Response to Reviewer #2

We are grateful to the reviewer for the thoughtful comments, which are very helpful for improving our manuscript. Our point-to-point responses to each comment are as follows (the reviewer's comments are in black text, our responses are in blue text, and revised texts that appear in the manuscript are in red text).

General Comments:

The manuscript by Hu et al. presents interesting new results on isoprene-derived SOA in a megacity in China and the response to the on-going reductions in anthropogenic emissions. In general, the results are well presented and discussed. The study includes both measurements and modelling results. One limitation, which the authors should keep in mind, is that the field measurements are conducted on a limited number of samples. The modelling approach balances this, but the text sometimes overstates the conclusions that can be drawn.

Specific comments:

Line 21: profoundly is a somewhat strange word to use here.

Response: We have replaced "profoundly" with "strongly".

L28: Mass spectrometry – please be more specific.

Response: We have specified the type of mass spectrometry in the text.

L26-27: "…were measured by gas chromatography-mass spectrometry and high-resolution liquid chromatography-mass spectrometry."

L38: "regulating atmospheric oxidizing capacity and aerosol reactivity to mitigate biogenic SOA formation" Is biogenic SOA formation really the major source of air pollution? How can atmospheric oxidizing capacity and aerosol reactivity be regulated?

Response: Thanks for the reviewer's comment. The contribution of biogenic SOA to ambient particulate matter varies significantly across different locations and seasons. For example, observational studies revealed that biogenic SOA contributed to approximately half of total fine organic aerosol in southeastern US (Liao et al., 2007; Zhang et al., 2018). Modelling studies found that biogenic sources accounted for 61% of SOA on average in China, with its proportion reaching 75% in summer but dropping to 24% in winter (Hu et al., 2017). In addition, the proportion of biogenic SOA increased with anthropogenic emission reductions in recent years (Zhang et al., 2024). These results suggest an important contribution of biogenic SOA to PM pollution.

The atmospheric oxidizing capacity is largely governed by the concentrations of oxidants including hydroxyl radicals, ozone, and nitrate radicals, which can be regulated by reducing anthropogenic emissions such as VOCs and NOx (Fiore et al., 2024; Dai et al., 2024). The aerosol reactivity is controlled by its concentration and composition, which can be regulated by reducing anthropogenic emissions such as VOCs, NOx, and SO_2.

In the revised manuscript, this sentence has been revised as follows:

Line 36-39: "These findings highlight pathway-specific iSOA responses to emission reductions in a megacity and the importance of targeted anthropogenic emission reductions for mitigating biogenic SOA formation through regulating atmospheric oxidizing capacity and aerosol reactivity."

L46: Heald et al., 2008 -is there a newer reference.

Response: Thanks. The reference has been updated here as follows:

L44-46: "…the oxidation of isoprene contributes significantly to the formation of secondary organic aerosol (SOA) in the troposphere, estimated to 19.6 TgC yr^{-1} (Kelly et al., 2018)."

L58-59: "The gaseous IEPOX can be taken up into aqueous aerosol and undergo acid-catalyzed reactions to form polyols, organosulfates (OSs), and oligomers." This sentence needs references.

Response: Thanks. We have added references to this sentence.

L57-60: "The gaseous IEPOX can be taken up into aqueous aerosol and undergo acid-catalyzed reactions to form polyols, organosulfates (OSs), and oligomers (Hettiyadura et al., 2019; Lin et al., 2012; Surratt et al., 2010)."

L61: "abundantly measured" it is unclear what you mean – where they measured a lot or in high concentrations?

Response: We want to mean here the IEPOX-SOA tracers were measured a lot. To be more precise, we have replaced "abundantly" with "extensively".

L67: The sign before 45% is missing.

Response: Thanks. The sign before 45% is "~", and we have added it.

L100: Which iSOA tracers?

Response: The iSOA tracers here include polyols tracers such as 2-MTs and 2-MG. We have replaced the sentence here with a clearer statement.

Lines 98-102: "Furthermore, while the iSOA polyol tracers are formed in the particle phase, they can actively partition between gas and particle phases due to their semi-volatile characteristics (Fan et al., 2020; Isaacman-Vanwertz et al., 2016; Nguyen et al., 2015). As a result, considering their particle-phase concentration only may bias our understanding of the atmospheric abundance and chemistry of iSOA."

L108: cation ions -> cations

Response: We have revised this.

