***Response to Review Comments by Anonymous Referee #1 on "Chemical evolution of secondary organic aerosol tracers during high PM2.5 episodes at a suburban site in Hong Kong over 4 months of continuous measurement" by Q. Wang et al.***

**General Comments by Anonymous Referee #1:**

This manuscript reported a four-month's continuous bihourly measurement of various types of secondary organic aerosol (SOA) tracers originated from common specific volatile organic compounds (VOCs). The authors focused on the concentration change of the SOA tracers during high $PM_{2.5}$ episodes and found obvious mass increment of the SOA tracers during the episodes especially in summer and fall, suggesting enhanced SOA formation during the episode. Moreover, the chemical formation mechanism and ageing of the biogenic SOA formation during the episodes were examined with the measurement of multiple molecular tracers from single VOC precursor, which provide valuable insight into the formation mechanism under ambient atmosphere similar to this study area. In general, this manuscript is well written, and the dataset is large. The manuscript can be accepted with the following revisions:

**Response to General Comments:** We thank the reviewer for the comments and acknowledgment of the importance of our work. Below is our point-by-point response to each comment, marked in blue. Changes to be made to the main text are also marked in blue in the revised manuscript file.

1. As the focus of this work is to examine the SOA tracers, while most of which lack authentic standards for the quantification. The authors should elaborate more on how the identification and quantification was achieved (such as the retention time, quantification ions, etc.).

   **Response:** The measurement of individual SOA tracers via derivatization followed by GC/MS method have been well documented in previous studies. The mass spectra of the SOA tracers studied in this work have been reported in previous studies, for example, Claeys et al. (2004) for isoprene SOA tracers, Szmigielski et al. (2007) for α-pinene SOA tracers, Jaoui et al. (2007) for β-caryophyllinic acid, and Al-Naiema and Stone, (2017) for DHOPA. In this work, the SOA tracer species without authentic standards were identified by comparing their ambient mass spectra with the previously reported data and quantified by using surrogate compounds with similar structures and functional groups. We've added the retention time and quantification ions for all the measured organic markers in Figure S1.

   **Lines 106-111:** "Levoglucosan, 4-nitrocatechol, phthalic acid and pinonic acid, with available authentic standards, were identified and quantified by directly comparing to their standards. The remaining SOA tracers which do not have authentic standards were identified by comparing their ambient mass spectra with the previously reported data (Al-Naiema and Stone, 2017; Claeys et al., 2004; Jaoui et al., 2007; Szmigielski et al., 2007) and quantified using the surrogate compounds with similar structures and functional groups. The retention time and quantification ions of each species are shown in Figure S1."

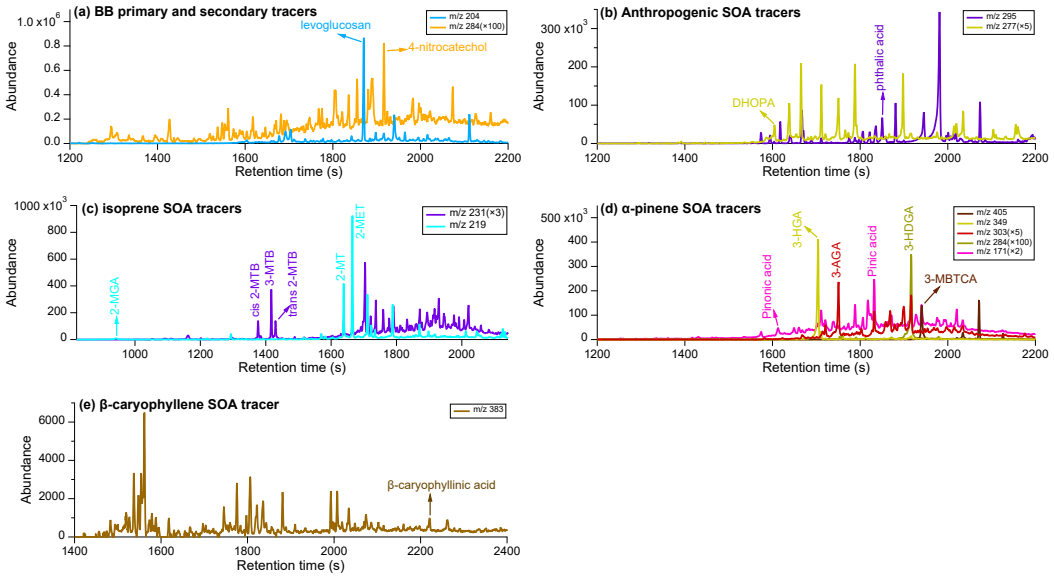


**Figure S1.** Example of the extracted ion chromatograms from select ambient sample (4 Sep. 2020 4:00 am) for the target organic markers studied in this work.

