Editor' comments

The reviewer raised an important remaining concern after the first round of revisions: "that the results (Figure 2-3) are biased towards Chinese cities", even though at least one study (referenced by the reviewer) outside of China exists that does consider reactive loss in determining emission ratios. Since this manuscript is a review article, I expect the authors to include all the relevant literature that exists on the topic globally. Therefore, the manuscript needs further revision. With the potential necessity to update figures, I rate the revision as "major".

Response: Thanks for your comments.

(1) This study primarily reviewed VOCs source apportionments considering reactive losses: including the methods of reducing reactive loss effects (*Section 3.1*), impacts of reactive losses (*Section 3.2*), estimation methods of reactive losses (*Sections 3.3 and 3.4*), and sources of VOC reactive losses (*Section 3.5*). There are a total of 170 relevant research papers (including one new paper) (on the line 109), of which 61 papers were identified as conducted in countries outside of China. These 61 papers mainly focus on methods of reducing the impacts of photochemical reactive losses in VOCs source apportionment research, especially the estimation methods of ambient VOC initial concentrations (i.e., the concentrations in the fresh emissions before any VOC species can undergo chemical reaction) and related parameters based on initial species emission ratios. Although these studies considered the impacts of reactive losses, they have almost not quantified reactive losses of ambient VOCs. As mentioned by reviewer in de Gouw's study (*https://doi.org/10.1002/2017JD027976*), although the impacts of reactive losses were considered when analyzing the sources of ambient OVOCs, they did not quantify the chemical reactive losses of ambient OVOC species. You can click on the link to check. Therefore, after reviewing all the relevant literature one by one, we presented the results of VOC reactive losses listed in the papers or the results that can calculate chemical losses (i.e., initial concentration minus observed concentration) in **Figs. 2** and **3** in the main text. Unfortunately, the results on the reactive losses of VOC species were mainly focused on research in Chinese cities (in addition to a newly published study in New York City, and a study in Mumbai; Borlaza-Lacoste et al., 2024, Kalbande et al., 2022), which was indeed the actual situation.

(2) According to the comments of the reviewer, we conducted a careful review and literature collection again using databases such as Science Direct (Elsevier), the Web of Science, Scopus, Springer, Wiley, and China National Knowledge Infrastructure (CNKI). We found that only two papers outside of China mentioned and analyzed the reactive losses of VOCs. One of the studies was conducted in Mumbai, India, but their research only included 9 VOC species and had substantial differences compared to VOC species involved in other cities in the world, making it difficult to compare (Kalbande et al., 2022). Therefore, we did not include the data from Mumbai in Figs. 2 and 3. In another paper in New York City (Borlaza-Lacoste et al., 2024), the reactive losses of ambient VOC species were estimated by the differences between the observed and initial concentrations. The related data of VOC reactive losses have also been added in Figs. 2 and 3 in the main text. Meanwhile, we have also added relevant

descriptive analyses in the text (on the lines 538-545). The more details are as follows. Additionally, we have added the chemical loss rate of VOCs in New York City in text for comparison with other cities (on the line 566). However, other studies outside of China have not quantified the reactive losses of the ambient VOC species. We have provided a detailed introduction and explanation of each relevant literature in our response to the reviewer below. You can click on the literature link to verify.

"To date, there were only 2 publications reporting quantitative research of VOC reactive losses conducted outside of China (i.e., Borlaza-Lacoste et al., 2024; Kalbande et al., 2022). The data of VOC reactive losses in the study conducted in New York City in USA (Borlaza-Lacoste et al., 2024) have been included in Figs. 2-3. However, due to the fact that only 9 VOC species were measured in the research conducted in Mumbai in India (Kalbande et al., 2022), the reactive loss data in their studies was difficult to compare and analyze with data from other cities, therefore, their estimated results were not shown in Figs. 2 and 3."

(3) In addition, we have rechecked the manuscript and made comprehensive revisions and improvements to the vocabulary, grammar errors, and other formatting issues.

References:

Kalbande, R., Yadav, R., Maji, S., Rathore, D. S., and Beig, G.: Characteristics of VOCs and their contribution to O_3 and SOA formation across seasons over a metropolitan region in India, Atmos. Pollut. Res., 13, 101515, https://doi.org/10.1016/j.apr.2022.101515, 2022.

