A point-to-point response to referee #3

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Title: "Reconstructed VOC emissions reveal hidden ozone precursors: Overlooked roles of primary OVOCs and unmeasured species"

Author(s): Sijia Yin, Gan Yang, Chuang Li, Qingyan Fu, Juntao Huo, Yuruo Fu, Rengqi Yan, Lei Yao, and Lin Wang

We sincerely thank referee #3 for his/her careful reading and constructive comments, which have helped us substantially to clarify and strengthen the manuscript. We address each comment in turn below. Reviewer's comments appear in *red italics*, our responses in black, and the revised manuscript text in blue. All changes are highlighted in the revised manuscript. Line numbers refer to those in the revised manuscript.

1. *This paper presented the comprehensive analysis of measured VOC concentrations and missing OH reactivity at a suburban site in Shanghai, China. Source apportionment was performed via the photochemical age-based parametrisation method allowing the attribution of OVOCs to primary and secondary anthropogenic sources, biogenic sources, and regional background. Eventually, the ozone formation potential was determined with the MIR method. To assess the impact of chemical aging during transport of the eventually measured air mass on the VOC distribution and the ozone formation potential, primary VOC and OVOC concentrations were estimated using the observed VOC concentrations and the OH exposure, derived from a species ratio method.*

This paper focuses on the comparison between observed VOCs and estimated primarily emitted VOCs and the implication on the OFP. Even though differences are somehow expected, the quantitative comparison gives an idea about the magnitude and the VOCs, either measured or reconstructed, contributing most to the OFP.

Overall, this paper highlights the importance of reconstructing VOC and OVOC emissions in the OFP analysis, which is commonly performed with observed VOC concentrations. Even though previous studies already investigated the impact of the reconstruction on the OFP, I believe that this study would be complementing if the comparison between the different studies is properly discussed. Therefore, I recommend this paper for publication after major changes have been done.

Response: We sincerely appreciate referee #3's positive assessment and his/her recommendation toward publication. We fully agree that a systematic comparison with previous studies strengthens the contribution of this work. To address this, we have:

(1) added a description of trace gas concentrations, meteorological parameters, and the total OH reactivity measured during the campaign (Sect. 2.1);

(2) included a quantitative comparison with the most relevant prior study (Zheng and Xie, 2025);

(3) expanded the discussion on the discrepancies between observed and reconstructed OFP, including a quantitative assessment of the OFP that would be missed if only PAMS species were monitored.

All revisions are detailed in the point-by-point responses below.

2. Major comments

2.1 *In general, I am missing more details about trace gas concentrations and meteorological parameters (temperature, relative humidity) measured during the campaign which would help the reader to understand the chemical conditions onsite. There is no information about the level of ozone and nitrogen oxides as well of the measured total OH reactivity. The measured total OH reactivity is not discussed at all in the paper. Even though the missing OH reactivity is obviously derived from the comparison of the total OH reactivity with the OH reactivity calculated from the VOC measurements, there is no comparison shown and no absolute numbers given. Furthermore, since the OH exposure determines the reconstruction, it is important to show this as well. How long is actually the estimated transport time of the air mass which plays into the OH exposure and thus into the reconstruction?*

Response: We thank referee #3 for this important suggestion. The chemical and meteorological context of the campaign is indeed essential for interpreting both the source-apportionment results and the reconstruction. A full characterization of the meteorology and total OH reactivity at the DSL site during this campaign was reported by Yang et al. (2022). Accordingly, we have made the following additions, which reads: (Lines 91-92) "In addition to VOC measurements, O_3, NO_x, temperature, and relative humidity were continuously monitored during the campaign."