2. The SOA tracers are widely used to estimate the SOC contribution either by the SOA tracer method or by receptor models such as Positive Matrix Factorization (PMF). Since the comprehensive measurement data are available, do the authors plan to conduct such analysis to get a more quantitative estimation of the SOA contribution?

   **Response:** Yes, we have a plan to conduct a more quantitative source analysis using PMF of a longer dataset and this will be presented in a future publication.

3. Lines 190-194: Why the authors only selected the "before-episode period" to evaluate the evolution of SOA tracers, instead of using all measured data?

   **Response:** The campaign-wide average is not representative to the normal condition specific to individual seasons, as the emission sources and meteorological conditions varied among seasons. The selection of the "before-episode period" can minimize the interference from the different meteorological conditions such as temperature and boundary layer height among seasons. This comparison (i.e., before-episode period vs. episodic period) can better examine the rapid formation of the high PM episodes.

4. The date format in the main text and Tables is not consistent. For example, "10 Jul. -7 Oct" in Line 135 vs. "Sep-01 12:00 PM" in Table 1. Please check and unify the format.

   **Response:** the time format in Table 1 and Table S1 and elsewhere have been revised accordingly. Please see the updated Table 1.

**Table 1.** Statistical summary of $PM_{2.5}$ at 15 HKEPD air quality monitoring stations and the HKUST supersite during the 11 episodes and the remaining non-episodic hours during the period of 10 Jul.-31 Dec. 2020. Meteorological parameters and gas pollutant data are from HKUST supersite.

| Episodes | Season | Time period | Duration (h) | Wind speed ($m\ s^{-1}$) | T (°C) | RH (%) | $O_3$ (ppb) | $NO_x$ (ppb) | Avg. $PM_{2.5}$, $\mu g\ m^{-3}$ | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | | | | | City-wide avg. | Min. | Max. | Max-to-Min ratio |
| EP1 | Summer | 1 Sep 12:00 PM – 4 Sep 3:00 PM | 76 | 1.33 | 29.7 | 78.0 | 63.4 | / | 37.6 | 32.1 | 44.2 | 1.4 |
| EP2 | Fall | 30 Oct 7:00 AM - 7:00 PM | 13 | 2.72 | 23.2 | 79.0 | 48.5 | 12.6 | 32.6 | 26.1 | 39.9 | 1.5 |
| EP3 | Fall | 2 Nov 7:00 AM – 4 Nov 9:00 PM | 63 | 3.93 | 23.2 | 65.5 | 57.6 | 12.0 | 30.7 | 26.6 | 37.8 | 1.4 |
| EP4 | Fall | 6 Nov 11:00 AM – 10 Nov 10:00 PM | 108 | 3.36 | 24.2 | 56.9 | 69.0 | 13.1 | 33.0 | 28.4 | 41.0 | 1.4 |
| EP5 | Fall | 24 Nov 12:00 PM - 7:00 PM | 8 | 3.44 | 22.4 | 79.1 | 58.4 | 8.22 | 29.7 | 22.1 | 43.0 | 1.9 |
| EP6N | Fall | 26 Nov 4:00 PM – 27 Nov 1:00 AM | 10 | 1.36 | 20.8 | 89.6 | 58.5 | 7.20 | 29.9 | 17.2 | 44.9 | 2.6 |
| EP7N | Winter | 3 Dec 1:00 AM - 10:00 AM | 10 | 6.13 | 15.6 | 67.7 | 26.2 | 11.2 | 31.0 | 24.3 | 38.9 | 1.6 |
| EP8 | Winter | 5 Dec 2:00 AM – 13 Dec 0:00 AM | 191 | 2.59 | 18.7 | 71.9 | 41.8 | 13.9 | 33.2 | 25.6 | 47.9 | 1.9 |
| EP9 | Winter | 19 Dec 1:00 PM – 25 Dec 10:00 PM | 154 | 3.62 | 16.3 | 68.4 | 38.2 | 11.3 | 28.4 | 20.6 | 41.9 | 2.0 |
| EP10 | Winter | 27 Dec 11:00 AM – 28 Dec 11:00 AM | 25 | 1.89 | 20.5 | 58.0 | 61.7 | 6.69 | 28.9 | 15.7 | 66.7 | 4.3 |
| EP11N | Winter | 29 Dec 8:00 PM – 30 Dec 4:00 AM | 9 | 3.70 | 18.5 | 72.1 | 32.2 | 21.3 | 33.8 | 22.8 | 43.4 | 1.9 |
| non-EP | / | 10 Jul 0:00 AM – 31 Dec 11:00 PM | 3533 | 2.88 | 25.0 | 78.9 | 44.8 | 9.10 | 12.5 | 10.4 | 16.9 | 1.6 |

