RC2: <u>'Comment on acp-2022-220'</u>, Anonymous Referee #2, 18 Apr 2022

Review on "Measurement Report: Effects of anthropogenic emissions and environmental factors on biogenic secondary organic aerosol (BSOA) formation in a coastal city of Southeastern China" for Hong et al.

The author conducted the field observation during summer and winter in the southeast of China, and discussed the formation of SOA tracers, especially BSOA tracers. The author found that the concentrations of SOA tracers were affected by photochemical oxidation in summer, and were affected by anthropogenic emissions in winter. They highlighted that anthropogenic emissions, atmospheric oxidation capacity and halogen chemistry have significant effects on the formation of BSOA in the southeast coastal area. The manuscript can provide unique data for SOA tracers in the coastal area, and clarified the influencing factors on SOA formation. However, there are still some content deficiencies and logical omissions in this manuscript, which need to be carefully revised. Overall, the manuscript could be accepted after addressing the following issues.

<u>Response:</u> Thank you very much for all the valuable comments and suggestions. We have addressed each comment in the following point by point and have revised the manuscript accordingly.

1. Line 147-149. How many times the samples were ultrasonically extracted during the pre-treatment, it should be shown in the manuscript.

<u>Response:</u> Thank you for your suggestions. The sentence was changed as follows:

Briefly, the filter samples were ultrasonically extracted with a mixture of dichloromethane and methanol (2:1, v/v) for 10 min three times.

2. Line 189-190. f_{soc} of isoprene was 0.155 ± 0.039 in study of Kleindienst et al., 2007, the author should recheck your content.

Response: Thank you for your suggestions. Corrected.

3. Section 2.5. The authors use both E-AIM IV model and ISORROPIA II model to calculate the aerosol pH. They need to discuss the correlation and difference between the results of two models, and explain which result is more reasonable for this manuscript. The authors should also explain which model they chose for the following discussions.

<u>Response:</u> Thank you for your good comments and suggestions. As the reviewer mentioned, E-AIM IV model and ISORROPIA II model are usually used to calculate the aerosol acidity. In this study, we compare them with each other. The comparison of H⁺insitu calculated by EAIM IV and ISORROPIA II were illustrated in the

following figure. We found that the H⁺insitu derived from ISORROPIA II agreed perfectly with those from E-AIM IV, and their trends matched perfectly with each other. For the two thermodynamic models, ISORROPIA II is widely used owing to its rigorous calculation, performance, and computational speed. Therefore, the results of ISORROPIA II calculation was just demonstrated in this study. To avoid the misunderstanding from the readers, we have deleted the introduction details of EAIM IV calculation.

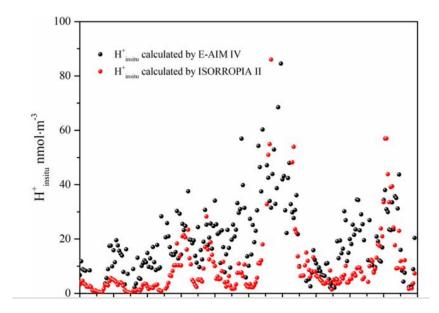


Figure Comparison of H⁺insitu calculated from E-AIM IV and ISORROPIA II.

The paragraph was rewritten as follows:

The forward mode of ISORROPIA II thermodynamic model was used to calculate the aerosol acidity (pH) (Fountoukis and Nenes, 2007). ISORROPIA II can calculate liquid water content (LWC), based on total $SO_4^{2^-}$, NO_3^- , CI^- , ammonia, non-volatile cations (Na⁺, K⁺, Ca²⁺, Mg²⁺), and meteorological factors (RH and T) (Rumsey et al., 2014; Guo et al., 2016). The pH value from ISORROPIA II was calculated using the following equation:

$$pH = - \ lg \biggl(\frac{1000 \times H^+}{LWC} \biggr)$$

where H^+ is the hydronium ion concentration loading for an air sample ($\mu g/m^3$).

Fountoukis, C., and Nenes, A.: ISORROPIA II: a computationally efficient thermodynamic equilibrium model for K^+ -Ca²⁺-Mg²⁺-NH₄⁺ -Na⁺-SO₄²⁻ -NO₃⁻-Cl⁻-H₂O aerosols, Atmos. Chem. Phys., 7, 4639-4659, 10.5194/acp-7-4639-2007, 2007.

Guo, H., Sullivan, A. P., Campuzano-Jost, P., Schroder, J. C., Lopez-Hilfiker, F. D., Dibb, J. E., Jimenez, J. L., Thornton, J. A., Brown, S. S., Nenes, A., and Weber, R. J.: Fine particle pH and the partitioning of nitric acid during winter in the northeastern United States, Journal of Geophysical Research: Atmospheres, 121, 10,355-310,376, https://doi.org/10.1002/2016JD025311, 2016.