Borlaza-Lacoste, L., Bari, M. A., Lu, C. -H., and Hopke, P. K.: Long-term contributions of VOC sources and their link to ozone pollution in Bronx, New York City, Environ. Int., 191, 108993, https://doi.org/10.1016/j.envint.2024.108993, 2024.

Reviewer #

The revised manuscript has addressed most of my comments. However, a concern remains that the results (Figure 2-3) are biased towards Chinese cities.

In the methods section, the authors have shown that 40% of studies were conducted outside of China. However, they argued that related studies on estimating photochemical reactive loss were all in Chinese cities.

Response: Thanks for your comments.

It needs to be noted that China has made a very substantial investment in measurements systems with much more data available that anywhere else. In addition, the rising ozone concentrations have made identifying VOC source a greater priority in China, thus leading to more publications. To specifically address your comment:

(1) First, we have thoroughly reviewed the literature related to this study and added one new study in New York City, USA (Borlaza-Lacoste et al., 2024). Currently,

a total of 170 papers have been identified in this study. We guaranteed that this study has covered all the articles currently published, and you can check and verify them in the database.

Through literature review, there are 61 papers conducted outside of China in this study, mainly involving studies on the VOC source apportionments during daytime and nighttime, photochemical-age estimation of the ambient VOC species, OVOCs source analyses, and methods for reducing chemical reactive losses using CMB model, etc. We found that only two papers outside of China mentioned and analyzed the reactive losses of ambient VOCs. One of the studies was conducted in Mumbai, India, but their research only included 9 VOC species (as shown in Table 1, Kalbande et al., 2022) and had substantial differences compared to VOC species involved in other cities in the world, making it difficult to compare. Therefore, to avoid misleading readers, we did not include the data from Mumbai in Figs. 2 and 3. In another paper in New York City (Borlaza-Lacoste et al., 2024), the reactive losses of VOC species were estimated by differences between the observed and initial concentrations. The related data of VOC reactive losses have also been added in Figs. 2 and 3 in the main text. Meanwhile, we have also added the relevant descriptive analyses in the main text (on the lines 538-545). The more details are as follows.

"To date, there were only 2 publications reporting quantitative research of VOC reactive losses conducted outside of China (i.e., Borlaza-Lacoste et al., 2024; Kalbande et al., 2022). The data of VOC reactive losses in the study conducted in New York City in USA (Borlaza-Lacoste et al., 2024) have been included in Figs. 2-3. However, due to the fact that only 9 VOC species were measured in the research conducted in Mumbai in India (Kalbande et al., 2022), the reactive loss data in their studies was difficult to compare and analyze with data from other cities, therefore, their estimated results were not shown in Figs. 2 and 3."

Table 1
Annual average concentration of VOCs (measured and PIC) along with their hydroxyl reactivity values at 298 K. .

VOC	Measured (ppbv)	PIC (ppbv)	kOH*10^{-12} at 298 K, cm^3. $molecule^{-1}.sec^{-1}$
Methanol	5.55	5.60	0.94
Acetonitrile	0.35	0.35	0.0263
Acetaldehyde	1.99	2.04	15
Acetone	2.18	2.18	0.17
Isoprene	0.59	1.48	100
2-butanone	0.56	0.57	1.22
Benzene	0.52	0.52	1.22
Toluene	1.07	1.13	5.63
C8 aromatics	0.76	0.86	15

Table 1 is cited from Kalbande, R., Yadav, R., Maji, S., Rathore, D. S., and Beig, G.: Characteristics of VOCs and their contribution to O_3 and SOA formation across seasons over a metropolitan region in India, Atmos. Pollut. Res., 13, 101515, https://doi.org/10.1016/j.apr.2022.101515, 2022.

(2) There is really nothing we can do to expand reporting studies that come from outside of China. All but the two we have added, the other reports did not quantify the chemical reactive losses of VOC species. We carefully classified and checked all the literature outside of China, and described the relevant contents of each paper in relation to this study. If you have any further suggestions or know of literature we have missed, please let us know. You can check for yourself the limitations of the other literature by clicking the relevant article link for verification. *The specific details are as follows:*

1) Buzcu-Guven and Fraser (2008), Buzcu and Fraser (2006), Kim et al. (2005), and Xie et al. (2006) conducted source apportionments using nighttime VOCs data in Harris County and Houston, USA, respectively. Zhao et al. (2004) and Buzcu and Fraser (2006) conducted source analyses of VOCs in Houston using both full day and nighttime data during the same period, and the results showed substantial differences between the two studies. Jain et al. (2022) conducted source analyses in Delhi, India, which showed that the factor profiles during the daytime and nighttime was consistent. The study by de Gouw et al. (2017) showed that VOCs can react with NO_3 radicals and O_3 at night, causing chemical losses, but did not do source apportionment with corrected data. The above papers were all related to the study of nighttime VOC source apportionments, corresponding to Section 3.1.1.1 in this study.