5. Figure 1: there are only two sites showing numbers near the column plot in the figure, are they the concentration values stated in the figure caption? How about the other sites?

**Response:** Yes, the numbers indicate the average $PM_{2.5}$ concentrations during the non-episodic period for each site (Figure 1). Previously, when converting the word file to pdf files, the numbers somehow were erased. We will fix the problem in the revised manuscript.

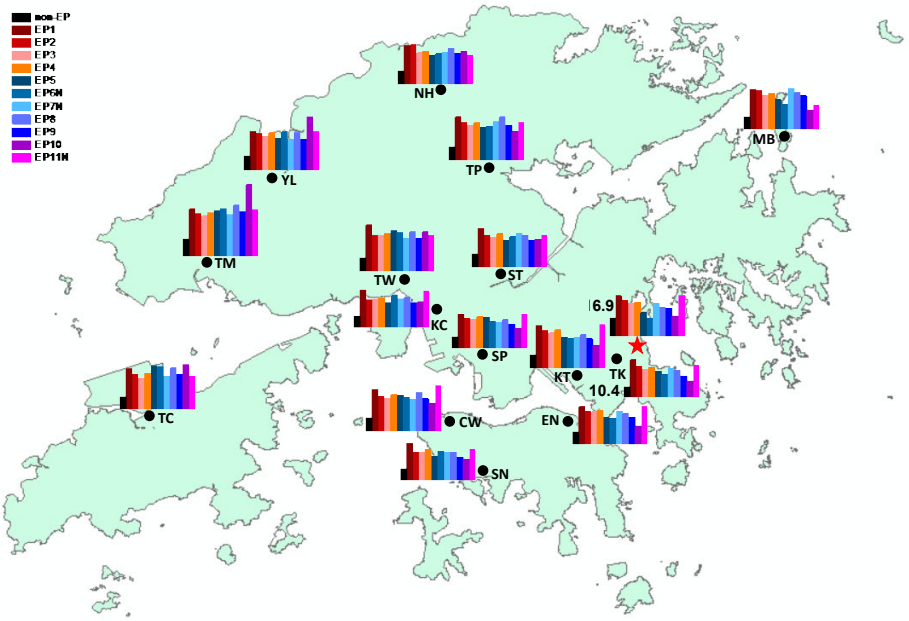


**Figure 1.** Geographical location of the 15 HKEPD general air quality monitoring stations (black dot) and the HKUST supersite (red star) in HK. Among the 15 HKEPD stations, MB is the rural site while others are general urban stations with different microenvironments. Column plot shows the average $PM_{2.5}$ concentrations ($\mu g\ m^{-3}$) during non-episodic hours (in black, with numbers indicating the concentration values) and the eleven episodes (in various colors).

