

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 all the constructive comments from two reviewers. The following texts are the response to the reviewers with normal italic font is original review comments, green font for authors' responses, and the blue italic font for changes in the revised version.

Review comments

Authors' response

Texts from the revised version

Reviewer 1

Chen et al. investigated the effect of COVID-19-related policies on aerosol composition in London, particularly how the source apportionment of organic aerosol varied before and after the implementation of different policies (lockdown and Eat Out to Help Out (EOTH)). They found that the lockdown due to COVID-19 substantially reduced levels of primary organic aerosol (POA = hydrogen-like + cooking + biomass burning OAs (HOA + COA + BBOA)), lowering them by approximately 50% compared to pre-COVID levels. In contrast, oxygenated organic aerosols (OOA = less-oxidized OOA (LO-OOA) + more-oxidized OOA (MO-OOA)) were not as significantly affected. However, when EOTH was introduced, POA levels—especially COA—increased, highlighting cooking activities as an important source of urban air pollution.

This study is a compelling example of why policymakers should consider unintended consequences when implementing policies. However, several aspects need improvement before publication. My primary recommendation is for the authors to reorganize the results and discussion sections to create a smoother flow that readers can easily follow and ensure the content aligns with the title. I suggest restructuring sections to first introduce the regional characteristics, then clearly describe the similarities and differences in OA characteristics during the pre-COVID, COVID, and EOTH periods, while avoiding a time series-based presentation of the results.

That said, the authors present an excellent dataset that clearly illustrates changes in the characteristics of OA across the pre-COVID, COVID, and EOTH periods. Accordingly, I suggest a major revision before publication in ACP. Additional comments are listed below.

We appreciate the positive feedback from the reviewer, and we agree that this study has strong policy implications on cooking emissions in the urban environment. We also agree with the reviewer regarding the restructuring of the manuscript and have

implemented this in the corrected manuscript. The response to the additional comments is provided below.

Major comments:

The ME-2 solution selection process should be described in greater detail. I expect to see more information in the SI on how the current solution differs from those obtained using a lower or higher number of factors. Additionally, it would be beneficial for the authors to explain how varying the α -value influences the source apportionment solutions. Lastly, I noticed that the α -value in Figure 2 is presented in a 0.## format, whereas the measurement section states that the α -value was adjusted in increments of 0.1. The authors should provide a comprehensive description of the source apportionment analysis, including the aspects mentioned above.

Determining the number of factors is done during the seasonal PMF step. Doing rolling PMF with different number of factors is not feasible as it takes substantial time to do so. To clarify our selections of the number of factors, we've added two sentences as below in the main text in section 2.4 (line159-162 in the clean version):

“Adding an additional factor resulted in split of COA factor, decreasing it to four factors caused mixing between the MO-OOA and COA factors. Therefore, 5 factor-solution was determined across the whole year.”

The sensitivity analysis of different α -values was initially conducted during the pre-test of seasonal PMF. The best solution was chosen as the base case solution for further bootstrap analysis based on factor profiles, time series, diurnal cycles, etc. Inappropriate α -values are typically easy to identify as the factor profiles/diurnal cycles will become mixed. In addition, the bootstrap analysis of the seasonal PMF enabled random α -value approach (Canonaco et al., 2021; Chen et al., 2021) by using the constraints retrieved from base case solution, which means the α -values for each constraint is allowed to vary from 0.1 to 0.5 with a step of 0.1 to evaluate the rotational uncertainties and temporal variabilities of PMF with 100 iterations (Chen et al., 2021), it has covered all the possible combinations of different α -values. Using the criteria-list mentioned in Table S1, we are able to select “good” PMF runs automatically with minimized subjective judgements. Similarly, the rolling PMF applies bootstrap using the random α -value approach from 0.1 to 0.5 with a step of 0.1 for HOA, COA, and BBOA with 50 iterations per rolling window (i.e., 14 days). The α -value showed in Figure 2 is the averaged α -values with 2 decimal places for selected PMF runs for all the windows throughout the year. That's why the averaged α -values have two digits. To avoid confusion, line 174-183 (clean version) has been revised as follows:

“Rolling PMF was conducted with a time window of 14 days and a step of 1 day by constraining primary factor profiles of HOA, COA, BBOA in Fig. S2 (averaged bootstrap results) and two additional unconstrained factors with bootstrap resampling and the random α -value option (0.1-0.5, step of 0.1, 50 iterations/window). A criteria list including selections based on both time series and factor profiles as shown in Table S1 was applied as per Chen et al. (2022). With the help of t-test in temporal-based criteria (1-3), we can minimize subjective judgements in determining the environmentally reasonable results. Eventually, 3,166 runs (14.1%) of the PMF runs were selected across different rolling windows across the whole year to average as the final results (utilized α -values were averaged to two decimal places) with 4.9 % unmodelled data points, which is comparable with other rolling PMF analyses (Chen et al., 2022).”

As the detailed methodology have been described in (Chen et al., 2022) with step-by-step instructions, and the scope of this study is to focus on the OA characteristics under different policies. Therefore, more detailed descriptions of the pre-test PMF is not provided.

1. *I expect to see a more detailed description in Section 3.1, not just the changes in mass concentration. For example, how did the fraction of non-refractory species vary by season or implementation of policies? How was this particular period different from the past PM variation from literature? Also, when comparing the PM characteristics between pre-lockdown vs. post-lockdown, I think it should be more clearly compared by season.*

The scope of this study is to highlight the influences of COVID-related policies on source apportionment results of organic aerosols. In general, the COVID-related policy did not have significant influences on PM species (except for BC) and the seasonal variabilities of PM species was quite similar to other European sites as described in Bressi et al. (2021). Therefore, the variabilities of PM species over different periods were briefly discussed except for the BC, which is discussed extensively.

2. *I recommend the authors revisit the discussion regarding OA factors because I see multiple points that are difficult to agree with the authors' perspectives. For example, it is hard for me to recognize a strong seasonality in Figure 3 while only the effects of COVID-related policies are noticeable. In addition, I think there is a small lunchtime peak in the diurnal of COA if you zoom in. Additionally, the authors should add more detailed discussion by comparing with previous studies that were conducted in other major cities around the world. I added further details in the specific comments below.*

We agree that Figure 3 is difficult to see the seasonality of OA sources. This is common in the UK, which is influenced by changeable weather systems, and seasonality of OA sources in another London site (urban background) have been discussed extensively in (Chen et al., 2022). Both datasets showed a similar seasonality for all the sources.

The statement about COA diurnal variation during lockdown has been amended to avoid confusion, as indeed it still showed small lunchtime peak, likely due to takeout activity of some restaurants that were still active as well as potential increases for residential cooking activities at that time. Line 291-294 (clean version).

“The diurnal variation of COA and BBOA during lockdown showed much less intensity overall but the distinctive lunchtime peak remained as the pre-lockdown; and the evening peak reduced its intensity (Fig. 3). This is because the takeout activities of some restaurants were still active as well as the potential increases for residential cooking activities during lockdown.”