L109: The text is about recent changes but one of the references is from 2014.

Response: We have updated the reference to newer ones.

L104-107: "For example, $PM_{2.5}$ mass concentrations dropped significantly, with a marked reduction in water-soluble inorganic ions such as sulfate, ammonium, and cation ions (Liu et al., 2023a; Zheng et al., 2018; Liu et al., 2023b; Geng et al., 2024)…"

L113: later -> latter

Response: We have corrected this typo.

L119: comprehend -> understand

Response: We have revised this.

L149. What is KPA?

Response: KPA refers to ketopinic acid that has been widely used as a surrogate standard in the quantification of iSOA polyol tracers. We have replaced KPA with its full name ketopinic acid.

L151. Please provide information about sources of authentic standards.

Response: Thanks. We have added the sources of the standards.

L149-150: "…2-MG and 2-methylerythritol were quantified using their authentic standards (Toronto Research Chemicals, 99.8%)."

L171-172: Can this depend on the vendor of the ESI inlet and MS?

Response: Yes, the ESI efficiency is affected by various factors, including the type and configuration of the ESI inlet and MS as well as their specific operating conditions. Considering that the ESI efficiency obtained from one instrument may not be directly applied to another instrument, we have deleted the discussion in lines 171-172.

L181: Please be a bit more specific about the distance and the surrounding areas i.e. emissions.

Response: We have added a more detailed description about this.

L169-173: "Temperature, relative humidity (RH), as well as the concentrations of trace gases and $PM_{2.5}$ were measured at a state-controlled air quality monitoring station on the Xuhui Campus of Shanghai Normal University, which is surrounded by residential areas and commercial districts and 4.5 km southwest of the $PM_{2.5}$ sampling site of this work."

L194-195: Please discuss the uncertainty associated with this approach.

Response: Previous studies have found that lacking gas-phase inputs of ammonia could lead to underprediction of pH using thermodynamic equilibrium models, such as ISORROPIA and E-AIM (Hennigan et al., 2015; Guo et al., 2015; Song et al., 2018). Guo et al. (2015) found that the pH values were underestimated by 1 unit on average in southeast U.S. when using only aerosol ammonium data as inputs in ISORROPIA model. Similarly, Song et al. (2018) found that a 10-fold increase in gas-phase NH_3 concentrations roughly corresponds to a 1 unit increase in pH in the ammonia-rich atmosphere like Beijing. In addition, we found that the average difference between aerosol pH predicted by using aerosol ammonium only and gas-plus-particle-phase ammonia as inputs was about 1 unit in 2015 and 2019. Thus, we inferred that the lack of gas-phase concentrations of ammonia might lead to underestimation of pH by ~1 unit and increased the output pH estimated using aerosol ammonium only as input by one unit in 2021 to reduce the uncertainty arising from lack of gas-phase NH_3 in pH estimation.

We have added the following discussion to the revised manuscript.

L182-195: "Additionally, the 2021 pH was predicted by the ISORROPIA-II using particle-phase-only concentrations of ions as input due to lack of gas-phase NH_3 data. Previous studies have found that lacking gas-phase inputs of ammonia could lead to under-prediction of pH using thermodynamic equilibrium models, such as ISORROPIA and E-AIM (Hennigan et al., 2015a; Guo et al., 2015; Song et al., 2018). Guo et al. (2015) found that the pH values were underestimated by 1 unit on average in southeast US when using only aerosol ammonium data as inputs in ISORROPIA model. Similarly, Song et al. (2018) found that a 10-fold increase in gas-phase NH_3 concentrations roughly corresponds to a 1 unit increase in pH in the ammonia-rich atmosphere like Beijing. In addition, we found that aerosol pH in 2015 and 2019 predicted using aerosol ammonium only as input in our study was on average 1 unit lower than that predicted using gas-plus-particle-phase ammonia as input in Zhou et al. (2022). Thus, we inferred that the lack of gas-phase concentrations of ammonia might lead to underestimation of pH by ~1 unit in the present study and increased the output pH estimated using aerosol ammonium only as input by one unit to represent aerosol acidity in 2021."

L224: It is nice that the authors bring forward this uncertainty, but it is unclear how it affects the results presented here and whether it should have been tested if the magnitude of the effect is the same in the current study.

Response: Thanks for the reviewer's comment. Reviewer #1 has also raised this concern (comment #1) and we have addressed it in detail in our responses to his/her comments.