and (Lines 202-214): "During the daytime (07:00–18:00 local time), the mean temperature and relative humidity were 28.7 °C and 75.8%, respectively. Ambient O_3 concentrations ranged from 24.0 to 324.0 µg m^{-3} (mean ± one standard deviation: 127.1 ± 58.4 µg m^{-3}), and NO_x averaged 14.0 ± 6.3 ppbv, indicating conditions favorable for photochemical oxidation of VOCs. Consistent with this chemical environment, the daytime measured total R_{OH} averaged 39.9 ± 20.9 s^{-1} (Yang et al., 2022), comparable to values reported at other suburban Chinese sites with substantial biogenic and photochemical influence. For example, Yang et al. (2017) reported total R_{OH} of ~30–40 s^{-1} at the suburban Heshan site in the Pearl River Delta during summer. During our campaign, the OH exposure of anthropogenic VOCs, derived from the ethylbenzene/m&p-xylene ratio method (Roberts et al., 1984), was estimated to be 3.2 ± 3.2 × 10^{10} molecules cm^{-3} s, corresponding to a photochemical age of approximately 4.4 h by assuming a mean daytime OH concentration of ~2.0 × 10^6 molecules cm^{-3}. For biogenic VOCs, the OH exposure estimated using a sequential reaction model (Stroud et al., 2001) was 7.9 ± 5.1 × 10^9 molecules cm^{-3} s, equivalent to a photochemical age of 1.1 h. Overall, the air masses sampled at DSL site on average underwent 1–4 h of integrated photochemical processing prior to arrival; therefore, the observed OVOCs are expected to reflect contributions from both primary emissions and substantial secondary production.”

2.2 *The quantitative comparison of the results with previous studies (e.g. Zheng and Xie (2025)) should be included in the paper. How do the results about the OFP and the top drivers compare? How does the measured total OH reactivity compare with other studies focusing on ozone pollution in China?*

Response: We thank referee #3 for this constructive suggestion. We agree that situating our results in the context of previous studies strengthens the discussion. Two dedicated comparisons have been added:

(1) In Sect. 3.3, a paragraph that quantitatively compares our reconstructed-vs-observed OFP biases and our top OFP drivers with those of Zheng and Xie (2025), which reads (Lines 386-400): “Our results are qualitatively consistent with, yet quantitatively and conceptually distinct from, those of Zheng and Xie (2025), who compared reconstructed primary emitted versus ambient VOC concentrations at three sites in the Sichuan Basin. Both studies found that NMHCs, particularly alkenes and aromatics, were underestimated in OFP estimation when using observed rather than primary emitted concentrations, while OVOCs’ OFP were overestimated. Specifically, Zheng and Xie (2025) found that OVOCs’ OFP in Chengdu decreased from 171.9 to 88.4 μg m^{-3} when emitted rather than ambient concentrations were used (~95% overestimation), consistent with the 42.6% overestimation identified here; conversely, the OFP of reactive NMHCs increased, mirroring the 31.7% underestimation for NMHCs at the DSL site. For individual species, isoprene, acetaldehyde, propylene, m&p-xylene, ethylene, toluene, and o-xylene were consistently identified as key O_3 precursors, reinforcing their priority status in O_3 control strategies. Notably, a key distinction is that our study covers a far broader range of VOC species, encompassing 321 measured VOC species

(113 NMHCs and 208 OVOCs), together with R_{OH} inferred unmeasured species, compared to the 99 species (86 NMHCs and 13 OVOCs) analyzed by Zheng and Xie (2025). This broader coverage enables us to attribute 33.2% and 14.5% of initial TOFP to primary emitted OVOCs and unmeasured species. The two studies are thus complementary rather than hierarchical: Zheng and Xie (2025) additionally resolved nighttime NO_3/O_3-driven alkene loss, whereas our reconstruction focuses on the daytime OH-dominated window to characterize a broader range of oxygenated and unmeasured species."

and (2) In Sect. 3.1, a short comparison that places our measured total R_{OH} in the context of values reported at other representative Chinese sites, which reads (Lines 204-207): "Consistent with this chemical environment, the daytime measured total R_{OH} averaged 39.9 ± 20.9 s^{-1} (Yang et al., 2022), comparable to values reported at other suburban Chinese sites with substantial biogenic and photochemical influence. For example, Yang et al. (2017) reported total R_{OH} of ~30–40 s^{-1} at the suburban Heshan site in the Pearl River Delta during summer."