As mentioned in the introduction, there are not many OA source apportionment studies during the lockdown periods around the world. Majority of studies were in China, where the lockdown time was different from the UK and the rest of the world. Also, most of studies only showed the effects of lockdown, which demonstrated the decrease of primary organic aerosol (POA) sources. However, the unique EOTHO policy in the UK cannot be compared with other cities.

3. *In some sections, it was hard to follow the authors' discussion due to the lack of results or references that support authors' opinions. A couple of examples can be found in Section 3.2. If the authors can provide any results (or references) that show changes in temperature, GDP changes, travel, economic activities, vehicle mileage, etc., it would be much more supportive of the authors' discussion in Section 3.2.3 and 3.2.4, where focused on policy-related effects on OA.*

Citations have been provided to support authors' opinions

4. *I recommend that the authors add a separate section at the beginning of Section 3.2 to describe the typical OA characteristics in the region. This would help guide readers and establish a clearer baseline for understanding the impact of the policies on OA characteristics. Additionally, I suggest restructuring the paper as follows: (1) general OA characteristics of the region, (2) pre-lockdown, (3) lockdown, and (4) EOTHO. From my perspective, the current structure, which includes an overall time series and diurnal section, weakens the paper's main argument and leads to repetitive statements (e.g., long-range transport, seasonality of photochemistry).*

We appreciate reviewer's comment, and we have revised the section 3.2 according as shown in the clean version.

Specific comments:

1. *Line 119: Is this ACSM deployed with a standard vaporizer? Or a capture vaporizer?*

It's a standard vaporizer. Revised as follows:

"Quadrupole ACSM (Q-ACSM, Aerodyne, Ltd., Ng et al. (2011)) with a standard vaporizer provides 30-min mass loadings of chemical species within non-refractory submicron aerosol (NR-PM₁), including NH₄, NO₃, SO₄, Cl, and OA.

2. *Line 218: What kind of activities are related to elevated PM₁ events? Did air mass originate from northern continental Europe and also be related to agricultural activities? Please specify."*

Agreed. Additional sentences have been added as follow:

"PM₁ increased by 95% in lockdown spring (Mar 26th–May 31st, 2020) compared to pre-lockdown spring (Mar 1st–Mar 25th, 2020). Specifically, Org, SO₄, NO₃, NH₄, and Cl all increased by 87%, 211%, 73%, 237%, and 132%, respectively. Except for BC, which decreased by 52%. This is due to the polluted air mass originating from mainland Europe and the enhanced agricultural emissions in spring from the UK and wider continental Europe (Aksoyoglu et al., 2020)."

3. *Line 220: Would only volatility be related to lower NO₃ concentration in summer? How about NO_x/NH₃ emission or other factors that can affect the formation of inorganic NO₃?*

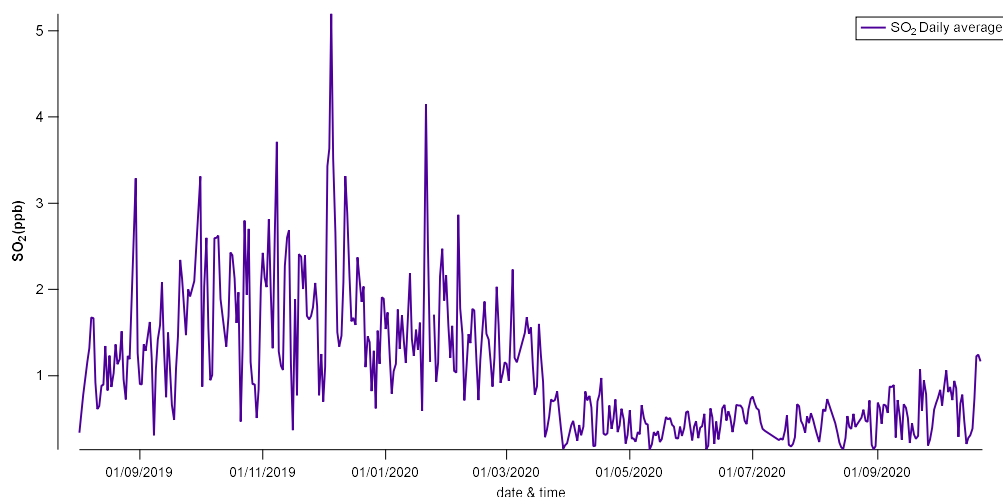
The main reason is its volatility but there are also less agricultural emissions in summer compared with spring and fall seasons. Revised as follows:

“NO₃ concentration reduced in summer 2019 and 2020 as expected compared to spring or fall seasons due to the volatility of NH₄NO₃ and lower agricultural emissions, while SO₄ concentrations increased in summer due to enhanced photochemistry.”

4. Line 221: Please add a citation and describe briefly how SO₄ is formed via photochemistry. Also, how did SO₂ change throughout the measurement period?

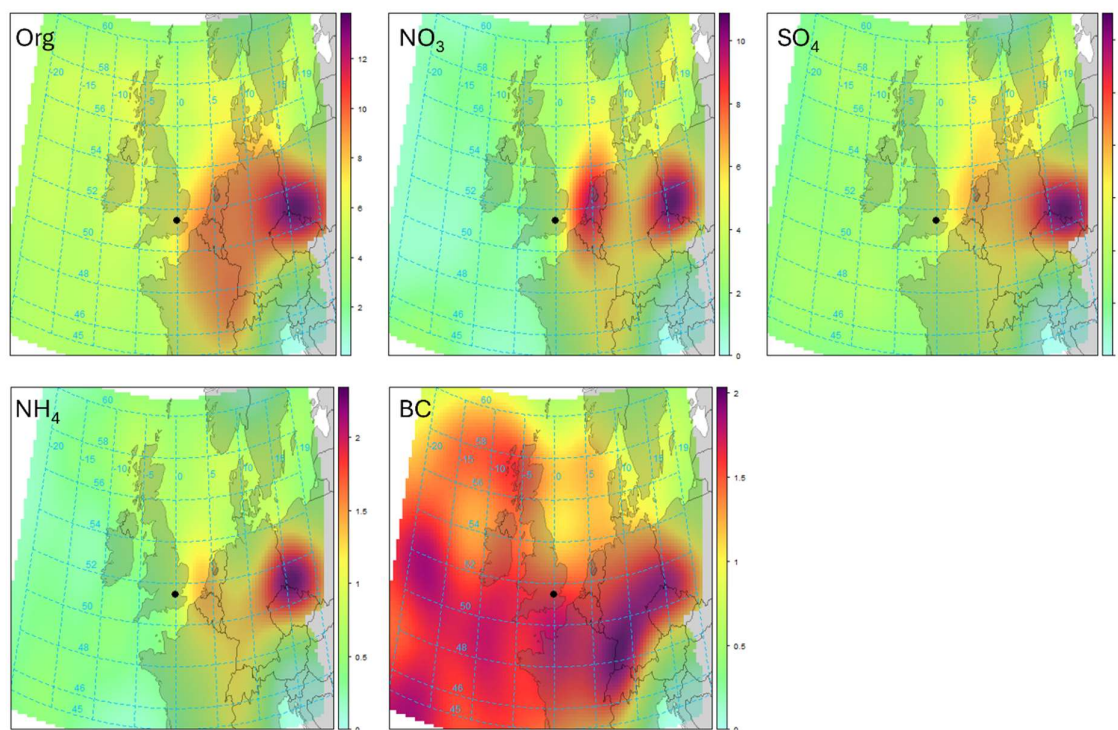
Two citations (Bressi et al., 2021; Chen et al., 2022) have been added where it showed same seasonal variations of enhanced SO₄ in summer due to more intense photochemistry across Europe. Also, Figure S7 (also shown in point 5) was added, which demonstrated the SO₄ was originated from Eastern Europe.