The matrix effect factors and relative ionization efficiencies reported by Bryant et al. (2021) may not be valid for the present study, given that these factors could vary significantly across studies due to the differences in ambient samples analyzed and in analytical instruments and specific instrumental conditions. Therefore, we have conducted a set of experiments to evaluate the matrix effect of iSOA OSs and the uncertainties in the abundance, trend, and relative ratios of different iSOA tracers in the present study and added relevant discussions to the revised manuscript (see below).

Lines 221-255: "Previous studies have demonstrated that the concentrations of 2-MT-OS and 2-MG-OS were significantly underestimated due to matrix effect by using reversed phase liquid chromatography-mass spectrometry (RPLC-MS) (Hettiyadura et al., 2015; Bryant et al., 2020; Bryant et al., 2021). In the present work, because of lack of authentic standards of isoprene-derived OSs, we are not able to quantify the absolute value of underestimation in the concentration of 2-MT-OS and 2-MG-OS due to the matrix effect. However, using ambient $PM_{2.5}$ samples with different concentrations, we can quantify the relative extent of underestimation in OS concentrations due to matrix effect in different samples, which allows for an evaluation of uncertainties in the abundance, trend, and relative ratios of different iSOA tracers in this study. In the matrix effect experiments, the extracts of ambient $PM_{2.5}$ samples with different concentrations were mixed and the measured signals of 2-MT-OS and 2-MG-OS in mixed extracts were compared to the sum of OS signals detected separately in individual extracts. The concentrations of $PM_{2.5}$, sulfate, as well as 2-MT-OS and 2-MG-OS in ambient samples used for this evaluation are listed in Table S3. The relative matrix effect factor (F_{matrix}), defined as the ratio of the measured OS signals in mixed extracts to the sum of OS signals measured in each extract before mixing, are used to evaluate the matrix effect of OSs. A F_{matrix} value of less than 1 indicates the presence of matrix effect.

As shown in Figure S2, the F_{matrix} values were significantly smaller than 1 in both summer and winter, indicating that the signal responses of 2-MT-OS and 2-MG-OS in mixed extracts were largely suppressed due to the matrix effect. Notably, F_{matrix} exhibits a significant negative dependence on the reduced mass (μ, μg m^{-3}), a proxy used to represent effective mass loadings of the mixed $PM_{2.5}$ extracts, defined as:

$$\mu = \sqrt{m_1 * m_2/(m_1 + m_2)} \tag{1}$$

where m_1 and m_2 are the $PM_{2.5}$ mass loading of individual samples. This observation suggests that the concentrations of iSOA OSs in $PM_{2.5}$ samples collected in 2015 were underestimated more than those in 2021, given that ambient $PM_{2.5}$ concentrations declined by 39.8% and 47.0% from 2015 to 2021 in summer and winter, respectively.

As $PM_{2.5}$ concentrations in ambient samples used for the matrix effect evaluation generally represent lower or upper ends of $PM_{2.5}$ concentrations during the observation period (see Table S3), the relative differences in measured F_{matrix} values at varying reduced mass (Figure S3) may roughly reflect the differences in the extent of underestimation in OS concentrations for samples collected across 2015-2021. During summer, the F_{matrix} value decreased from 0.71 to 0.63 for 2-MT-OS and from 0.85 to 0.58 for 2-MG-OS with increasing reduced mass, indicating that the concentrations of these two iSOA OSs were a factor of 1.2 and 1.5 more underestimated in 2015 than in 2021 due to matrix effect. Similarly, during winter the F_{matrix} values of iSOA OSs decreased from 0.9 to 0.6 with rising reduced mass, implying a factor of 1.5 greater underestimation in OS concentrations in 2015 than in 2021."

Lines 371-379: "However, the inter-annual trend of iSOA OSs could be altered due to the matrix effect. The measured concentration of 2-MT-OS exhibited a decreasing inter-annual trend, while 2-MG-OS showed insignificant variation between 2015-2021. Accounting for the significantly larger matrix effects in 2015 samples compared to 2021 samples (see Section 2.3), the true concentrations of 2-MT-OS would decrease more sharply and 2-MG-OS might also exhibit a declining trend."

Lines 411-413: "However, given the significant underestimation of iSOA OSs due to matrix effect and overestimation of C_5-alkene triols due to their potential artifact formation, the true concentration of iSOA OSs would predominate over that of polyol tracers."

