**Response to reviewer #1**

This study characterized the diel dynamics of various organic markers in aerosols using a TAG system at urban Hong Kong during a summer period. The high time resolution observations of organic markers allowed to identify specific sources or processes that had crucial contributions to organic aerosols. This effort promoted our understandings of the sources of organic aerosols in urban HK, and the manuscript was well written. However, there have been some studies using TAG to apportion the sources of organic aerosols at urban/suburban areas. This study just looks like a supplement of TAG data in urban HK. Therefore, I recommend a major revision on the structure of writing/descriptions to point out some interesting findings. Detailed comments are as follows:

> *We are grateful for the review and constructive comments. Revisions have been made accordingly and are highlighted in blue throughout the text. For details, please see our point-by-point responses in italics below.*

**1.** The title looks inappropriate. First, the study was only conducted in a summer period less than one month. I feel the results could not represent "An overview of organic aerosols" in urban HK. Second, as stated by the authors, the concentrations of organic markers were not quantified, and some important markers such as monoterpene derived SOA tracer were not identified. In addition, nitroaromatics, which are key brown carbon species, were not detected. Nitroaromatics may also help evaluate the contributions of oxidation products from biomass burning and vehicle emissions to organic aerosols. Third, the title did not carry impressive information.

> *Thanks for the insightful comments on the title. With these points in mind, we have modified the title as follows.*

> **In-situ measurement of organic aerosol molecular markers in urban Hong Kong: temporal variations and source apportionment in summer**

**2.** The manuscript is mainly discussing the influence of different air masses (continental, coastal, and marine) on organic markers and OM sources. However, looking at Figure 3, there was only one day when air mass was from continental regions; only two days when air mass was from coastal regions and there was a one-day long maintenance of instruments when air mass was from coastal regions. One-day long observation is not representative. Therefore, grouping the data by continental, coastal, and marine air masses is not appropriate. For example, it is odd that cooking emissions had an insignificant contribution to OM when air mass was from continental region if the sampling site is surrounded by restaurants. This may be due to cooking plums did not significantly affect the sampling on just that day. It is feasible to regard the periods when continental and coastal winds were prevailing as cases to discuss the influences on organic markers but cannot regards the results as common situations when continental and coastal air mass dominated.

*Indeed, the number of days with continental or coastal air was too few to be representative. We treat them as special cases, and the related discussions are revised in the text.*

> However, the fraction of air masses originating from and passing over the mainland was very low (6%, red trajectories), same for those arrived at the site along the coastline of eastern and southern China (7%, orange trajectories). From a representative perspective, we discuss them as special cases, and the continental air and coastal air are labelled as Case I and Case II, respectively.

*For details, please refer to lines 87-90 and other places with Case I and Case II.*

*The low contributions of primary cooking emissions in the cases with continental and coastal air could be due to i) more conversion through oxidation, ii) uneven distribution of restaurants, and iii) sampling bias, which are discussed as follows.*

> However, the levels of PCOA markers were much lower in the cases with continental and coastal air than in the marine air. On one hand, this could be partially explained by the quicker chemical losses, due to the higher concentrations of oxidants, *e.g.,* $O_3$ (Table S3). Oleic acid with a carbon-carbon double bond in the molecule can be efficiently oxidized by $O_3$ and other oxidants (Zeng et al., 2020; Wang et al., 2021). The argument is supported by the fact that 9-oxononanoic acid, a typical OCOA tracer (Lyu et al., 2021), in the two cases was 1.47 – 2.00 times the level in the marine air. On the other hand, the differences could be related to the uneven distribution of restaurants on different air paths. As shown in Figure 1, the restaurant density in the areas where the continental and coastal airflows passed is substantially lower than south of the sampling site. Besides, it cannot be ruled out that the limited number of samples in the two cases coincidently did not capture the cooking emissions well.

*For details, please refer to lines 195-203.*

> Although the mass concentration of $PM_1$-OM attributed to OCOA in Case I (0.67 μg $m^{-3}$) and Case II (0.73 μg $m^{-3}$) was higher than in the marine air (0.47 μg $m^{-3}$), the percentage contributions were much lower (6.5% and 9.3% in Case I and Case II, respectively). Overall, the SOA contributions increased significantly with the rise in $PM_1$-OM concentration (Figure S10), consistent with the findings at a suburban site that $PM_{2.5}$ pollution events witnessed a notable rise in SOA tracer levels (Wang et al., 2022). However, it is noteworthy that PCOA made much less and even negligible contributions to the $PM_1$-OM in the two cases. The reasons might be the same as those for the deficiency of PCOA markers, as discussed in section 3.1.