2.3 *At the end, it was mentioned that the results help understanding previous discrepancies between field observations, emission inventories, and source measurements. How much do the findings solve these discrepancies? In addition, the authors state that the emission control strategies focus too much on a limited set of NMHCs such as PAMS. How much do PAMS contribute to the total OFP? In other words, how much is the OFP underestimated by only considering PAMS?*

Response: We thank referee #3 for this constructive comment, which directly targets the practical implications of our findings. We have addressed both questions quantitatively by adding two paragraphs in Sect. 4 (Atmospheric implications):

(1) We discuss the extent to which our reconstructed emissions reconcile the apparent observation–inventory–source-profile discrepancies, which reads (Lines 411-420): "These reconstructed emissions help to reconcile previously reported discrepancies between ambient VOC observations and emission inventories. Previous inventories and direct source measurements in urban and suburban environments indicate that anthropogenic sources contribute roughly 70% of the total VOC burden, with OVOCs comprising approximately 20–65% of total VOC emissions (Gu et al., 2021; Ma et al., 2022a; Ou et al., 2015; Yan et al., 2024). Our reconstructed initial concentrations closely reproduce this source-level composition (OVOCs: 40.8%; anthropogenic origin: 71.5%). By contrast, ambient observations show a markedly reduced anthropogenic primary fraction (48.9%), alongside a substantially elevated OVOC proportion (52.4%), reflecting photochemical aging and secondary OVOC formation

during atmospheric transport, which obscure the original VOC emission profile."

and (2) We present how much of the initial TOFP is captured by the 56 PAMS species and, by difference, how much would be missed under a PAMS-only monitoring strategy, which reads (Lines 427-434): "Current emission control strategies remain narrowly focused on a limited set of NMHCs (e.g., PAMS species) (Gao et al., 2025). However, our results show that the 56 PAMS compounds account for only ~40% (173.0 μg m^{-3}) of the initial TOFP, implying that exclusive reliance on PAMS monitoring would leave ~60% of the total initial OFP unaddressed. The unaccounted fraction is composed of primary OVOCs (33.2%), reactivity-inferred unmeasured VOCs (14.5%), and non-PAMS NMHCs detected by Vocus-PTR (12.2%). These findings underscore the necessity of integrating these species into emission inventories, routine monitoring networks, and regional chemical transport models to improve the accuracy of O_3 predictions."

3. *Technical corrections*

3.1 *In general, values have too many decimals which is not necessary.*

Response: We have reviewed all numerical values throughout the manuscript and reduced concentrations, percentages, and OFP metrics to one decimal place. For example, "431.96 ± 319.19 μg m^{-3}" has been revised to "432.0 ± 319.2 μg m^{-3}", and "6.84 ± 4.41 s^{-1}" to "6.8 ± 4.4 s^{-1}", etc.

3.2 *There are often articles missing. Here are few examples: (a) Line 14,19,21:**the** ozone formation potential, (b) line 17: **an** OFP*

Response: Corrected throughout the manuscript, which reads

(Line 14): "However, conventional estimates of the ozone formation potential (OFP)…";

(Line 19): "the OFP of unmeasured VOCs were inferred by";

(Line 21): "leading to pronounced discrepancies in the OFP estimation…";

(Line 27): "…OVOCs and unmeasured species exhibited an OFP comparable to NMHCs";

and those identified by the authors.

3.3 *There are references missing:*

Line 86: Fuchs et al. (2017) (https://doi.org/10.5194/amt-10-4023-2017) should be cited here as it compares different kOH measurements including CRM.