Table S3. Major components in eight $PM_{2.5}$ filter samples that used for estimating matrix effect of 2-MT-OS and 2-MG-OS.

Season	Sample	Sampling date	$PM_{2.5}$ (μg m^{-3})	SO_4^{2-} (μg m^{-3})	2-MG-OS (ng m^{-3})	2-MT-OS (ng m^{-3})
Summer	High 1	2015/7/28	35.66	7.74	13.04	157.89
	High 2	2019/7/24	31.76	4.58	12.36	100.61
	Low 1	2019/8/5	10.04	2.29	0.58	0.83
	Low 2	2021/8/5	12.21	1.48	0.18	0.18
Winter	High 3	2022/1/2	77.76	6.28	1.80	0.47
	High 4	2022/1/3	65.38	5.41	1.54	0.41
	Low 3	2021/12/25	18.04	3.42	0.49	0.07
	Low 4	2021/12/26	23.71	3.01	0.70	0.08

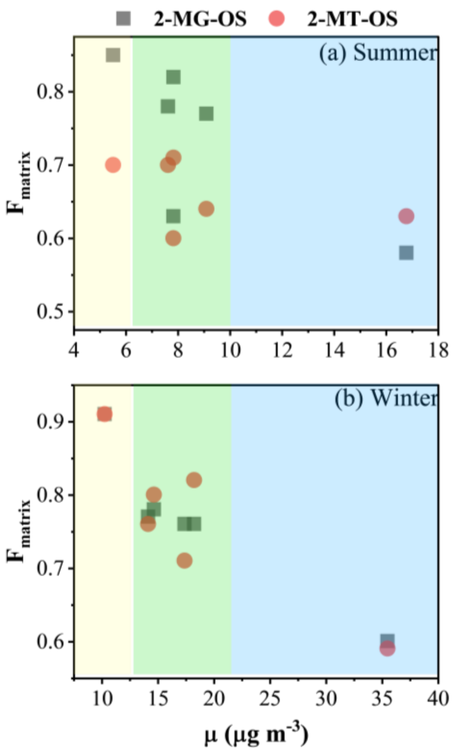


Figure S2. Relative matrix effect factors (defined as the ratio of signal response in mixed extracts to the sum of signal response in individual extracts, F_{matrix}) of 2-MT-OS and 2-MG-OS under different mixture type in summer (a) and winter (b) (yellow background represents low-plus-low mixture, green background represents high-plus-low mixture, and blue background represents high-plus-high mixture).

L299: Are the values statistically significantly different?

Response: Results from the t significance test indicated no statistically significant differences in ozone concentrations among the years 2015, 2019, and 2021, though the average values of MDA O_3 had a slight decrease.

L302: Dramatic is not a correct scientific word to use here. It would be nice if the authors could list some numbers.

Response: Thanks. The word "dramatic" was replaced by "drastic", and the proportion of major composition of $PM_{2.5}$ have been added there.

L331-333: "During the observation period, $PM_{2.5}$ concentrations decreased by 56.5%, with concentrations of major components, including sulfate, ammonium, and OM, decreasing by 51.8%, 40.6%, and 39.1%, respectively (see Figure 1)"

L303: Are there other types of nitrate than aerosol nitrate?

Response: There are no other types of nitrate than aerosol nitrate. For simplicity, we have replaced "aerosol nitrate" with "nitrate".

L305: Please also state the average values.

Response: Thanks, we have added the average values for the fraction.

L335-337: "Overall, OM was the most abundant component in $PM_{2.5}$, accounting for 10.2-72.7% (average 22.6%) of total $PM_{2.5}$ mass, followed by sulfate (6.8-45.2%, average 17.7%), nitrate (0.5-32.6%, average 17.0%), and ammonium (1.1-18.2%, average 8.8%)."

L308: Please avoid the use of the word dramatic. Aerosol pH – are the differences statistically significantly different?