SO₂ concentration decreased significantly with a step change during lockdown and remained at a low level until the end of the measurement period as shown below with a rapid drop (20%) in cargo volumes in the UK in the 2nd quarter of 2020 (Simmonds and George Finch, 2021). This may be the reason that SO₄ in lockdown summer remained at a lower level when long-range transported air mass did not have a big influence in London. On the contrary, the enhanced SO₄ concentration during lockdown spring is due to the air mass is originated from the sea (e.g., MSA) or long-range transported continental air masses.



5. Line 222: Please refer to which figure describes long-range transport. In addition, I would like to see if SO₄ or other pollutants are indeed related to long-range transport via CWT or PSCF backtrajectory analysis.

The CWT of Org, NO₃, SO₄, NH₄, and BC, which confirmed our statement that NH₄, NO₃, and SO₄ are mainly originated from long-range transport. The CWT of Chl is not included as the non-refractory chloride in the atmosphere in Europe is often low (Bressi et al., 2021), therefore, the Chl measurement by ACSM is around detection limit with high uncertainties. And, yes, Figure S7 is added in the end of this sentence.



6. Line 229: Wouldn't the reduced BC concentration from winter to spring be related to less heating or energy consumption?

In central London, residential heating is mainly from natural gas, not wood burning. Also, BC in the measurement site is a roadside station, where vehicle exhaust emission is expected to be the main source of BC. The aethalometer source apportionment model for BC often results in noisy or negative solid fuel BC contribution in the measurement site even during wintertime. Therefore, we are confident the reduced BC concentration from pre-lockdown winter to pre-lockdown spring is mainly caused by the reducing vehicle mileages.

7. Line 241: I find it odd that the weekday comparison of BC concentration appears suddenly in this section. Since it is not a critical discussion point of the paper, I suggest removing it.

Thank you for the suggestion. It's an important message that the BC also went up with EOTH0 from Mon-Wed with the increased traffic and cooking activities since the EOTH0 was only valid from Mon to Wed (Aug 3rd–Aug 31st, 2020). To clarify this, the sentence has been revised as follows:

“Since the EOTH0 was only in place from Mon to Wed, BC concentrations (likely due to increased traffic and cooking emissions) increased on Mon-Wed compared to post-lockdown but before EOTH0 (Jun 24th–Aug 2nd, 2020) (Fig. S10).”

8. Line 256: From my perspective, strong seasonality in OA factors is not as evident as the differences between the pre-COVID and lockdown periods. This is because the total OOA did not change significantly, and the PMF OOA separation carries more uncertainties compared to POA-related factors.

It is true that seasonality of OA sources not strong and here is clearly out-weighted by the effects of COVID-related policies. However, the seasonality of OA sources was still considerably obvious as shown in the figure below, which has been added as Figure S3.

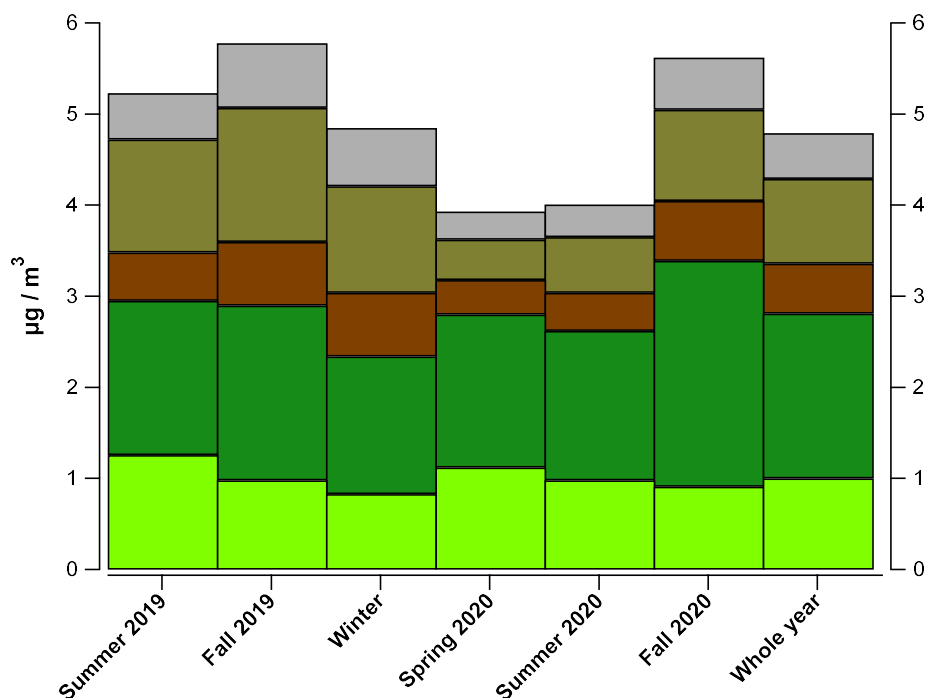


Figure S 1 Stacked mass concentration of OA sources for different seasons during the measurement period.

The text was also amended to avoid confusion:

“OA factors also showed considerable seasonality besides the effects from COVID-related policies (Fig. 4 and Fig. S3).”

It is true OOA sources have larger uncertainties than POA factors in PMF analysis as they have larger fractions in OA as well as they are often not constrained with ME-2 during PMF analysis. However, the variabilities of OOA mass concentration and fractions are still non-negligible according to this previous Europe-wide study (including another urban background site in London, Chen et al., 2022). Therefore, the seasonality of OA factors was still briefly discussed in this manuscript.

9. Line 258: *Would there be any relationship between the decrease in heating and energy consumption?*

The decrease in spring compared to winter could partially be due to the decrease in heating and energy consumption, while the heating and energy consumption in central London are mainly from natural gas and renewable energy instead of solid fuel combustions. Therefore, the decrease in POA (HOA, COA, and BBOA) is not mainly caused by this reason. Here, we clarify the statement by adding following sentences:

“It’s worth mentioning that the reduced POA concentrations in warm season was not caused by reduced residential heating and energy consumption since Central

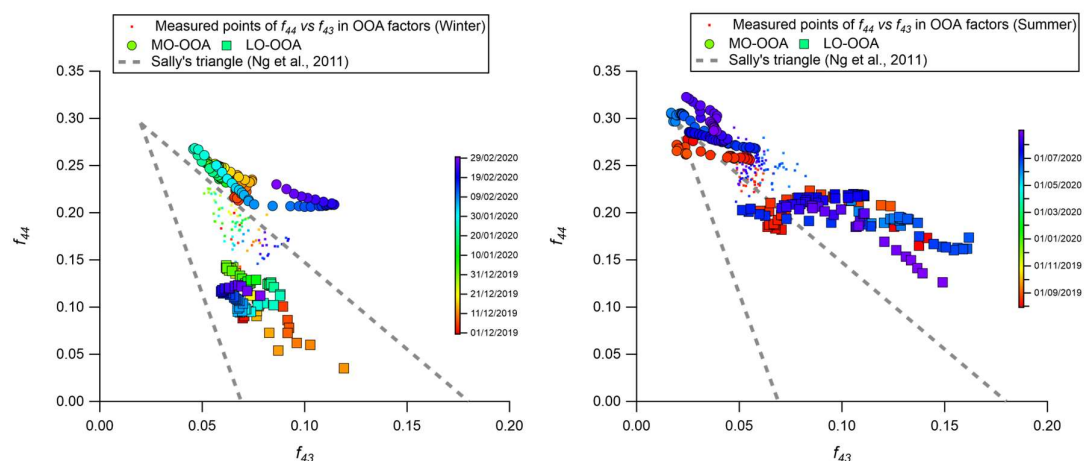
London mainly uses natural gas and renewable energy instead of solid fuel combustions.”