Rumsey, I. C., Cowen, K. A., Walker, J. T., Kelly, T. J., Hanft, E. A., Mishoe, K., Rogers, C., Proost, R., Beachley, G. M., Lear, G., Frelink, T., and Otjes, R. P.: An assessment of the performance of the Monitor for AeRosols and GAses in ambient air (MARGA): a semi-continuous method for soluble compounds, Atmos. Chem. Phys., 14, 5639-5658, 10.5194/acp-14-5639-2014, 2014.

4. Section 3.1. In my opinion, it is clearer to list the average concentrations of these air pollutants during summer and winter, daytime and nighttime in Supporting Information as a Table.

<u>Response:</u> Thank you for your good suggestions. The details have been shown in Table S1.

Index	Winter		Summer	
	Daytime	Nighttime	Daytime	Nighttime
$PM_{2.5}(\mu g/m^3)$	40.3±18.7	45.1±17.0	19.4±9.70	14.1±6.00
$PM_{10}(\mu g/m^3)$	61.1±27.2	68.9 ± 25.0	36.5±17.5	30.3±9.70
$O_3(\mu g/m^3)$	45.7±25.4	37.6±16.8	80.3±46.2	24.2±11.8
$CO(mg/m^3)$	0.70 ± 0.10	0.70 ± 0.10	0.30±0.10	0.30±0.10
$SO_2(\mu g/m^3)$	2.90 ± 1.80	2.10±0.90	8.30±1.00	$7.80{\pm}1.40$
$NO_2(\mu g/m^3)$	33.0±8.50	32.3±9.00	12.2 ± 6.50	18.7 ± 7.40
T(°C)	16.8 ± 2.60	14.6 ± 1.70	36.0±2.70	31.2±1.00
P(kPa)	100.9 ± 0.20	100.9±0.20	99.5±0.20	99.6±0.20
RH (%)	60.7±9.50	69.5 ± 5.80	55.0±6.90	67.7±3.30
WD(°)	159.0±14.3	151.3±12.7	191.5±16.9	194.0 ± 30.8
WS(m/s)	1.50 ± 0.40	1.10 ± 0.70	1.40 ± 0.30	0.80 ± 0.20

Table S1 Comparisons of criteria air pollutants and meteorological parameters during the daytime and nighttime in winter and summer

 Line 250. The average concentrations of SOA_M, SOA_I and SOA_c in winter and summer should be given. As the author determined to discuss "total SOA tracers" (Line 249), the concentration of ASOA should also be shown here.

<u>Response:</u> Thank you for your good comments. These sentences have been added in the revised manuscript, as follows:

The average concentrations of total SOA tracers in winter and summer were 37.3 and 111.3 ng m⁻³, respectively. The predominance of SOA_M (26.6 ng m⁻³), followed by ASOA (4.60 ng m⁻³), SOA_I (4.35 ng m⁻³) and SOA_C (1.76 ng m⁻³) was observed in winter while SOA_I (54.4 ng m⁻³) and SOA_M (47.8 ng m⁻³) in summer were the main contributors to total SOA tracers, followed by ASOA (6.64 ng m⁻³) and SOA_C (2.45 ng m⁻³).

6. Line 250-252. The author showed that "In summer, BSOA tracers showed much higher concentrations in the daytime than in the nighttime, while inverse results were observed in winter", the specific concentrations of BSOA tracers in daytime and nighttime of summer and winter should be displayed here.

Response: Thank you for your suggestions. Corrected.

In summer, BSOA tracers showed much higher concentrations in the daytime (149.3 ng m⁻³) than in the nighttime (60.1 ng m⁻³), while inverse results were observed in winter (30.4 ng m⁻³ and 35.0 ng m⁻³ in the daytime and nighttime, respectively)

 Line 252-258. Instead of using "for example" here, the author could display the average concentrations of SOA tracers (including SOA_I, SOA_M, SOA_c and ASOA tracers) during day, night, summer and winter in the Supporting Information as a Table directly.

<u>Response:</u> Thank you for your good suggestions. The details have been shown in Table S2.

Table S2 Comparisons of different types of SOA tracers (ng m^{-3}) during the daytime and nighttime in winter and summer

SOA tracers	Winter		Summer	
	Daytime	Nighttime	Daytime	Nighttime
SOAI	3.79±2.37	4.91±3.75	81.9±66.2	26.8±24.8
SOA _M	24.9±8.51	28.3±13.0	64.5±38.5	31.2±27.2
SOA _C	1.70 ± 0.81	1.82 ± 0.77	2.83±1.97	2.06 ± 2.11
Sum of BSOA	30.4±11.1	35.0±17.1	149.3±96.9	60.1±52.9
ASOA	3.80±1.99	5.35±2.72	9.00±5.98	4.28±2.96
Total SOA	34.2±12.8	40.4±19.6	158.3±102.5	64.4±55.8

8. Line 275-279. As the concentrations of SOA tracers were higher in summer than winter, and the f_{soc} values were constant in this manuscript, it was not surprisingly that the concentrations of SOC in summer was higher than that in winter. And this result could not demonstrate that the contributions of SOA tracers to SOC in summer was higher than those in winter.