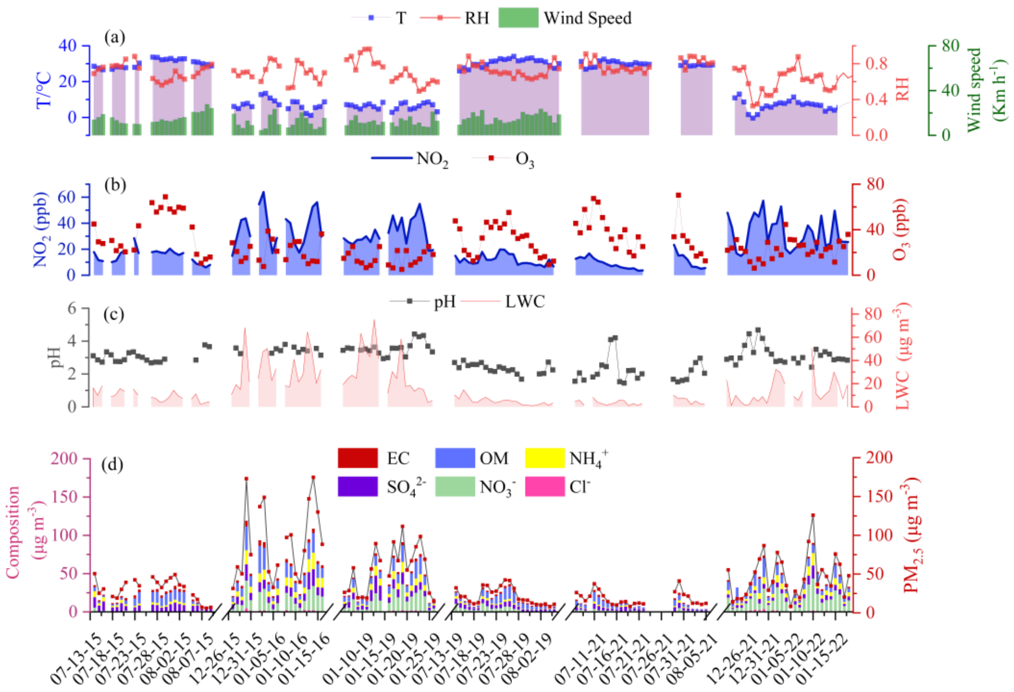
Response: Thanks. We have conducted a t-test for aerosol pH and LWC, and the result shows that the differences in aerosol pH and LWC across different years are significantly different ($p < 0.05$).

We have modified the sentence as follows:

L337-340: "Ascribed to the strong decrease of inorganic ion concentrations (in particular sulfate), aerosol LWC decreased from 9.14 ± 4.51 μg m^{-3} in 2015 to 4.40 ± 2.76 μg m^{-3} in 2021 ($p < 0.05$). Aerosol pH decreased from 3.2 ± 0.4 in 2015 to 2.5 ± 0.9 in 2021 ($p < 0.05$)…"

Figure 1: The figure is very small and it is difficult to see details.

Response: We have updated Figure 1 to a clearer version.



L334: Please state standard deviations.

Response: Thanks. We have included the standard deviations for the concentration values.

L408-409: "The annual average particulate concentration of the total iSOA polyol tracers (including 2-MTs, 2-MG, and C_5-alkene triols) were 36.1 ± 63.3, 33.4 ± 70.5, and 18.7 ± 47.1 ng m^{-3} in 2015, 2019 and 2021…"

L361: What do you mean by median?

Response: We have replaced "median" with "moderate".

L365-367: Please clarify this sentence.

Response: We have deleted this sentence since we have added detailed discussion about the modeled result of individual iSOA tracers in the revised manuscript.

L368: Is it consistent or different?

Response: Thanks. There is a typo here and we have deleted the word "in" to emphasize the consistence between the model simulation results in our study and previous studies.

L396-398: It seems like a bold statement to say that the concentrations in winter decrease when the values are 0.8 ± 0.3, 0.8 ± 0.2, and 0.7 ± 0.4.

