Author's response

Referee #1

We thank the Referee for taking the time to read and evaluate our manuscript. We appreciate the highly competent, detailed, and constructive comments provided. The insightful suggestions for revision have been immensely helpful, and we are confident that addressing them will significantly enhance the overall quality, clarity, and impact of our work. Below we respond point by point to the comments and indicate the corresponding manuscript text revisions that we have made. Additionally, we corrected a few minor spelling errors we found.

Referee #1 comments

My main concern with the paper is that some of the conclusions seem overstated or based on a single outlying point. For example, the discussion of Figure 6 on lines 366-375 is a little misleading. *It is only the Winter 2022 TrOA that looks different. The other three TrOA factors fall in line with HOA, SV-OOA and most of the LV-OOA factors, though with slightly higher f values. Yet, in the conclusions, they call out a pronounced signal at m/z 61 for TrOA (line 410 and again at lines 433-434). They need to rephrase the conclusions so that they reflect the actual data on TrOA from all four campaigns.*

Response: We appreciate the Referee's concerns regarding the conclusions drawn from the limited data points. It is crucial to address these constraints, as drawing broad conclusions from a single data point risk overextending the findings.

To ensure complete alignment between our results, discussion, and conclusions, we have revised the conclusion section. Specifically, we have removed the suggestion that the $fC_2H_5O_2^+$ to $fC_2H_4O_2^+$ ratio can serve as a reliable indicator of TrOA. "TrOA differed from both HOA and BBOA based on the $fC_2H_5O_2^+$ to $fC_2H_4O_2^+$ ratio which may serve as an indicator for TrOA." was changed to "TrOA differed from BBOA based on the $fC_2H_5O_2^+$ to $fC_2H_4O_2^+$ ratio."

In conclusion we also changed sentence: "Additionally, TrOA exhibited a pronounced signal for $C_2H_5O_2^+$ (at m/z 61), which was particularly high in winter 2022 data." to be: "Additionally, TrOA exhibited a pronounced signal for $C_2H_5O_2^+$ (at m/z 61) in Winter 2022 data." to point out that exceptionally high $C_2H_5O_2^+$ was found only during one campaign, Winter 2022.

Additionally, we changed the coloring of TrOA area in Figure 6 to lessen the importance of one data point (Winter 2022) and added a colored area for HOA to present more clearly the difference between TrOA and HOA in terms of f values.

The discussion of size distributions on lines 331-337 seems to argue that there is mixing between the TrOA and BBOA-influenced factors. Yet, in the conclusions, the TrOA is assumed to be entirely traffic-related. Again, they should rephrase the conclusions to reflect the data in the paper.

Response: We agree with the Referee that the discussion on mass size distributions created an unintended contradiction with our other conclusions regarding the origin of TrOA. To resolve this inconsistency, we have carefully revised both sections to ensure alignment throughout the manuscript.

The discrepancy regarding the sources of TrOA arises primarily from differences in mass resolution between the two analyses. The mass-based size distributions were calculated using unit mass resolution (UMR) m/z values, whereas the source

apportionment was conducted utilizing high-resolution (HR) ions. Because a single UMR m/z typically comprises a mixture of multiple HR ions, including both hydrocarbon and oxygenated fragments, it can be attributed to several PMF factors. Consequently, size distributions derived from UMR data lack most of the chemical specificity required to separate individual sources.

Based on this comment, we modified text related to mass size distributions:

In results and discussion we replaced "m/z values characteristic of each OA type" by "m/z values typical for each OA type".

We also added text regarding the lack of the chemical specificity of UMR m/z's: "However, it should be noted that UMR m/z's can consist of several high-resolution (HR) ions, and furthermore, the same UMR m/z's, as well HR ions, can be attributed to several sources. Therefore, the mass size distributions for UMR m/z's can only be considered indicative for different sources."

Additionally, we modified the paragraph regarding the sources of m/z 60 and m/z 61 (based on mass size distributions) considerably to say that they were clearly associated with a mixture of traffic and biomass burning -related OA.

We removed following text related to TrOA in Conclusions: "and its mass size distribution peaked at larger particle sizes," to make conclusions clearer.

Also, in the conclusions (line 430), please add a brief summary of why SV-OOA is considered to be traffic-related since the rest of the paragraph is about how different it is from traffic indicators. Since SV-OOA has multiple sources, the statement that OA from traffic reaches 62% seems overstated. Maybe reframe this paragraph as OA from traffic might still be underestimated, even including HOA and TrOA, if some SV-OOA is traffic-related.

Response: Text related to SV-OOA was modified in Conclusions.

We added: "as its concentrations were higher on weekdays than on weekends" and "SV-OOA may also originate from other sources such as biogenic emissions during the growing season or aged biomass-burning emissions during the colder months."

