 2019 to 2020, and advanced source apportionment approaches to quantify the PM composition and OA sources before, during and after the UK national lockdown and EOTHO scheme. This study provides valuable insight into PM sources and composition in a global mega city and how air quality responds to abrupt changes in emissions from different sources. Importantly, it helps to establish the importance of cooking as a source of PM and uniquely associates biomass burning organic aerosol with commercial cooking emissions. This provides crucial information to policy makers as they attempt to reduce exposure to air pollution in urban areas."

In this revised version, the authors have included the results from deweathering analyses. However, no details are provided regarding the modelling methodology, model configurations, validation approach, and/or cross-validation results. Such critical information is essential and should be explicitly described. Additionally, the meteorological parameters considered (i.e., relative humidity, wind speed, wind direction, and temperature) are rather limited and insufficient to represent the complexity of meteorological influences on atmospheric aerosol variability. It is strongly recommended that the authors include other relevant meteorological indicators such as planetary boundary layer height and air mass trajectory cluster analysis. Relevant references that may help improve this section include Grange and Carslaw (2019, Sci. Total Environ.) and Shi et al. (2021, Sci. Adv.).

Thank you for reviewer's comment on the deweathering analyses in our work. We have now included additional detail of the modelling methodology, model configurations, validation approach, and/or cross-validation results.

Regarding the inclusion of additional meteorological indicators other authors (e.g. Grange and Carslaw, (2019)) have suggested boundary layer height, air mass cluster, or back trajectory information would be beneficial to include to deal with pollutants primarily controlled by regional scale process. However, the aim of this study is understanding how COVID-related policies affect primary/local emission sources (i.e., BC, HOA, COA, and BBOA), which will not be affected significantly by regional processes, and we do not feel that the additional metrics will improve the quantification of these PM components. It is consistent and comparable with previous studies (Font et al., 2022; Grange et al., 2021; Yao and Zhang, 2024) that includes similar meteorological parameters (i.e., wind speed, wind direction, relative humidity, temperature). It has already achieved excellent model performances with an R^2 larger than 0.77, which are comparable with previous studies (Font et al., 2022; Grange et al., 2018, 2021; Grange and Carslaw, 2019; Krechmer et al., 2018; Shi et al., 2021; Yao and Zhang, 2024) All of which have provided scientifically insightful findings. Thus, the overall conclusion will not be changed even with the new analysis. To clarify this, we have added necessary text in the main text (Section 2.5 and 3.1).

Table S 1 Boot regression trees model performance on the testing dataset for each PM species/sources

	Slope	*R^2 (Pearson)*	*RMSE*
BC	*0.98*	*0.94*	*0.46*
Org	*1.01*	*0.93*	*1.80*
NH_4	*1*	*0.87*	*0.39*
NO_3	*1.02*	*0.92*	*1.52*
SO_4	*1.01*	*0.94*	*0.63*
HOA	*0.97*	*0.77*	*0.34*
COA	*0.99*	*0.82*	*0.66*
BBOA	*1*	*0.86*	*0.29*
MO-OOA	*1.01*	*0.92*	*0.64*
LO-OOA	*1.01*	*0.89*	*0.45*

To expand on model performance, we have now included the performance of our BRT model for each PM species/source in Table S2 in SI as shown above. The performance are good to excellent with slopes from 0.97 to 1.02 and R^2 (Pearson) from 0.77 to 0.94, which are comparable and consistent with previous studies (Font et al., 2022; Grange et al., 2018, 2021; Grange and Carslaw, 2019; Krechmer et al., 2018; Shi et al., 2021; Yao and Zhang, 2024). As suggested by Grange et al. (2018) and Yao and Zhang, (2024), BRT models generally suffer from overfitting, however, this does not appear to be significant in our study (i.e., generally consistent good performances for both training and testing datasets). This is in contrast to random forest models, which normally provide poorer statistical agreement (Grange et al., 2018; Yao and Zhang, 2024). However, as our BRT model already provides good to excellent predictions without overfitting, we do not consider it necessary to perform random forest model by including additional meteorological parameters. Previous authors have also demonstrated that the BRT and random forest models show generally similar results (Yao and Zhang, 2024). Therefore, as the scope of this study is not deweathering and different models with different parameters will not change the results significantly, especially it will not change our conclusions how COVID-related policies will affect the primary emissions, we have decided not to perform the additional analyses reviewer suggested. However, to clarify the model performance, rationale for model and parameter selection, an additional section has been added in the Methodology section (Section 2.5 and 3.1) as follows:

"2.5 Meteorological normalisation using boot regression tree model

Meteorological normalisation, also known as deweathering analysis, has been conducted using the "worldmet" R package (Carslaw, 2025) to build boot regression tree (BRT) models for all resolved OA factors from PMF as well as chemical species measured. Considered variables, included relative humidity, wind speed, wind direction, and air temperature trend, hours of the day (local time), day of the week, Julian dates, week of the year as suggested by (Carslaw, (2025). While Grange and Carslaw (2019) have also suggested boundary layer height, air mass cluster, or back trajectory information would be beneficial to include to deal with pollutants primarily controlled by regional scale process. However, the aim of this study is understanding how COVID-related policies affect primary/local emission sources (i.e., BC, HOA, COA, and BBOA), which will not be affected significantly regional process, therefore additional metrics will most likely not improve the quantification of these PM *components. It is consistent and comparable with previous studies (Font et al., 2022; Grange et al., 2021; Yao and Zhang, 2024) that includes similar meteorological parameters (i.e., wind speed, wind direction, relative humidity, temperature). In addition, since the trained BRT models are sufficiently good even without considering boundary layer height and back trajectories, performing random forest model will not improve the model significantly, nor change the results drastically as suggested by Yao and Zhang (2024). Thus, in this study, only BRT models were trained and the meteorological effects subsequently removed (i.e., relative humidity, wind speed, wind direction, and air temperature) on all PM/OA.*

3 Results and Discussions

3.1 Model performance of meteorological normalisation

The performance of each model (for individual species/source) is shown in Table S2 with slopes from 0.97 to 1.02 and R^2 (Pearson) from 0.77 to 0.94, which have similar or somewhat better performances compared with previous studies (Font et al., 2022; Grange et al., 2018, 2021; Grange and Carslaw, 2019; Krechmer et al., 2018; Shi et al., 2021; Yao and Zhang, 2024). As shown in the SI (Fig. S8 and Fig. S9) and the lower panel of Fig. 5, meteorological effects were generally considerable, especially for Pre-lockdown Spring period, while it does not change the conclusion of the effects from lockdown and EOTHO policies. Therefore, the main results presented in this study are based on the original measurements."

Overall, the work may be more appropriately considered as a measurement-report type submission. Major revisions are necessary before the manuscript can be considered suitable for publication in ACP.

Thank you for the comment, however, we strongly disagree with the reviewer on this point. This work has potentially high impact for policy makers internationally, it provides clear evidence of cooking as a major source on $PM_{2.5}$ in urban areas, which is currently an under-recognised source, and co-emission of BBOA which has previously been wholly ascribed to wood burning. It therefore has the potential to provide important scientific justification for further studies and mitigation approaches. Such mitigation has been suggested in London government guidance but not yet acted upon. This is especially important as cities such as London have substantially reduced traffic emissions yet still do not meet international air quality targets.

Furthermore, the scope of this work fits perfectly with the special issue we have submitted to (https://acp.copernicus.org/articles/special_issue1175.html), which specifically focus on following aspects:

- quantifying the spatial and temporal extent of stay-at-home policies on the European atmosphere, at both local and regional scales,
- evaluating the impact of lockdown measures on the formation of secondary pollutants,
- documenting the impact of reduced emissions (including air-traffic emissions) on cloud properties and occurrence, and
- estimating the "missing" emissions using observation–model approaches. The outcome from the special issue aims to provide an in-depth analysis of the

perturbation induced by the repeated lockdowns on the complex atmospheric system.

This work is therefore not only scientific significant, aligns with the scope of this special issue of the ACP, but is also impactful for policies both locally and worldwide.

In terms of writing, specific sentence improvements could be made. For instance:

1. *Line 17 sentence could be revised to: "Organic aerosol (OA), a major component of submicron particulate matter (PM1), has significant impacts on both human health and climate. Quantifying its sources is therefore crucial for developing effective mitigation strategies."*

Revised

2. Lines 19 sentence could be rewritten as: "Positive matrix factorisation (PMF) applied to aerosol chemical speciation monitor (ACSM) mass spectral data offers a robust approach for quantifying OA sources."

Revised

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