10. Line 261: Wouldn't photolysis affect the evaporation of some semi-volatile components in OOA? How about the f_{44} & f_{43} comparison by season?

Agreed, this is a typo from our side. I meant contributions. The sentence is revised as follows:

“The OOA factor concentrations remain relatively consistent across seasons, while its contributions were larger during the warmer seasons. This is because both high temperature and strong irradiation will enhance the photochemistry and evaporation of POA sources and the increased biogenic volatile organic compound (VOC) emissions lead to high OOA production despite the evaporation of semi-volatile OOA.”

Also, the f_{44} vs f_{43} for winter and summer are provided below, where the seasonalises as well as positions of resolved OOAs are consistent with other studies across Europe (Canonaco et al., 2015; Chen et al., 2021, 2022).



11. Line 261: Where can I find increased VOC emissions in summer? Please provide any related references or data.

It should be increased biogenic VOC emissions in summer. The sentence is therefore revised:

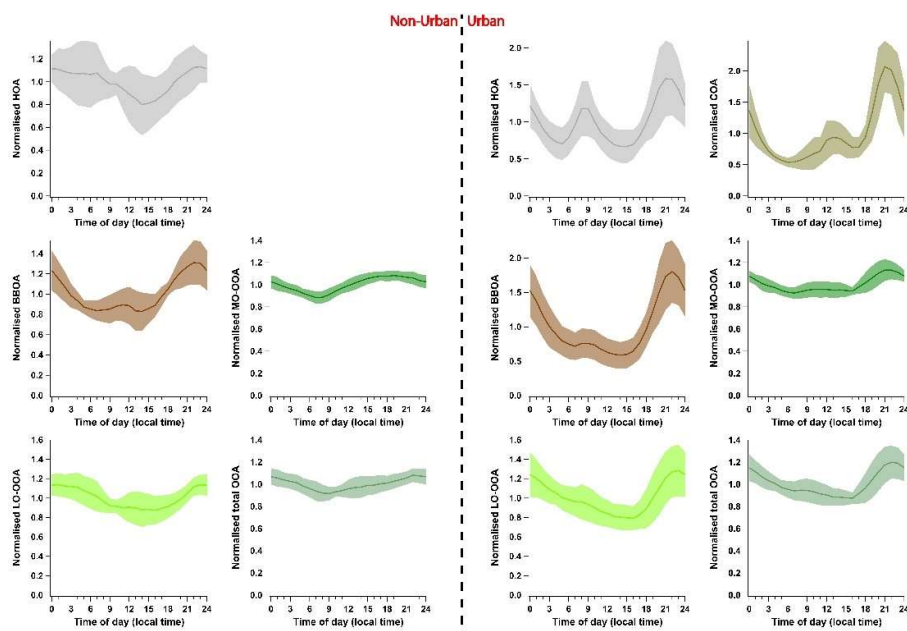
“The OOA factor concentrations remain relatively consistent across seasons, while its contributions were larger during the warmer seasons. This is because both high temperature and strong irradiation will enhance the photochemistry and evaporation of POA sources and the increased biogenic volatile organic compound (VOC) emissions lead to high OOA production despite the evaporation of semi-volatile OOA.”

12. Figure 2 right panel: I suggest having an independent axis for the OA data of the lockdown period.

We want to demonstrate the difference in diurnal cycles for all OA factors before, during and after lockdown instead of showing the diurnal variations of OA factor for each period. Therefore, we believe the same axis is important to convey this message. Thus, the right panel of Figure 2 is kept as it is.

13. Line 275-276: What does the smaller diurnal variation of MO-OOA compared to LO-OOA suggest? Could LO-OOA be associated with a specific source? Please provide a more detailed discussion.

MO-OOA across Europe in general shows smaller diurnal variation compared to LO-OOA as suggested in Fig.4 in Chen et al. (2022) shown below. Also, LO-OOA is also known as semi-volatile OOA (SV-OOA), which showed lower concentration during the daytime compared to the nighttime due to the effect of evaporation during the day and shallow boundary layer in the evening.



14. Line 279: If you zoom in, there could be a small COA peak during lunchtime. Please see Specific Comments #12.

Thanks a lot for pointing this out, we therefore changed the sentence accordingly:

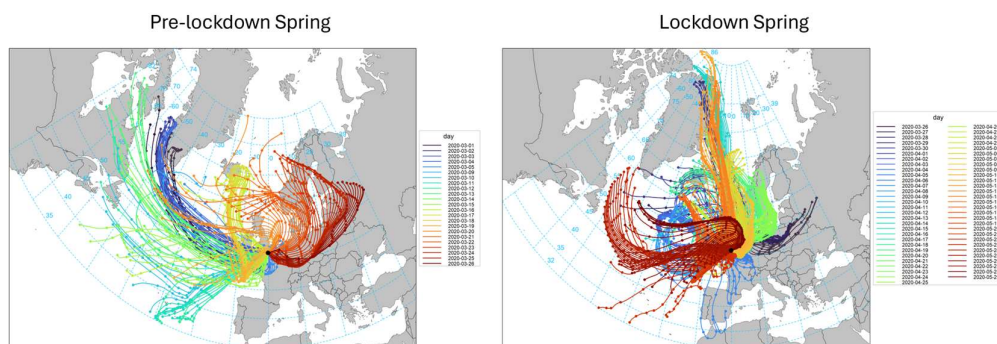
“The diurnal variation of COA and BBOA during lockdown showed much less intensity overall but the distinctive lunch peak remained as shown in the pre-lockdown; and the evening peak reduced its intensity (Fig. 2).”

15. Line 285-289: I believe this part does not fit well in the “Diurnal Cycle” section. Instead, it would be more appropriate in the next section, where the impact of the lockdown is discussed.

This part has been moved to the next section now as suggested.