We agree with the Referee that the statement of OA from traffic reaching 62% might be be overstated. Therefore we replaced text: "When SV-OOA was included, the total OA contribution from traffic reached 62 %, which should be considered a maximum estimate." by "Consequently, OA from traffic may still be underestimated, even when HOA and TrOA are included, if a fraction of SV-OOA is traffic-related."

Lines 148-149: The authors state that both the tungsten and laser vaporizers were in use. Were they switching the laser on and off? If so, how often? If the laser was on continuously, they should state that explicitly.

Response: Laser vaporizer was on all the time, not switched on/off. We added clarification to text: "and both vaporizers were on all the time".

Why are no results presented for the black carbon measurement with the SP-AMS? Did the authors include the Cx signals in PMF? I wonder if that might help explain some of the separation into HOA and TrOA. Maybe one is black carbon-rich and one is black carbon-poor.

Response: We agree with the Referee that linking PMF factors to refractory black carbon (rBC) via carbon cluster Cx signals is a compelling approach. However, we intentionally omitted Cx signals from the PMF matrices due to variations in instrument performance across the different field campaigns. Because these studies were conducted over an extended period (2018–2024), laser intensity and alignment were not perfectly identical throughout. Notably, a critical laser system malfunction in 2022 necessitated a complete replacement of the laser assembly.

We acknowledge that the variations in laser intensity and alignment can potentially also influence the mass spectra and relative ionization efficiency (RIE) of organics, however, we argue that this effect is substantially less critical than it is for Cx fragments. This is due to the fact that the laser and tungsten vaporizers consistently operated across all field campaigns, providing a stable baseline for organic ionization that mitigates major instrument-induced variance.

Minor comments:

Line 37: Sea salt is listed as an anthropogenic source of OA. I don't understand the connection. Either delete it from the list of anthropogenic sources or include an explanation.

Response: "Sea salt" has been deleted

Line 69: "80% less aged OA (POA + SOA)" is confusing. Should that be "80% less aged OA (aged POA + SOA)"?

Response: "80% less aged OA (POA+SOA)" was referring to OA measured after the oxidation flow reactor (potential aerosol mass, PAM) that was a mixture of POA and SOA. We agree that the sentence was unclear, and therefore it was modified to be only "80% less aged OA". There is a citation for the article of Saarikoski et al. (2024) at the end of the sentence, so the details of the PAM measurements can be found there.

Line 176 (and elsewhere): The figures in the SI need to be re-ordered (and re-numbered) in the order they are mentioned in the text.

Response: Figures in supplemental material have been re-numbered according to their order in the manuscript, and the figures referred only in supplemental material have been named as Figure A and Figure B. The structure of supplemental material has also changed slightly by moving the description of source apportionment analyses for Spring 2018 and Spring 2024 data to the end of supplementary material, after all figures. We think that this change has made supplemental material easier to read.

Line 194-195 (and elsewhere): This list of m/z's is in order of highest to lowest signal but I think it would be less confusing to read if the m/z's were listed in numerical order.

Response: The order of the m/z's has been changed to follow numerical order as suggested by the Referee.

Figure 2: I would add the H:C and O:C to each panel for easier comparison among the campaigns.

Response: H:C and O:C values have been added to Figure 2.

Figure 3. I would move the TrOA label in the figure to beside the light grey oval. It is a little lost above BBOA and CoOA.

Response: TrOA label has been moved in Figure 3 as suggested by the Referee. We also found a mistake in the H:C values for LV-OOA presented in Figure 3. H:C values for LV-OOA were too large, and therefore with the new H:C values, the colored area for LV-OOA was shifted lower in vertical H:C axes. Text was not changed as there was no text related to the elemental ratios of LV-OOA.

Line 266: I am not sure what the phrase "but all oxygenated ions tend to be pronounced in the SOA mass spectra" means in this context. I think the point of the last two sentences is that although C2H5O2+ has not been observed in primary emissions it has been observed in SOA formed from those emissions. Maybe say that more directly.

Response: Text: **"**but all oxygenated ions tend to be pronounced in the SOA mass spectra" has been removed to clarify the text.

Lines 280-282: This statement isn't necessarily true unless the authors tell us what number size distribution the SCA factor has and how that compares to the size cutoff in the SP-AMS. I would also insert "mass-based" between "SP-AMS" and "measurements."

Response: The sentence "Because the SCA factor is derived from particle number size distribution measurements using DMPS, whereas TrOA is based on SP-AMS measurements, they correspond to different particle size ranges and therefore contribute differently to OA mass" was removed to make text clearer.

Line 301: Insert "(at m/z 43)" after C2H3O+ since this is the first time this ion is mentioned. In fact, it might be helpful if the authors included the m/z every time an ion is mentioned.

Response: "at m/z 43" was added, and the whole text has been corrected to be in the form ion (at m/z) when mentioned for the first time.