Line 92: Give a reference for PAPM

Response: Both references have been added, which reads (Line 94): "R_{OH} was measured using a comparative reactivity method (CRM) (Sinha et al., 2008; Fuchs et al., 2017)”, and (Line 101): “The PAPM (De Gouw et al., 2005, 2018) assumes that…”

3.4 *Since different techniques are applied, it is important to make sure to be precise in the wording. There are different situations where it is unclear for the reader what is meant.*

3.4.1 *Line 93: It sounds like the PAPM neglects the photolysis of OVOCs. However, in line 52 it is mentioned that the photolysis of OVOCs is also considered by the PAPM.*

Response: We have revised the sentence at line 93 to ensure the consistency with the description in the introduction, which reads (Lines 102-103): “···and that reactions with OH radicals and photolysis dominate the photochemical removal of OVOCs”

3.4.2 *Line 94: I would add the corresponding emission rates, like “anthropogenic primary emissions (ER_OVOC), …” This will help the reader to understand what is shown in the plots later.*

Response: The emission-ratio notation has been introduced explicitly, which reads (Line 103-105): “ambient OVOC concentrations can be attributed to anthropogenic primary emissions (characterized by emission ratio ER_{OVOC}), anthropogenic secondary formation (characterized by emission ratio $ER_{precursor}$), biogenic sources (characterized by emission ratio $ER_{biogenic}$), and regional background,”

3.4.3 *Lines 106,107: It is not clear to me how these two sentences connect. Currently, it reads like a least-squares fit was done because the OH radicals are dominating the VOC oxidation. However, I guess the authors intended to say that only this specific time window is considered because OH radicals dominate the VOC oxidation then.*

Response: Thank you for pointing out this ambiguity. We have revised the text to make the logic explicit, which read (Lines 117–119): “Since OH radical reactions dominate VOC consumption during the daytime, only observations from 07:00 to 18:00 local time were used for the PAPM fitting, ensuring that the underlying assumption of OH-driven oxidation (Eq. 1) is satisfied.”

3.4.4 *Line 107: I would add the fit parameters here to make it as clear as possible.*

Response: Added, which reads (Lines 119-121): “The source contributions of OVOCs were then determined by a least-squares fit, with the fitted emission ratios and background levels summarized in Tables S3 and S4.”

3.4.5 *Lines 113-115: How are the sources of NMHCs derived?*

Response: A clarifying sentence has been added to indicate that NMHCs are treated as directly emitted (no secondary formation), and that their source attribution (anthropogenic versus biogenic) is based on compound identity, which reads (Lines 131–133): "Unlike OVOCs, NMHCs are treated as directly emitted compounds with no secondary formation pathway. Among them, isoprene, monoterpenes ($C_{10}H_{16}$), and sesquiterpenes ($C_{15}H_{24}$) are classified as biogenic VOCs, whereas other measured NMHCs are categorized as anthropogenic VOCs."

3.4.6 *Line 139,140: What is used for the correlation? The different emission rates?*

Response: We have clarified that the background fraction of missing R_{OH} was compared against the three tracers used in the MLR (i.e., C_2H_2, $[\text{isoprene}]_{\text{source}}$, and O$x$), which reads (Lines 157–158): "The background fraction of missing R_{OH} exhibited no correlation with $[C_2H_2]$, $[\text{isoprene}]_{\text{source}}$, or $[\mathrm{O}x]$, hinting that it could not be attributed to the three source categories represented by these tracers."

3.4.7 *Line 147,148: Why is it here the "jth source at the observation site" and before only "jth source"? I recommend to be consistent, in this specific case with regard to lines 113-115, 120,149.*

Response: We have unified the text to consistently use "the j^{th} source", which reads (Lines 166–168): "where $[\text{unmeasured VOCs}_{i,j}]_{\text{t}}$ denotes the ambient concentrations of unmeasured VOC_i originating from the j^{th} source, $[\text{VOCs}_{i,j}]_{\text{t}}$ represents the concentrations of measured VOCs from the j^{th} source, respectively."