*For details, please refer to lines 338-344.*

**3.** For the PMF analysis, it is not a good way to keep a factor which is unexplainable. The authors are encouraged to improve the PMF analysis by modifying inputs or others. As highlighted by the authors, cooking emissions should be a major source of OM at the

sampling site, while the contribution from cooking oxidation products to OM could not be evaluated. A previous study (Huang et al., Comparative Assessment of Cooking Emission Contributions to Urban Organic Aerosol Using Online Molecular Tracers and Aerosol Mass Spectrometry Measurements, Environ. Sci. Technol. 2021, 55, 14526-14535) used azelaic acid, nonanoic acid, and 9-oxononanoic acid to indicate the oxidation of cooking emissions. I noticed azelaic acid has been detected in this study (No. 20 in Figure 2). The authors may try to conduct an analysis to evaluate how significant of cooking oxidation is in contributing to OM mass.

*Excellent comments, especially regarding the aged cooking organic aerosol. We have made some adjustments based on the suggestions here and below and re-run the PMF model.*

*In this sampling campaign, both azelaic acid and 9-oxononanoic acid were detected by the TAG. Azelaic acid and oleic acid were similar in time series and diurnal patterns, therefore a moderate correlation between them ($R^2$ = 0.57), indicating that a considerable fraction of azelaic acid was derived from primary cooking emissions and/or quick formation from primary emissions. No similar result was found for 9-oxononanoic acid. However, the diurnal pattern of azelaic acid to oleic acid was quite consistent with that of 9-oxononanoic acid to oleic acid. In particular, the peaks several hours after the evening peak of oleic acid showed a clear evidence that primary cooking emissions experienced chemical aging. Based on these, we selected 9-oxononanoic acid as the tracer of aged cooking organic aerosol. In fact, Huang et al. (2021) also found that the loading of 9-oxononanoic acid in the factor of aged cooking organic aerosol was notably higher than that of the other tracers (e.g., azelaic acid, nonanoic acid).*

*To identify the factor of vehicle emissions, we used a hopane species as the tracer in the original manuscript. However, due to the lack of authentic standard, we do not have enough confidence in the identification of this hopane. Moreover, it did not show any correlation with $NO_x$ or any peak in the morning or evening rush hours. Hence, we have reservations about using this species. Instead, we adopt $NO_x$ as the tracer of vehicle emissions. First, the diurnal pattern of $NO_x$ was very similar to that observed at a roadside site 350 m away. Second, there was no correlation between $NO_x$ and levoglucosan, the biomass burning tracer, even in the hours with elevated levels of levoglucosan.*

*The new tracers, i.e., 9-oxononanoic acid, azelaic acid and $NO_x$, are discussed in before the source apportionment.*

*Lines 182-189:*

Although the marine air was supposed to be relatively clean, it contained elevated levels of $SO_2$ and $NO_x$, which correlated well with each other ($R^2$ = 0.76). This phenomenon was in line with that at the three roadside sites in HK, but was not identified at the general urban sites (Figure S3). Therefore, the close relationship between $SO_2$ and $NO_x$ on the days with marine air were likely attributable to local vehicle emissions rather than ship emissions. For all the air masses, we also note that

the variation of $NO_x$ throughout the day was highly consistent with that observed at a nearby roadside site (Figure S4). Moreover, there was no correlation between $NO_x$ and levoglucosan, even during the period with elevated levels of levoglucosan, eliminating the likelihood of high biomass burning contribution. Therefore, $NO_x$ is regarded as a tracer of vehicle emissions in this study.

*Lines 244-247:*

Oleic acid was obviously enhanced at noon and in early evening, when intensive cooking activities took place in restaurants and at home, same for fatty acids, azelaic acid, and fructoses (Figure S7). The consistency of azelaic acid with oleic acid ($R^2$ = 0.57) indicated that a considerable fraction of the azelaic acid was derived from or quickly formed in primary cooking emissions.

*Lines 253-257:*

As a tracer of OCOA, 9-oxononanoic acid got higher levels at night. This was in line with the observations in an indoor air quality study (Lyu et al., 2021), and was due to the $O_3$-initiated oxidation of cooking emissions that peaked in the early evening. Although the diurnal pattern of 9-oxononanoic acid was totally different from that of oleic acid and azelaic acid, the variation in the ratio of 9-oxononanoic acid to oleic acid was consistent with that in the azelaic acid to oleic acid ratio (Figure S9), both reflecting the chemical aging of cooking emissions.