Figure 5. Panels a and c show a clear difference between 60 vs 73 and 61 vs 73, but I'm not sure panels b and d (in terms of f values) add to the discussion. They show the same trends but are much noisier. In addition, the f values for the PMF factors are displayed in a much better way in Figure 6. I would skip panels b and d in Figure 5 and the associated discussion.

Response: Panels (b) and (d) were deleted from Figure 5 and the text associated with them was removed.

Line 370: I am not sure what the authors mean by the CoOA factor "can be mixed" with TrOA. The time series and mass spectra are distinctly different. Maybe the authors mean "mistaken for?" Or maybe rephrase this sentence to point out the similarity in f space without drawing a conclusion.

Response: "can be mixed with" was replaced by "mistaken for" as the sentence was unclear previously.

SI Table S3: What is "episode/episodes" referring to? There should be an asterisk on Summer-Autumn 2019.

Response: The episodes are referring to long-range transport episodes described in Saarikoski et al. (2023) and Teinilä et al. (2025) in more detail. "LRT" was added before "pollution episode" in text and "long-range transport" before episode/episodes in supplemental material to clarify the episodess. An asterisk was added to Summer-Autumn 2019.

SI Figures S8 to S11: The authors could consolidate these diurnals with the figures of the PMF factors. They are only mentioned once in the text and don't need to take up so much space. What is the vertical blue line in Fig. S10a?

Response: Figures S8-11 were combined. Vertical blue line in Fig. S10a was a mistake and was removed.

SI Figure S12: I think you could skip this figure. Everything discussed with respect to this figure is easily observable in the mass spectra in Figure 2.

Response: Figure S12, and the sentence referring to it, have been removed.

SI Figure S17: I would add a point for a typical COA since that is commonly what is associated with higher fC4H7+ than fC4H9+.

Response: That was a very good idea. $fC_4H_7^+$ and fC_4H_9+ values for cooking OA have been added to Figure S11 (Figure S17 in previous version) as suggested by the Referee.

We also added text: "For the comparison, $fC_4H_7^+/fC_4H_9^+$ was also presented for cooking-related OA (Mohr et al., 2012). COA was located close to TrOA having slightly larger $fC_4H_7^+/fC_4H_9^+$ than TrOA (Fig. S11)."

Typos:

Line76: Should be "gas recirculation" not "gas circulation"

Response: "gas circulation" replaced by "gas recirculation"

Line 101: Insert "wear" after "brake"

Response: "tire wear and brake wear" replaced by "tire and brake wear"

Line 118: Delete "of" before "six"

Response: "of" deleted

Line 124: Use "after the pandemic" rather than "after the COVID-19"

Response: modified as suggested by the Referee

Line 214: Substitute "the pattern" for "that"

Response: modified as suggested

Line 281: Define DMPS

Response: The sentence regarding the DMPS measurements has been removed from the manuscript.

Throughout paper: Capitalize "Autumn" in "Summer-autumn"

Response: "Summer-autumn" has been replaced by "Summer-Autumn" is text and figures.

SI, top of page 5: Insert "with" before "the highest"

Response: "with" added

SI, page 7: "Unknow" should be "unknown"

Response: "Unknown" changed to "unknown" in Figure S5

Insert "where" before "the eighth"

Response: "where" added

"eight" should be "eighth"

Response: "eight" changed to "eighth"

Fig. S6b does not show the 8th factor gaining mass. Delete the figure reference.

Response: Figure reference deleted

Fig. S3 does not show diurnals. Should be S6 instead.

Response: Fig. S3 has been replaced by Fig. S2 as the numbering of supplemental figures has changed.

"4-factor" should be "5-factor"

Response: "4-factor" has been changed to "5-factor"

Referee #2

We thank the Referee for thorough evaluation of our manuscript. We appreciate the highly competent, and constructive comments provided. These insightful suggestions for revision have been very helpful, and we are convinced that addressing them will significantly enhance the overall quality, clarity, and impact of our manuscript. Below we respond point by point to the comments and indicate the corresponding manuscript revisions that we have made. Additionally, we corrected a few minor spelling errors we found.

Referee #2 comments

line110, the statement "its sources were identified using PMF" is somehow misleading. PMF factors in AMS data are not always direct sources, but rather chemically or process-defined components that can sometimes be linked to sources after interpretation.

Response: "sources" was replaced by "chemically distinct or process-related components"

Fig. 2. The diurnal cycles are presented as normalized concentration. Please clarify which parameter was used for normalization.

Response: Concentrations in diurnal plots were normalized to the cumulative concentrations over the whole day. Clarification was added to the figure caption.