3.4.8 *Line 150: It is not clear to me what you mean by "equivalent concentrations of measured VOCs". You do measure concentrations, so this wording is confusing.*

Response: We have revised this sentence to remove the ambiguity, which reads (line 169): "Based on the equivalent concentrations derived from Eq. (6),"

3.4.9 *Line 152: How were the unmeasured VOCs associated with secondary formation determined?*

Response: A clarifying sentence has been added, which reads (line 171-172): "Note that unmeasured VOCs attributed to secondary formation were excluded from this calculation, as secondary OVOCs are produced during atmospheric transport rather than emitted at the source."

3.4.10 *Line 155: Isn't Eq. (7) the same as Eq. (2)? In line 151 it is mentioned that the unmeasured VOCs are similarly treated as the measured ones.*

Response: The two equations have an identical mathematical form but apply to different categories of species. Eq. (2) uses directly measured concentrations, whereas Eq. (7) uses concentrations inferred from missing R_{OH} for the unmeasured species. We have made this distinction explicit, which reads (Line 176-177): "Eq. (7) follows the same formulation as Eq. (2), but is specifically applied to unmeasured VOCs using their inferred concentrations rather than directly measured values."

3.4.11 *Line 177: "fitted results". For the understanding, I would add here a reference to the corresponding equation and section. Same for the caption of Figure 1.*

Response: References to the corresponding equation and section have been added in both the main text and the figure caption, which reads

(Line 217-220): "The fitted results from Eq. (1), with the fitted parameters summarized in Tables S3 and S4, show good agreement with the observed concentrations (R = 0.40–0.90) and reconstruct the time series well, as illustrated for formaldehyde, acetaldehyde, and acetone in Fig. 1.",

and (Caption of Fig. 1, lines 222-223) "Time series (a, c, e) and scatter plot (b, d, f) of the PAPM fitting result from Eq. (1) (using the emission ratios and background levels in Tables S3 and S4) versus the measured concentrations of formaldehyde, acetaldehyde, and acetone."

3.5 *Line 22,24: Since this is the abstract it is not clear yet what the authors mean by "reconstructed NMHCs/OVOCs". To clarify this, I would rather write "reconstructed primarily emitted NMHCs/OVOCs"*

Response: Revised throughout the abstract, which reads (Lines 22–24): "the OFP contributions from reconstructed primary emitted NMHCs (52.3%) were underestimated by 31.7% when derived from observed concentrations for this site, whereas those from reconstructed primary emitted OVOCs (33.2%) were overestimated by 42.6%."

3.6 *Line 35: I do not think that it is relevant here whether the VOCs were added in smog chambers or came from numerical simulations. I would cancel this as it confuses the reader.*

Response: The phrase "added in smog chambers or numerical simulations" has been removed, which reads (line 37-40): "By definition, the MIR coefficient quantifies the mass of O_3 formed per unit mass of a freshly emitted VOC (i.e., before any photochemical processing) (Carter, 1994, 2010); a physically consistent OFP must therefore be evaluated from the initial emission concentrations rather than from ambient observations.”

3.7 *Line 87: “…, respectively” can be cancelled as it does not apply.*

Response: Removed.

3.8 *Line 89: What is “zero gas”? Is it pure synthetic air or is it compressed air? I would be more specific here.*

Response: The reagent has been specified, which reads (Line 97): “…first in the presence of pure nitrogen gas and then in the presence of ambient air.”

3.9 *There are some sentences with a wrong syntax or grammer:*

3.9.1 *Line 99: “their precursors to relative C2H2” should be “their precursors relative to C2H2”*

Response: Corrected.

3.9.2 *Line 103: “ratio of OVOCs to isoprene from biogenic emissions” should be “ratio of OVOCs from biogenic emissions relative to isoprene”*

Response: Corrected.

3.9.3 *Line 193: “high contributions to MVK” should be “high contributions of MVK”*

Response: Corrected.