*With these changes, there is no unexplainable factor, and we got a factor representing oxygenated cooking organic aerosol (OCOA). Please refer to section 3.3 for details of the revisions.*

**4.** It was interesting to identify the periods lasting a few days when trough played a role. The meteorological parameters such as RH, Temperature, solar radiation, wind speed showed significant differences between "trough" days and "non-trough" days. I think it is valuable to focus on discussing the variations of organic tracers, especially SOA tracers, during the two distinct periods. It is good to see that in the manuscript the authors have pointed out the increases of phthalic acid and DCAs during "trough" days, indicating an aqueous formation of the species. The authors are encouraged to find more markers that had significant difference between the periods. Hopefully, the authors can evaluate how significant of aqueous formation is in contributing to OM mass. This would be a very interesting part.

*Thanks. In fact, a wide range of POA and SOA tracers experienced an increase on the 'trough' days. We discussed some of the increases in different parts of the manuscript, such as nocturnal enhancement of 2-MT (section 3.2), positive response of phthalic acid to the product of radiation and relative humidity (section 3.3), and potentially aqueous photochemical formation of OHDCA (section 3.3). The revised manuscript integrates these discussions and includes some additional discussions on other species. However, the*

*discussions on aqueous processes are speculative, and it is hard to quantify the contributions to PM$_1$-OM.*

In fact, the troughs witnessed enhancements of a wide range of species relative to their levels in the marine air without troughs. The highest change ratio for the POA markers was 5.3 for benzo[a]pyrene. However, more significant increases were observed for many SOA markers, including DHOPA, C$_5$-alkenetriols, OHDCA species, and phthalic acid, implying the role of processes other than transport and atmospheric mixing. It is noteworthy that the troughs featured lower temperature and much weaker radiation, while the RH and LWC were higher. A recent study demonstrated the aqueous photochemical formation of OHDCA in HK (Huo et al., 2024). While it is unknown whether this was also responsible for the increases of the other SOA markers, a moderate correlation (R$^2$ = 0.41) was identified between phthalic acid and the product of RH and UV during the trough periods and was not found at other times (Figure S6). Besides, the rise in C$_5$-alkenetriols resembled the changes in 2-MTs, which mainly occurred at night and could also be linked to aqueous processes, as discussed below. Therefore, the high RH and LWC might be one of the leading factors, if not all, of the SOA enhancements in the presence of troughs.

*For details, please refer to lines 224-234.*

**5.** Some inconsistencies may exist. For example, in Figure 7a, cooking emissions (dark yellow) constituted a significant part of OM sources during June 15th to 17th (continental and coastal air masses dominated according to Figure 3), while in Figure 7b and text, cooking emission was a very minor source of OM during the period. Another example is the X axis of Figure S8 shows a OM range of 0-22 μg/m$^3$ while the Y axis of Figure 7a only reached 15 μg/m$^3$. If X axis of Figure S8 shows the observation result, then there should be an unresolved percentage in Figure S8.

*We sincerely appreciate the careful review. The errors have been corrected. Please refer to the updated Figure 7, Figure S10 and discussions in the text.*

**6.** line 185-188: the explanation may need to be modified. Just like I mentioned above, one-day observation for continental air mass dominated period may not capture the influence from cooking emission. During fall-winter period, most air masses may come from the north, and I guess you may still find the contribution from cooking emissions if a long period of observation is available.

*As stated above, the reasons for lower levels of cooking emission tracers in the two cases with continental and coastal air could be multifaceted. The explanation has been revised, and please refer to our response to comment #2.*

**7.** line 227: why not examine the correlation between pyrene and hopanes? $NO_x$ can also be emitted from biomass burning.

*We tried but did not see any correlation between them. In fact, this comment reminded us to revisit the hopane data. As stated in the response to comment #3, we do not have enough confidence in the species identification and data quality. Instead, NOx seemed to be a better indicator of vehicle emissions.*

**8.** line 290: add "respectively" after $PM_1$-OM.

*Accepted with thanks.*

**9.** line 318: please try to add azelaic acid in PMF analysis to see if cooking oxidation factor can be resolved.

*As stated above, we use 9-oxononanoic acid as the tracer of OCOA. Please see our response to comment #3 for details.*

**10.** Figure 1. How about show the locations of restaurants in the map?

*Accepted. We show a heatmap of restaurant distribution in HK. Please see the updated Figure 1.*

**11.** Figure 3. Please add the time series of DCAs.

*Thanks for the suggestion. We have added the time series of succinic acid, a representative DCA species we measured. Its variations are also briefly discussed. Please see the updated Figure 3.*