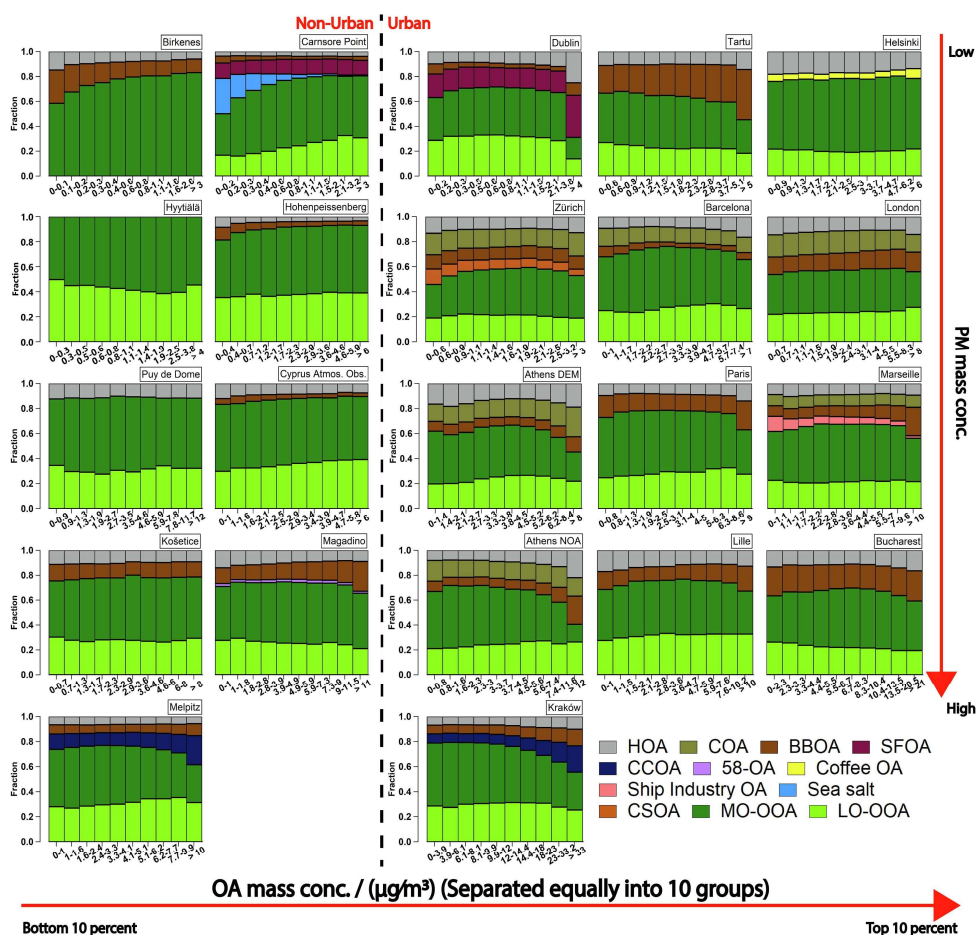
16. Line 289: Figure S3: What was the backtrajectory like before the lockdown? I believe this information should be included to confirm whether the change in PM composition is indeed attributable to the lockdown policy.

The back trajectory plots for pre-lockdown spring and lockdown spring are shown below and has been added as Figure S5, where trajectories are coloured by dates. The high OOA concentration dates at the end of pre-lockdown spring and lockdown spring are clearly showed influences by continental airmass from Europe.



17. Line 293-296: Does the data used to generate Figure 4 encompass the entire period? If so, I believe this information represents a key OA characteristic of central London and should be moved to the beginning of Section 3.2, rather than placed under the “Diurnal” section, to emphasize its significance. Additionally, making comparisons with other major cities, such as New York and Beijing, would be beneficial.

Yes, it includes the entire period and now has been moved up to the beginning of the section 3.2. The comparisons had been done in (Chen et al., 2022) as shown below, where London site is an urban background station measured from 2015-2018. However, comparisons with other major cities are out of the scope of this study.

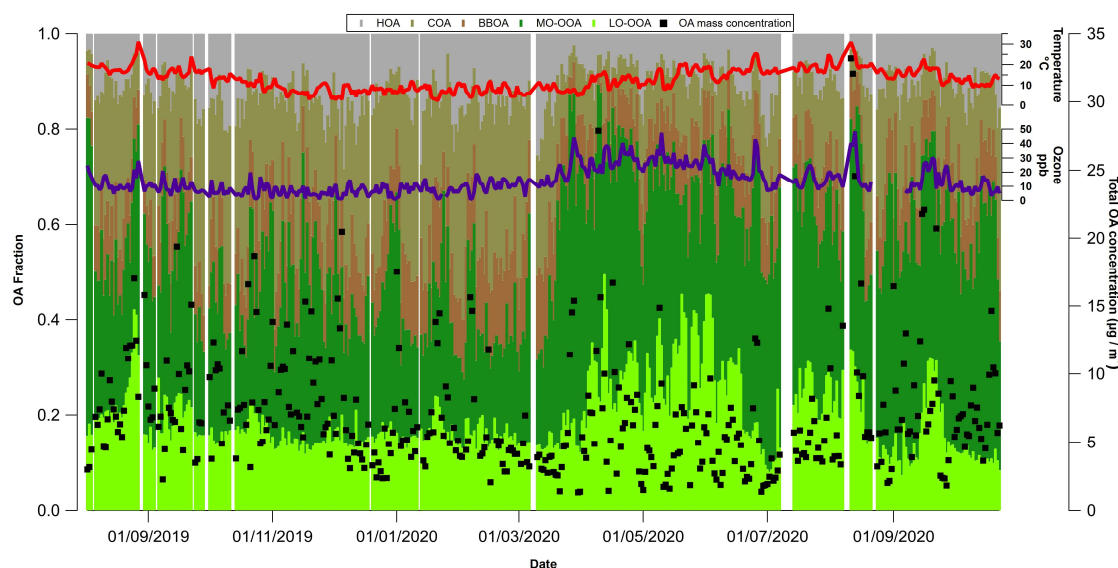


Nevertheless, we revised the sentence to compare with other European cities and another source apportionment results in London urban background site as shown below:

“This suggests that cooking emissions in Central London are responsible for elevated OA concentrations, which was also the case in Athens as shown in Chen et al. (2022).”

18. Line 307: Please add a figure in SI that shows a temperature variation.

Added the following figure in SI as Figure S4.



19. Line 313: Please include any results that allow readers to determine whether photochemistry was increased in pre-lockdown spring compared to March 2020.