References:

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2) Roberts et al. (1984) and Parrish et al. (1992) introduced the method of calculating the photochemical-age using two species ratio method. Parrish et al. (2007) found that the photochemical age-based parameterization method has limitations in estimating the initial concentrations, but it is still an effective method. Bertman et al. (1995), Stroud et al. (2001), de Gouw et al. (2005), and Roberts et al. (2006) estimated the photochemical ages of ambient VOCs using a sequential reaction model. Kornilova et al. (2016), Rudolph and Czuba (2000), and Saito et al. (2009) used the isotopic hydrocarbon clock method to estimate the photochemical ages of VOCs. de Gouw et al. (2018) proposed a calculation method for nighttime O_3 exposure. Na et al. (2004) found that the aromatic hydrocarbons used to calculate Δt mainly come from solvent sources and petrochemical sources. The above papers were all related to the estimation method of photochemical ages of VOCs, corresponding to Section 3.1.1.2 in this study.

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Mintz and McWhinney (2008) analyzed ambient VOC sources in industrial areas in Canada using principal component analysis (PCA) method. They found that the concentrations of halogenated hydrocarbons could be affected by long-distance transportation, corresponding to Section 3.1 of this study. Kuhn et al. (2004) suggested that isoprene mainly originate from biogenic emissions. Wiedinmyer et al. (2001) investigated the concentrations of isoprene and isoprene reaction products. The above article corresponds to the Section 3.3 of this study. Brown et al. (2007) and Warneke et al. (2007) provided relevant data in Tables S1-S3 and S5-S6 of the Supplementary Materials. Yadav et al. (2016) analyzed the influencing factors of VOC chemical reactive losses, which corresponds to the Section 3.4 of this study.

References:

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analyses based on CMB. In addition, Meng et al. (1997) introduced the effects of ambient ozone and its precursors on particulate matter (PM) in the atmosphere. The above papers provided theoretical support for the introduction, Sections 3.1, 3.3, and 3.4 of this study. Meanwhile, these papers did not quantify the chemical reactive losses of ambient VOCs.

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First, I disagree with this statement. For instance, in Section 3.2 of de Gouw et al., 2018, the reactive losses of OVOC were considered when determining the OVOC emission ratios.

Response: Thanks for your comments. According to your comments, you are referring to the contents of Section 3.1.2 in the text. We have rechecked the studying contents of de Gouw et al. (2018). Prof. de Gouw introduced the species emission ratios of ambient OVOCs and the source apportionment method of OVOCs based on photochemical-age parameters. For formulas and principles of source apportionment method for OVOCs, this study has provided a detailed descriptions in Section 3.1.2 in the main text (on the lines 360-423). Although the source analyses method of OVOCs did consider the impacts of chemical losses, it has not been quantified (on the lines 407-410). If you still have a question, we have provided the link to this article below. Thanks!

Reference:

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2018.

Second, if this statement is accurate, the authors should discuss the potential bias in the results of these studies conducted outside China due to the absence of quantifying the reactive losses.

Response: Thanks for your advices.

We have added related analyses in the text on the relative lack of quantitative analysis of VOCs reactive losses outside of China (on the lines 546-550). Due to the lack of quantitative studies on ambient VOC reactive losses in the cities outside of China, the comparative analyses of the differences in reactive losses of VOCs in different cities and the impacts of losses on air secondary pollution from a global perspective in this study could be insufficient.

However, due to the fact that the study objectives in literature outside of China (all but the two we have added) were not to quantify chemical reactive losses of ambient VOC species. Therefore, although there was a lack of estimation of reactive losses, it did not necessarily mean that their research was biased. As described earlier, there were indeed shortcomings in analyzing the correlations between VOC reactive losses and secondary pollution formation from a global perspective. We have also added relevant descriptive analysis in the main text (on the lines 546-550).