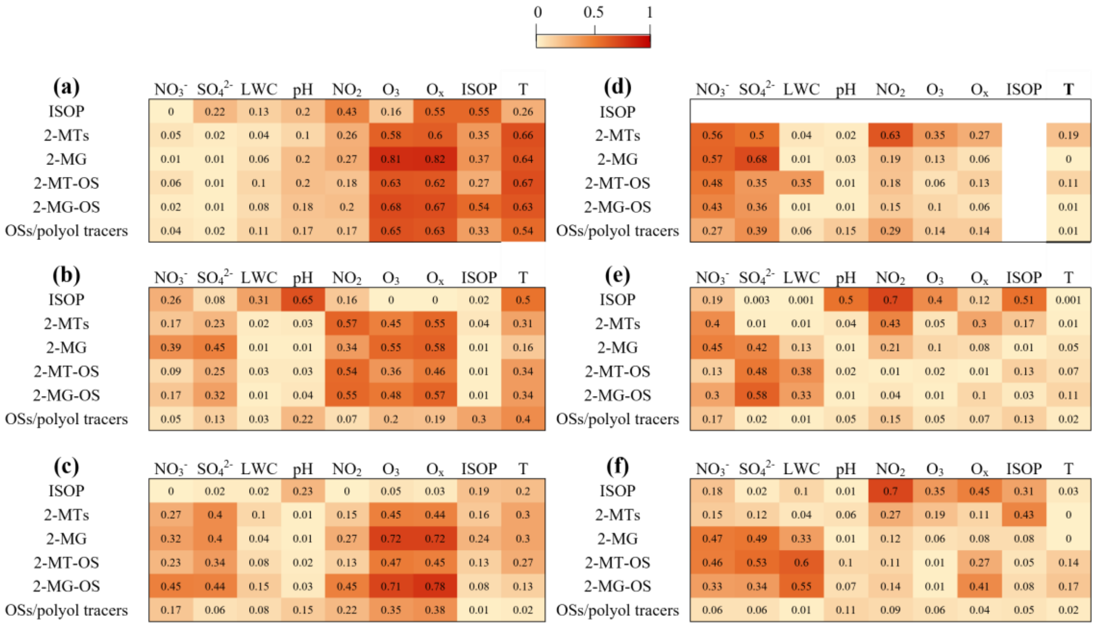
Response: We have deleted this statement since the large uncertainties arising from thermal degradation of C_5-alkene triols and the matrix effect of iSOA OSs would make it difficult to discuss the inter-annual trend of the IEPOX-SOA/HMML&MAE-SOA ratio.

L400-403: These sentences need clarification.

Response: Thanks. These sentences have been deleted since large uncertainties existing in quantification of C_5-alkene triols makes it difficult to discuss the trend of the ratios of IEPOX-SOA/HMML&MAE-SOA.

Figure 4: It is not possible to read the numbers in the heatmap.

Response: Thanks. The heatmap has been replaced with a clearer version.



(a)

	NO_3^-	SO_4^{2-}	LWC	pH	NO_2	O_3	O_x	ISOP	T
ISOP	0	0.22	0.13	0.2	0.43	0.16	0.55	0.55	0.26
2-MTs	0.05	0.02	0.04	0.1	0.26	0.58	0.6	0.35	0.66
2-MG	0.01	0.01	0.06	0.2	0.27	0.81	0.82	0.37	0.64
2-MT-OS	0.06	0.01	0.1	0.2	0.18	0.63	0.62	0.27	0.67
2-MG-OS	0.02	0.01	0.08	0.18	0.2	0.68	0.67	0.54	0.63
OSs/polyol tracers	0.04	0.02	0.11	0.17	0.17	0.65	0.63	0.33	0.54

(b)

	NO_3^-	SO_4^{2-}	LWC	pH	NO_2	O_3	O_x	ISOP	T
ISOP	0.26	0.08	0.31	0.65	0.16	0	0	0.02	0.5
2-MTs	0.17	0.23	0.02	0.03	0.57	0.45	0.55	0.04	0.31
2-MG	0.39	0.45	0.01	0.01	0.34	0.55	0.58	0.01	0.16
2-MT-OS	0.09	0.25	0.03	0.03	0.54	0.36	0.46	0.01	0.34
2-MG-OS	0.17	0.32	0.01	0.04	0.55	0.48	0.57	0.01	0.34
OSs/polyol tracers	0.05	0.13	0.03	0.22	0.07	0.2	0.19	0.3	0.4

(c)

	NO_3^-	SO_4^{2-}	LWC	pH	NO_2	O_3	O_x	ISOP	T
ISOP	0	0.02	0.02	0.23	0	0.05	0.03	0.19	0.2
2-MTs	0.27	0.4	0.1	0.01	0.15	0.45	0.44	0.16	0.3
2-MG	0.32	0.4	0.04	0.01	0.27	0.72	0.72	0.24	0.3
2-MT-OS	0.23	0.34	0.08	0.02	0.13	0.47	0.45	0.13	0.27
2-MG-OS	0.45	0.44	0.15	0.03	0.45	0.71	0.78	0.08	0.13
OSs/polyol tracers	0.17	0.06	0.08	0.15	0.22	0.35	0.38	0.01	0.02

(d)