As shown in Figure S4 (point 18), the increased temperature and ozone level were evident with the uptick of the OOA fractions. Also, the back trajectory analysis proof that the long-range transportation of continental air mass was responsible for this increase. To be more clear, we revised the sentence as follows:

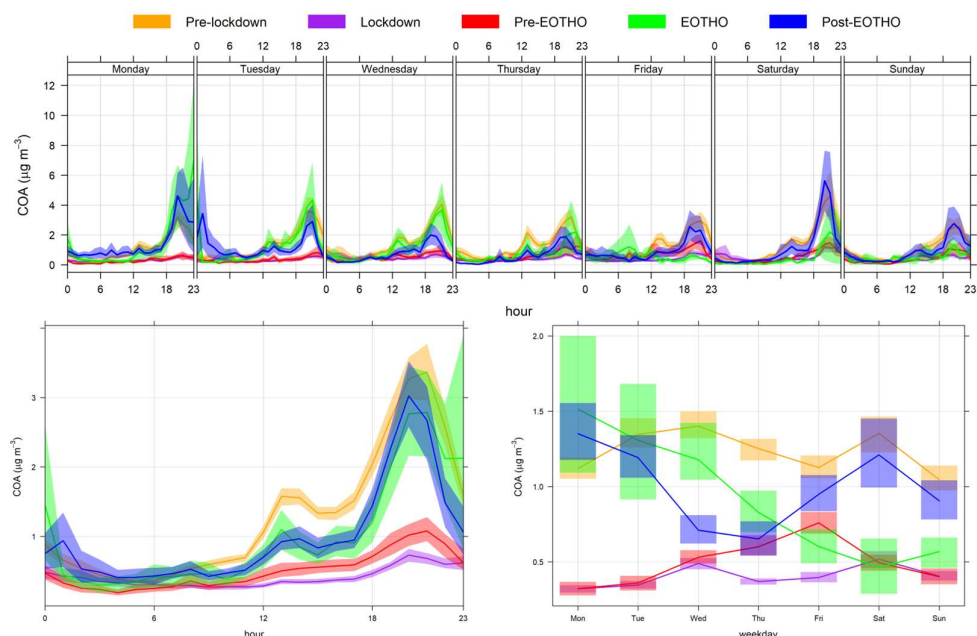
“MO-OOA and LO-OOA increased by 136% and 279%, respectively due to long-range transportation of airmasses from continental Europe (Fig S5 and Fig. S6) and increased photochemistry (enhanced temperature and ozone levels in Fig. S4) compared to the first 25 days in Mar 2020.”

20. Section 3.2.4: I suggest relocating the POA part of this section to the front to emphasize the impact of EOTH0.

Agreed, moved to the end of this section now.

21. Line 341 & 345: The fact that EOTH0 operated only from Monday to Wednesday may explain why COA did not return to pre-COVID levels. Therefore, Figure 6 should include a pre-COVID weekday plot and discuss if the difference in COA for the rest of the week would be responsible for the COA not being returned to pre-COVID levels.

Agreed, the figure 5 and figure 6 are merged into one figure to show the weekly cycles as well as shown below:



22. Line 342: From Figure 5, it is hard to recognize a 10% increase in COA from EOTHO to post EOTHO. Please show a figure that clearly shows such a difference or remove this statement.

True, this was a miscalculation from our side. This statement has been deleted as it's at the same level between EOTHO and post EOTHO.

23. Line 370: Which cities do you refer to? Please specify and add citations

This statement is true but not really important to this study, therefore, excluded from this sentence as follows:

“Clearly detecting this change confirms the presence of COA (20% to OA) as an important source of OA in London and the importance of commercial cooking as a source.”

Minor comments:

1. Line 47: Please add references that show PM_{2.5} composition-dependent health effects and hospitalization:

- Pye, H. O. T., Ward-Caviness, C. K., Murphy, B. N., Appel, K. W., and Seltzer, K. M.: Secondary organic aerosol association with cardiorespiratory disease mortality in the United States, *Nature Communications*, 12, 7215, 10.1038/s41467-021-27484-1, 2021.
- Joo, T., Rogers, M. J., Soong, C., Hass-Mitchell, T., Heo, S., Bell, M. L., Ng, N. L., and Gentner, D. R.: Aged and Obscured Wildfire Smoke Associated with Downwind Health Risks, *Environmental Science & Technology Letters*, 11, 1340-1347, 10.1021/acs.estlett.4c00785, 2024.

Added

2. Line 49: Please add references that show source apportioned PM association with health effects owing to oxidative stresses:

- Vasilakopoulou, C. N., Matrali, A., Skyllakou, K., Georgopoulou, M., Aktypis, A., Florou, K., Kaltsonoudis, C., Siouti, E., Kostenidou, E., Błaziak, A., Nenes, A., Papagiannis, S., Eleftheriadis, K., Patoulas, D., Kioutsioukis, I., and Pandis, S. N.: Rapid transformation of wildfire emissions to harmful background aerosol, *npj Climate and Atmospheric Science*, 6, 218, 10.1038/s41612-023-00544-7, 2023.
- Liu, F., Joo, T., Ditto, J. C., Saavedra, M. G., Takeuchi, M., Boris, A. J., Yang, Y., Weber, R. J., Dillner, A. M., Gentner, D. R., and Ng, N. L.: Oxidized and Unsaturated: Key Organic Aerosol Traits Associated with Cellular Reactive Oxygen Species Production in the Southeastern United States, *Environmental Science & Technology*, 57, 14150-14161, 10.1021/acs.est.3c03641, 2023.
- Daellenbach, K. R., Uzu, G., Jiang, J., Cassagnes, L.-E., Leni, Z., Vlachou, A., Stefenelli, G., Canonaco, F., Weber, S., Segers, A., Kuenen, J. J. P., Schaap, M., Favez, O., Albinet, A., Aksoyoglu, S., Dommen, J., Baltensperger, U., Geiser, M., El Haddad, I., Jaffrezo, J.-L., and Prévôt, A. S. H.: Sources of particulate-matter air pollution and its oxidative potential in Europe, *Nature*, 587, 414-419, 10.1038/s41586-020-2902-8, 2020.

added

3. Line 56: Please add references related to PMF analysis on OA:

- Jimenez, J. L., Canagaratna, M. R., Donahue, N. M., Prevot, A. S. H., Zhang, Q., Kroll, J. H., DeCarlo, P. F., Allan, J. D., Coe, H., Ng, N. L., Aiken, A. C., Docherty, K. S., Ulbrich, I. M., Grieshop, A. P., Robinson, A. L., Duplissy, J., Smith, J. D., Wilson, K. R., Lanz, V. A., Hueglin, C., Sun, Y. L., Tian, J., Laaksonen, A., Raatikainen, T., Rautiainen, J., Vaattovaara, P., Ehn, M., Kulmala, M., Tomlinson, J. M., Collins, D. R., Cubison, M. J., Dunlea, J., Huffman, J. A., Onasch, T. B., Alfarra, M. R., Williams, P. I., Bower, K., Kondo, Y., Schneider, J., Drewnick, F., Borrmann, S., Weimer, S., Demerjian, K., Salcedo, D., Cottrell, L., Griffin, R., Takami, A., Miyoshi, T., Hatakeyama, S., Shimono, A., Sun, J. Y., Zhang, Y. M., Dzepina, K., Kimmel, J. R., Sueper, D., Jayne, J. T., Herndon, S. C., Trimborn, A. M., Williams, L. R., Wood, E. C., Middlebrook, A. M., Kolb, C. E., Baltensperger, U., and Worsnop, D. R.: Evolution of Organic Aerosols in the Atmosphere, *Science*, 326, 1525-1529, 10.1126/science.1180353, 2009.
- Zhang, Q., Jimenez, J. L., Canagaratna, M. R., Allan, J. D., Coe, H., Ulbrich, I., Alfarra, M. R., Takami, A., Middlebrook, A. M., Sun, Y. L., Dzepina, K., Dunlea, E., Docherty, K., DeCarlo, P. F., Salcedo, D., Onasch, T., Jayne, J. T., Miyoshi, T., Shimono, A., Hatakeyama, S., Takegawa, N., Kondo, Y., Schneider, J., Drewnick, F., Borrmann, S., Weimer, S., Demerjian, K., Williams, P., Bower, K., Bahreini, R., Cottrell, L., Griffin, R. J., Rautiainen, J., Sun, J. Y., Zhang, Y.