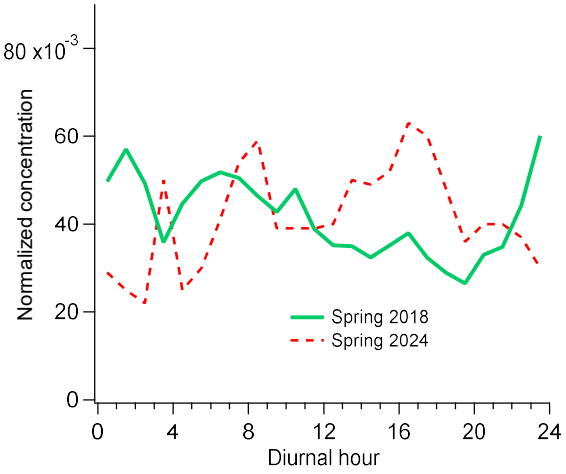
line210-213, It is difficult to attribute the relatively flat HOA diurnal pattern in spring 2018 solely to airflow and temperature. Moreover, the cycle does not appear truly flat but instead shows a noticeable peak around midnight. Are the air masses in the Helsinki region primarily influenced by southerly or southwesterly winds during the warmer season (spring to autumn)? Considering that the 2018 and

2019 campaigns represent meteorologically similar periods in terms of temperature and air mass influence, one might expect comparable HOA diurnal patterns for these two seasons.

Response: Average diurnal trends for HOA during Spring 2018 and Spring 2024, are shown in figure below. The Referee is correct that HOA peaked at midnight in Spring 2018, while during Spring 2024, the largest HOA concentrations were detected between 8 AM and 5 PM. Even though both campaigns were designated as spring campaigns, the campaign in spring 2018 was conducted much later in the season than spring 2024 campaign, and therefore spring 2018 campaign represented almost summer conditions in Finland. We didn't discuss much about meteorology during the campaigns in the article, since it was not the main topic of the paper, but average temperature and relative humidity during the campaigns were given in Table S1 in Supplemental material. It shows that the average temperature was much higher in Spring 2018 (13 °C) than in Spring 2024 (4.2 °C), as it was conducted mostly in May while the Spring 2024 campaign was conducted mostly in April.

The Helsinki region is primarily influenced by southwesterly and westerly winds but during spring also northern and northeastern winds are more common.

We added text to clarify the difference between two spring campaigns: "Although two campaigns were classified as spring campaigns, the meteorological conditions differed substantially between them. The Spring 2018 campaign was conducted later in the season than the Spring 2024 campaign, and consequently, air temperature was considerably higher during the 2018 campaign."



line213-214, it looks to me that HOA shows higher concentrations in the day than at night in the diurnal cycle (Fig. 2), contradictory to the statements here.

Response: A figure showing the average diurnal trend for HOA during Spring 2018 was given in previous response. The concentrations were smallest in the evening between 5 PM and 8 PM, and largest between 10 PM and 3 AM. The difference between day and night was not large but in general "HOA had higher concentrations at night than during the day" as stated in the manuscript.

line288-289, can you reason BBOA from a dominant local source?

Response: The dominant local source for BBOA was probably residential biomass combustion in fireplaces and sauna stoves. As mentioned in the manuscript, BBOA exhibited a pronounced evening maximum around 19:00 that coincides with the wood burning for heating purposes or warming up saunas. However, as mentioned in the manuscript, the measurement site was located at an area that was predominantly composed of apartment and industrial buildings, however, there is a residential area rather near (< 1 km) that consist of wooden houses with fireplaces and saunas. During certain wind direction and/or other meteorological conditions, the emissions from that residential area can affect the observed concentrations at Helsinki Supersite.

We modified text in Conclusions: "as there are no detached houses with fireplaces in the surrounding area" is now "as there are very few detached houses with fireplaces in the surrounding area"

line295, laser vaporizer should enable the detection refractory organic species due to the presence of BC. How does this capability affect the PMF factors compared to results from conventional thermal vaporizer results? This aspect is not discussed in the manuscript and would benefit from clarification.

Response: The Referee is correct that the laser vaporizer should enable the detection of refractory organic species due to the presence of BC. That aspect was not investigated in this article as the laser vaporizer was on all the time, not switching on/off, and the reference mass spectra with only the tungsten vaporizer was not available. The mention that the laser was on all the time has been added to text as suggested by the Referee 1.

In other studies, it has been found that the laser vaporizer increased the contribution of $C_2H_4O_2^+$ in total OA and oxygenated OA factors (Rivellini et al., 2020), while Wang et al. (2019) identified a substantial fraction of $C_2H_4O_2^+$ in BBOA when operating the AMS with a laser vaporizer only. More generally, OOA factors typically exhibit enhanced $C_2H_3O^+$ signals when measured with a laser vaporizer, whereas CO^+ and CO_2^+ signals tend to be more prominent when using a thermal vaporizer (Massoli et al., 2015; Lee et al., 2017). These details were already included in the article.

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