	NO_3^-	SO_4^{2-}	LWC	pH	NO_2	O_3	O_x	ISOP	T
ISOP									
2-MTs	0.56	0.5	0.04	0.02	0.63	0.35	0.27		0.19
2-MG	0.57	0.68	0.01	0.03	0.19	0.13	0.06		0
2-MT-OS	0.48	0.35	0.35	0.01	0.18	0.06	0.13		0.11
2-MG-OS	0.43	0.36	0.01	0.01	0.15	0.1	0.06		0.01
OSs/polyol tracers	0.27	0.39	0.06	0.15	0.29	0.14	0.14		0.01

(e)

	NO_3^-	SO_4^{2-}	LWC	pH	NO_2	O_3	O_x	ISOP	T
ISOP	0.19	0.003	0.001	0.5	0.7	0.4	0.12	0.51	0.001
2-MTs	0.4	0.01	0.01	0.04	0.43	0.05	0.3	0.17	0.01
2-MG	0.45	0.42	0.13	0.01	0.21	0.1	0.08	0.01	0.05
2-MT-OS	0.13	0.48	0.38	0.02	0.01	0.02	0.01	0.13	0.07
2-MG-OS	0.3	0.58	0.33	0.01	0.04	0.01	0.1	0.03	0.11
OSs/polyol tracers	0.17	0.02	0.01	0.05	0.15	0.05	0.07	0.13	0.02

(f)

	NO_3^-	SO_4^{2-}	LWC	pH	NO_2	O_3	O_x	ISOP	T
ISOP	0.18	0.02	0.1	0.01	0.7	0.35	0.45	0.31	0.03
2-MTs	0.15	0.12	0.04	0.06	0.27	0.19	0.11	0.43	0
2-MG	0.47	0.49	0.33	0.01	0.12	0.06	0.08	0.08	0
2-MT-OS	0.46	0.53	0.6	0.1	0.11	0.01	0.27	0.05	0.14
2-MG-OS	0.33	0.34	0.55	0.07	0.14	0.01	0.41	0.08	0.17
OSs/polyol tracers	0.06	0.06	0.01	0.11	0.09	0.06	0.04	0.05	0.02

Figure 4. Coefficients of correlation (r^2) between iSOA compounds and various influencing factors of iSOA formation in (a-c) summer and (d-f) winter of 2015, 2019, and 2021, respectively. The ISOP is the abbreviation of isoprene.

L429-431: The correlation is quite low, which should be reflected better in the text.

Response: We have used a more neutral description for the correlation, replacing "moderate" with "weak to moderate".

L434-436: The correlation is quite low to moderate, which should be reflected better in the text.

Response: We have replaced "well" with "moderately".

L439: The change is quite small. Was a statistical test performed to check this?

Response: A t-test was performed to evaluate the statistical significance of the changes in wintertime IEPOX-SOA, LWC, and sulfate concentrations across different years. The result shows that the changes are statistically significant ($p < 0.05$) during the observation period.

L451: What happens to isoprene when the oxidation capacity is lower? Does it not just take longer for the oxidation to occur?

Response: Yes, the lower oxidation capacity generally means lower level of oxidants, thus the oxidation of isoprene would be slower, which could lead to decreased formation of iSOA.

L496-498: Unclear sentence.

Response: The sentence has been rephrased as follows:

L564-566: "It also suggests that the $k_{\text{i,j-IEPOX}}^{-4}$ is more appropriate for predicting aerosol heterogeneous reactivity toward IEPOX than other four sets of kinetic parameters."

Figure 6: Describe better what the numbers/years for the cases are.

Response: The x-axis label in Figure 6 has been revised and its meaning is explained in the figure caption. In addition, we have added the measured data to this figure according to Reviewer #1's suggestion.