M., and Worsnop, D. R.: Ubiquity and dominance of oxygenated species in organic aerosols in anthropogenically-influenced Northern Hemisphere midlatitudes, *Geophysical Research Letters*, 34, <https://doi.org/10.1029/2007GL029979>, 2007.

added

4. Line 62 and 63: Please add references related to ACTRIS & ASCENT:

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added

5. Line 248: Please add “tracers” after “m/z”.

added

6. Line 253: relocate “respectively” to Line 254, after “59% (26% to PM1)”

done

7. Line 299-302: I believe this should be combined into one sentence. Please revise.

Revised as follows:

“OOA concentrations were relatively unaffected with some variability before, during, and after the lockdown since long-range transportation of airmasses from the continental Europe as observed for NH₄, NO₃, and SO₄.”

8. Line 301&312&320&333: Please refer to the figure that describes the long-range transport of different chemical species.

Added

9. Line 305: Please add a reference (or add a figure in SI) about the GDP variation.

Added a reference

10. Line 326-327: Please add references about the changes in travel, economic activities, and vehicle mileage during the measurement period.

Added references accordingly.

11. Line 359-362: The statement is difficult to follow. Please revise.

Revised as follows:

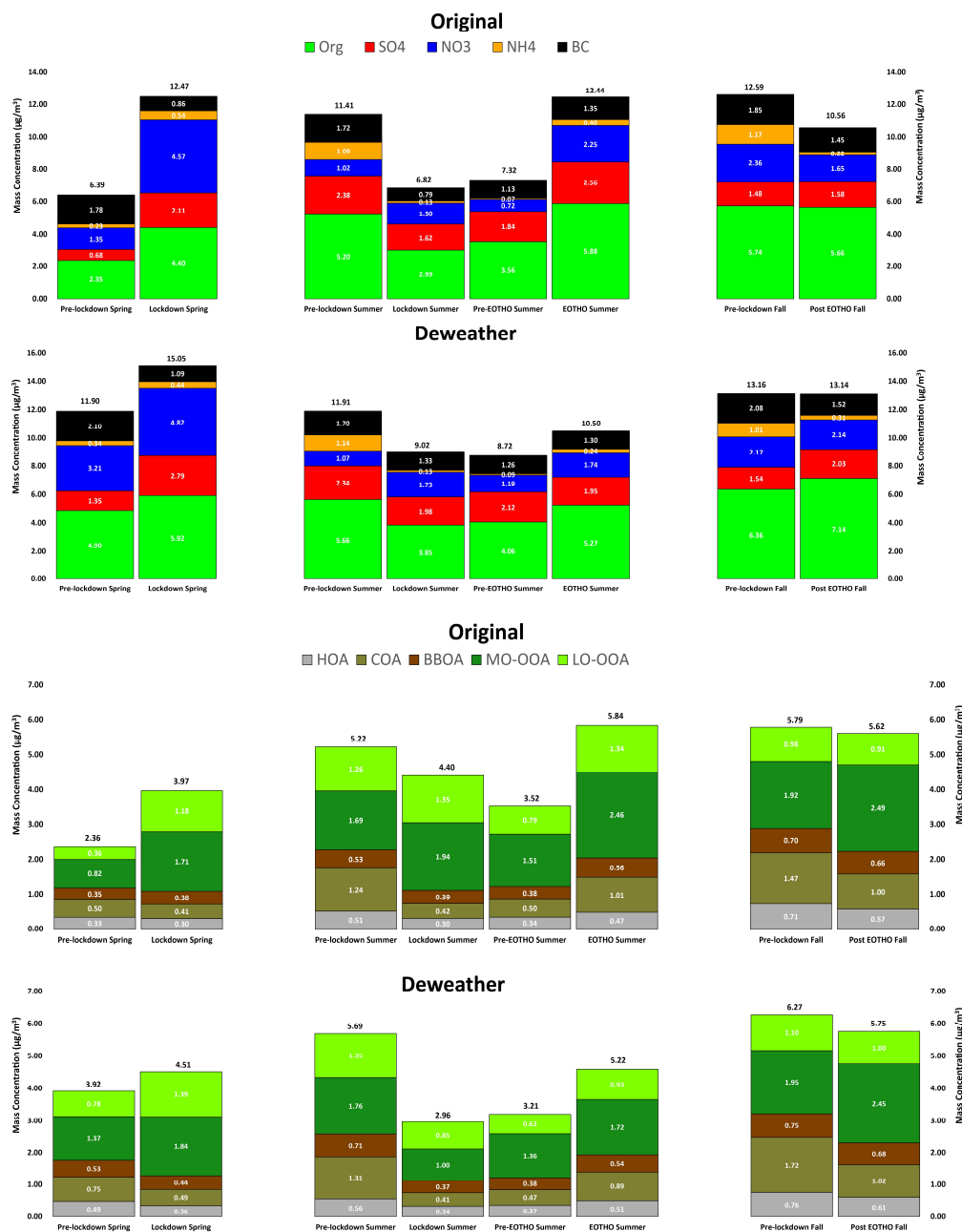
“PM concentrations increased at the beginning of the lockdown (Mar–Apr 2020) despite reduced economic activities, which was caused by long-range transported airmasses instead of primary emissions through examining the source apportionment (and inorganic PM composition).”

Reviewer #2

Chen et al. present an original study on the impact of COVID-19 related social distancing policies in London, UK on the composition of atmospheric aerosols. Aerosol mass spectrometer measurements combined with source apportionment analysis allowed a dedicated focus on the different organic fractions. They highlight a sharp decrease in primary organic aerosols (eg traffic and biomass burning) during lockdown period (LD), but a significant increase of cooking organic aerosols during Eat Out To Help Out (EOTH) policies.

In this study, the impacts of COVID-19 policies have been estimated by comparing LD and EOTH periods to pre-periods, supposed to be representative of business as usual concentrations. After all the flourishing literature on this kind of study, I am rather concerned that the authors didn't take into account (nor at least discuss) the main limitation of this kind of analysis : the variability induced by meteorology. Did the authors check that their "business as usual" periods are representative, meteorologically speaking ? As examples, I don't think the pre-LD period is representative of the meteorological conditions during LD; conditions in June may also be different than in August. Precipitation, temperature, relative humidity, wind speed & direction, BLH (among others) can have a direct impact on the variability and the concentrations measured at a given site, and none of these are presented. This is especially dangerous when stating that component X has increased/decreased by Y% during lockdown (/EOTH), because these results are very important for stakeholders, and can also be easily understood by non-expert citizens. To this regard, methodology is a critical aspect of the work, and must be as robust as possible. I am afraid that this is not quite the case here. As a main major revision, I suggest the authors to : investigate the meteorological representativeness of the different periods, and discuss the potential impacts on the results. If representativeness is not achieved, I suggest either to change methodology (by taking meteorology into account), or reshape the presentation of the results, by avoiding as much as possible to present numbered decrease or increase results.