6. Figures 5-6: the figure numbers "(a)" and "(b)" in the figure are missing.

   **Response:** The figure numbers will be added.

7. Figure S8: The axis of Figure S8 is not readable.

   **Response:** The revised Figure S8 (now become Figure S9) is copied below.

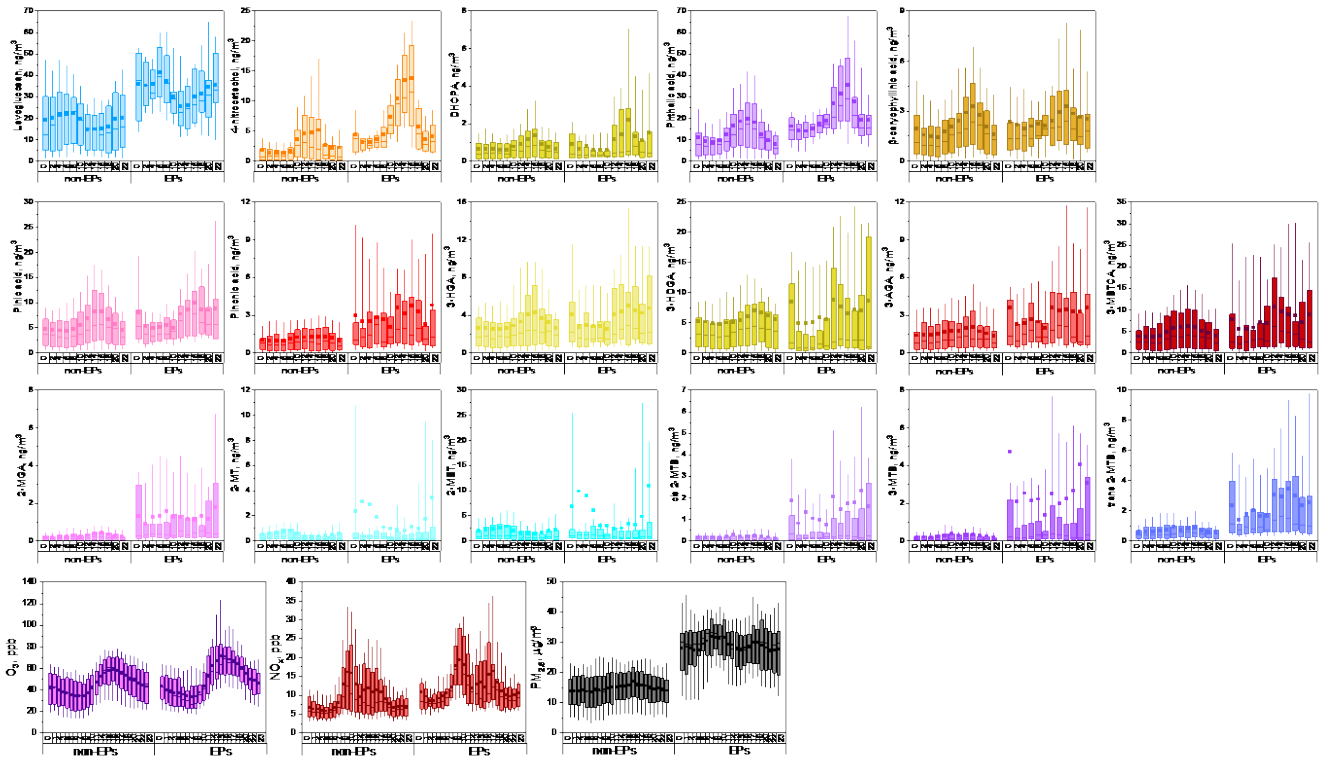

**Figure S9:** Diurnal variations of (a) levoglucosan, 4-nitrocatechol, DHOPA, phthalic acid, and β-caryophyllinic acid; (b) individual α-pinene SOA tracers; (c) individual isoprene SOA tracers, and (d) $O_3$, $NO_x$ and $PM_{2.5}$ under the non-episodic and episodic period from 30/8-31/12, 2020 at HKUST supersite.

8. Line 193: "were" should be "are".

   **Response:** Suggestion taken.

9. Line 230: "in Chow et al. 2016)." should be "in Chow et al. (2016))."

   **Response:** Suggestion taken.

10. Lines 297-298: The description is not consistent with the content of Figure 5. At EP6N, isoprene SOA tracer concentrations were higher in the time period before the episode.

    **Response:** we have revised the statement:

    **Lines 306-307: "**The isoprene SOA tracers were consistently higher during episodes than the before-episode periods except for one nighttime episode-EP6N (Figure 5)."

11. Lines 374-379: The description is not consistent with Figure 7.

    **Response:** The reviewer may have misunderstood the statement. Here we want to discuss the less decrease in concentration in winter compared with summer and fall for β-caryophyllinic acid (i.e., less seasonal contrast). We've re-phrased the statement as following:

    **Lines 383-385: "**It is plausible that the less decrease in concentration of β-caryophyllinic acid in both non-episodic and episodic periods in winter compared with those in summer and fall could be a result of the lower atmospheric oxidative capacity in winter leading to less degradation of β-caryophyllinic acid."

12. Line 403: "2-methyl tetrols" should be "2-methyltetrols".

    **Response:** revised as suggested.