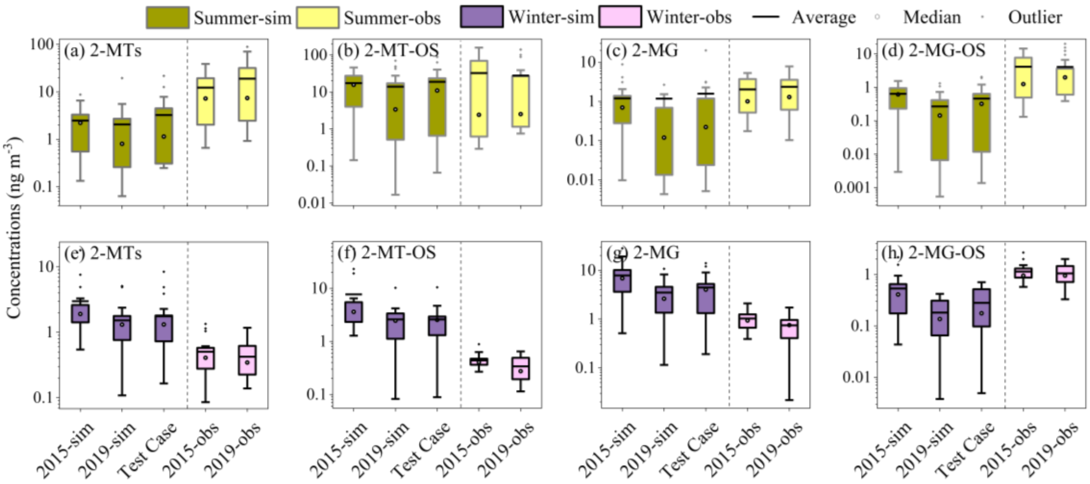


Figure 6. Simulated concentrations of 2-MTs, 2-MT-OS, 2-MG and 2-MG-OS in summer (a-d) and winter (e-h) in Case 1 (2015-sim and 2019-sim) and Test Case (simulations with 2015 emissions and 2019 meteorological conditions). The observed concentrations of 2-MTs, 2-MT-OS, 2-MG, and 2-MG-OS in 2015 and 2019 are also displayed (Detailed model-measurement comparisons are provided in Section 3.2; after accounting for matrix effect, 2-MT-OS would decrease more sharply and 2-MG-OS would show a descending inter-annual trend, consistent with model simulations).

L551: This line needs editing.

Response: Thanks. This sentence has been revised as follows:

L626-630: "On the other hand, MACR is known as a first-generation oxidation product of isoprene and an important intermediate for iSOA formation through NOx-dominant pathways (Nguyen et al., 2015), but it can also originate from primary sources, such as biological emissions (Jardine et al., 2012), residential wood burning (Gaeggeler et al., 2008), and vehicle exhaust emissions (He et al., 2009)"

L554: "Field studies have demonstrated" – this is a bold statement as there are differences between urban areas.

Response: This statement has been revised to improve clarity.

L630-632: "In certain urban areas, MACR arises primarily from vehicular emissions, as observed at the Heshan site in the Pearl River Delta region, China (Ling et al., 2019a) and in Houston, US (Park et al., 2011). "

L564: underappreciated is a loaded word not suitable here.

Response: We have replaced the word "underappreciated" with "underpredicted".

L567: three- year measurement gives the impression that samples for three complete years were studied. Please correct.

Response: We have replaced "a three-year measurement" with "an observation" and added "during the summer and winter in 2015, 2019, and 2021" to the end of this sentence to more accurately reflect the temporal coverage of the data.

L568: During the period 2015-2021, however only periods during three years were studied. Please correct.

Response: We have replaced "during the period 2015-2021" with "during the summer and winter in 2015, 2019, and 2021."

L574-575: "while HMML/MAE-SOA species (4.3, 4.2, and 4.3 ng m^{-3} in 2015, 2019, and 2021, respectively) did not decrease significantly". No they did not seem to decrease at all! Please correct.

Response: Thanks. We have deleted the word "significantly" in this sentence.

L579-581: Please check that this can be concluded based on the current study.

Response: Thanks. The correlation analysis showed that iSOA exhibited a good correlation with O_3 and Ox in summer and sulfate in winter. This indicates that the level of atmospheric oxidants (that largely determines the rate of isoprene oxidation) and sulfate aerosol (that plays a crucial role in the reactive uptake of isoprene-derived epoxide intermediates) are key factors driving iSOA formation in summer and winter, respectively. Based on this, we can conclude that "The atmospheric oxidation of isoprene to epoxide intermediates and their subsequent reactive uptake on aqueous aerosols are the key steps driving the formation of iSOA in summer and winter, respectively."

L582-583: This sentence is not clear.

Response: This sentence has been revised as follows:

L657-658: "The Ox-represented atmospheric oxidizing capacity and aerosol heterogeneous reactivity decreased significantly during the observation period…"

L599: Are biogenic emissions pollution?

Response: We have modified "PM pollution from biogenic emissions" as "PM formation from biogenic emissions".

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