We appreciate reviewer's positive comments on this manuscript. We also agree that the meteorological conditions can have significant impacts on the concentrations of PM/OA species. Therefore, we have conducted deweathering analysis using "worldmet" R package to minimize the meteorological effects (i.e., RH, air temperature, wind speed, and wind direction). It generally showed moderate effects for most of periods, except for the pre-lockdown spring period. More importantly, the effects of lockdown decreasing the primary organic aerosols (POA) and black carbon were even more evident for the deweathering analysis compared with the original dataset. Similarly, the EOTH increased the COA by 88% instead of 100% after deweathering analysis. In addition, given that the data included business as usual seasons without lockdown and EOTH policies, we have made figures below to isolate the effects from seasonality of the PM/OA species for both original and deweathered timeseries. Again, it demonstrated the evidence of how COVID-related policies had significant effects on both PM and POA mass concentrations as elaborated in the manuscript. Therefore, this study will present the original results by acknowledge the effects of meteorological effects via showing the deweathered results in SI.



Other major concerns :

- Introduction is not well structured. For instance, I don't know why the authors talk about aerosol mass spectrometers and source apportionment here, which is, in that case, more "material & methods" rather than an element of context justifying the interest of the study.

We appreciate reviewer's comment on the structure of the introduction. This part has been moved to Methodology section.

- Section 2.4 is way too long. We don't need a general description of how ME-2 works, the authors need instead to provide all valuable information showing how the final PMF solution was obtained. Additionnally, BBOA has a rather constant contribution to OA throughout the

different seasons (around 10-12%), even in summer. I am guessing that this may primarily come from the use of the rolling PMF rather than barbecue-ing or meat cooking. Plus, it is not clear if the authors constrained BBOA in their previous "summer" PMF. Did the authors also unambiguously find a solution with BBOA during summer only (with a profile resembling to winter BBOA ?) ? The authors may also check BC data (and Angstrom exponent) to support the discussion.

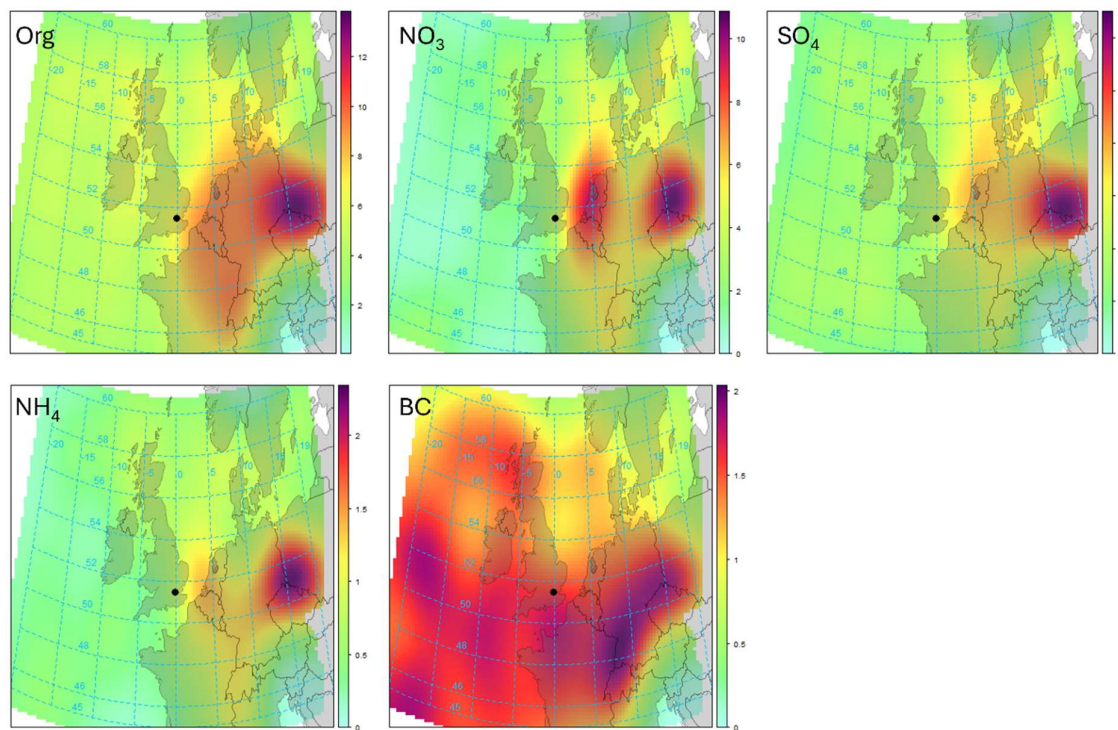
Agreed, general information about PMF, ME-2, etc have been removed now and more details in terms of how source apportionment was conducted have been added in this section.

BBOA was also found in summer PMF in a seasonal PMF analysis in August 2019 as well as the summer 2023 dataset in the same site. It showed a similar diurnal variation as COA since there is evidence showing restaurants in the UK do use solid fuel to cook to meet customers' demands in flavours as a Defra's report suggests (Defra, 2023). With sufficient proof from the Defra report, we think additional analysis on BC data (i.e., Angstrom exponent) is not necessary to support our statement. In addition, in a roadside site, like Marylebone Road, the Aethalometer model to resolve BC_{wb} is highly uncertain when BC is predominantly coming from traffic emissions.

Minor comments:

- Figure S3 (b) and (c) don't quite support long range transport. Maybe trajectory analysis (CWT, PSCF or cluster) would better help. Much more discussion is anyway needed concerning NO₃, because it may not only arise from long range transport. Trajectory analysis may also help to look at the occurrence of air masses (through eg cluster) throughout the different periods of the study. It would contribute to appreciate their meteorological representativeness.

The CWT of Org, NO₃, SO₄, NH₄, and BC are provided in SI, which confirmed our statement that NH₄, NO₃, and SO₄ are mainly originated from long-range transport. The CWT of Cl is not included as the non-refractory chloride in the atmosphere in Europe is often low (Bressi et al., 2021), therefore, the Cl measurement by ACSM is around detection limit with high uncertainties.



- Can the authors elaborate more on Figure S1, and especially how the different slopes obtained over time may impact the presented results. Is it an issue of ACSM calibration? or FIDAS measurements?

The ACSM RIE calibration factors during the measurement periods were averaged and then applied across the whole period, therefore, these discrepancies were not caused by calibrations, but rather different measurement techniques for FIDAS and ACSM. Therefore, the slope is often composition dependent. For instance, the low ACSM concentration but high FIDAS is caused by high fractions of refractory aerosols (e.g., sea salt) which cannot be measured effectively by ACSM. Also, the high ACSM+BC periods might be caused by the high fractions of ultra fine particles, which cannot be measured properly by the FIDAS. Therefore, the ACSM mass closure practice is a sanity check with independent measurements rather than fully trust one instrument over the other.

- Comparison with literature and previous results elsewhere is clearly missing in almost all result sections.

In introduction, there is comprehensive discussions of general phenomena of the influences from COVID lockdown in previous studies. We do agree, there is little comparison with previous studies in the results section. Therefore, we have added some sentences in section 3.2.2 to mention the similarity and dissimilarity from current study and literature

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