<u>Response:</u> Thank you for your comments. The sentence has been revised as follows:

The concentrations of SOC in summer was higher than that in winter, attributed to the increase of flourishing vegetation emissions and photochemical reactions under high temperature and strong solar radiation conditions.

9. Line 283-286. This sentence is confusing, why does the "obvious trend of diurnal variations of SOC_i" was "consistent with the isoprene emission", and why this result was compared with the trend in winter? Considering the coherence of context, maybe the author intended to explain the diurnal variation of SOC_i was obvious in summer and the variation was consistent with isoprene emission in summer? The authors should give more explanation about it.

<u>Response:</u> Thank you for your kindly comments. Exactly, as the reviewer mentioned, we try to demonstrate the diurnal variation of SOC_I was obvious in summer and the variation was consistent with isoprene emission in summer. We analyze the diurnal variation of isoprene concentrations during the wintertime and summertime, as shown in Fig.S3. These sentences have been rewritten in the revised manuscript, as follows:

An obvious trend of diurnal variations of isoprene-derived SOC in summer was observed, which was consistent with the diurnal pattern of isoprene concentration (Fig.S3). However, no similar trend was found in winter, attributed to the influence of low temperature on inhibiting the emissions of isoprene from various kinds of plants.

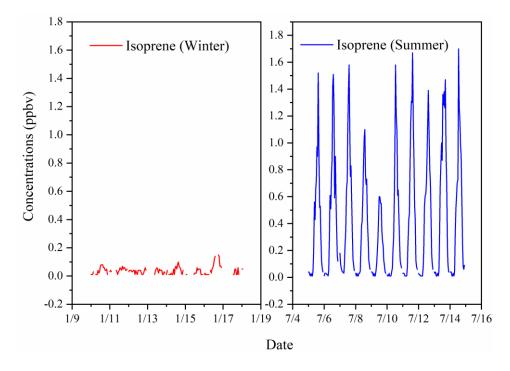


Fig.S3. Diurnal variation of isoprene concentrations during the wintertime and summertime

10. Figure 3. The legend of Figure 3 might be SOC₁, SOC_M, SOC_c and ASOC. <u>Response:</u> Thank you for your kindly comments. Corrected.

11. Line 306, it should be "SOA tracers", and Line 308, it should be "SOA tracers".

Response: Corrected.

12. Line 319. I think the first (PA and PNA) and later generation (HGA, AGA, HDMGA and MBTCA) products could only evaluate the aging degree of SOA_M, not all BSOA.

<u>Response:</u> Thank you for your comments. The sentence has been rewritten as follows:

The first (PA and PNA) and later generation (HGA, AGA, HDMGA and MBTCA) products were used to evaluate the aging degree of SOA_{M} .

13. Line 333-335. According to the logic of this section, it might be "Low ratio of HGA/MBTCA (~1.0) showed that α -pinene was the major precursor for SOA_M. The ratio of HGA/MBTCA with an average of 5.78 in Xiamen was high, suggesting the contribution of β -pinene to SOA_M".

Response: Thank you for your comments. Corrected.

14. Line 362. The author used the pH values calculated by ISORROPIA II here. Same as the Q3, the author should explain why they chose the pH calculated by ISORROPIA II, but not that calculated by E-AIM IV.

<u>Response:</u> Thank you for your kindly comments. As mentioned in Q3, for the two thermodynamic models, ISORROPIA II is widely used owing to its rigorous calculation, performance, and computational speed.

15. Line 380. Table 1 should be listed after this paragraph, which refers to table 1 for the first time.

Response: Thank you for your comments. Corrected.

16. As the contents of Figure 6 and Table 1 are similar, and the author has not discussed Figure 6 in detail, this figure should be moved to the supporting information section.

<u>Response:</u> Thank you for your kindly suggestions. Figure 6 was moved to the SI section, named Fig.S4.

17. Line 425-427. The author showed that "the correlations of SOA tracers in winter were found to increase with increasing NH₃ and chlorine ions in PM_{2.5}, while inverse results were observed in summer". The sentence is not rigorous, because NH₃ was not negative correlated with SOA tracers in summer as shown in Table 1.

<u>Response:</u> Thank you for your good comments. The correlations between SOA tracers and NH_3 was discussed in 3.6. The sentence has been rewritten as follows:

As shown in Table 1, most of SOA tracers in winter were correlated with the concentrations of chlorine ions in $PM_{2.5}$, while inverse results were observed in summer.