3.10 *Line 110: I believe the nitrate radical has not been introduced yet.*

Response: The full name has been added at first mention, which reads (Line 124): “…including deposition and reactions with the nitrate radical (NO_3) or O_3, are regarded to be negligible.”

3.11 *Line 129: I would add the scientific meaning of a, b, and c.*

Response: A description has been added, which reads (Line 146-148): “a, b, and c are regression coefficients representing the sensitivities of missing R_{OH} to the tracers of anthropogenic, biogenic and secondary sources, with fitted values of 0.39, 2.48, and 0.04 s^{-1} ppb^{-1}, respectively.”

3.12 *Line 130: “Cbackground is interception” seems to be lost.*

Response: This sentence has been rewritten for clarity, which reads (Lines 148-149): "The intercept, $C_{background}$ (2.27 s^{-1}), represents the baseline missing R_{OH} that is independent of these three source categories."

3.13 *Line 131: The unit of a,b,c should be ppb/s*

Response: We respectfully clarify that the units of the fitted coefficients (a, b, and c) are s^{-1} ppb^{-1}. In Eq. (5), missing R_{OH} has units of s^{-1}, whereas [C_2H_2], $[isoprene]_{source}$, and [Ox] are expressed in ppb. The coefficients must therefore have units of s^{-1} ppb^{-1} so that each term (e.g., a × [C_2H_2]) yields a correct unit of s^{-1}.

3.14 *Line 132,133: "derive" seems to be the wrong verb here. I would rather use "originate".*

Response: Corrected.

3.15 *Line 165: Why were these 130 OVOCs without MIR values excluded from the OFP analysis and not treated as unmeasured OVOCs?*

Response: Thank you for raising this point. These 130 OVOCs were excluded because reliable MIR values, required for OFP calculation, are not available. Treating them as "unmeasured VOCs" would require additional assumptions (e.g., assigning surrogate MIR values from chemically similar species) that would introduce unquantifiable uncertainties into the OFP estimation. To ensure the robustness of the OFP estimation, we therefore chose to exclude these compounds from the quantitative OFP calculation. We have clarified this rationale in the revised manuscript, which reads (Lines 188-190): "Including them without MIR values would require additional assumptions that would introduce unquantifiable uncertainties; therefore, to ensure the robustness of the results, these species were excluded from both the OFP calculations and the estimation of unmeasured VOCs."

3.16 *Line 171: "kOH" was not introduced yet. Since kOH is typically an abbreviation for the OH reactivity, which was also measured, I would not give this abbreviation to an OH reaction rate constant. In general, there is not a consistent notation of reaction rate constants and VOC species. In Eqs. (1), (2), (7), the OH reaction rate of a VOC species I is called ki. In Eq. (4), the VOC species is called Xi and the corresponding OH rate coefficient k(Xi+OH)*

Response: We have unified the notation throughout the manuscript. The abbreviation "k_{OH}" has been removed to avoid confusion with the total OH reactivity (R_{OH}). The OH radical reaction rate constant for species i is now consistently written as k_i, and Eq. (4) has been updated accordingly so that it is consistent with Eqs. (1), (2), and (7).

3.17 *Line 297, 298: I do not understand where these numbers are coming from. It is not consistent with Fig. 3.*

Response: We apologize for the lack of transparency in the original wording. The numbers are in fact consistent with Fig. 3, if the percentages are converted to absolute OFP values. Specifically:

- Observed OVOCs' OFP (Fig. 3a): 42.9% × 476.9 μg m^{-3} ≈ 204.6 μg m^{-3}.
- Reconstructed primary emitted OVOCs' OFP (Fig. 3b): 33.2% × 432.0 μg m^{-3} ≈ 143.4 μg m^{-3}.
- Overestimation: (204.6 − 143.4) / 143.4 ≈ 42.6%.

The corresponding calculation for unspecified OVOCs yields an overestimation of more than 50%. To allow these calculations to be verified directly from the figure, we have added the absolute OFP values to the text.

Revised (lines 342–345): "the OFP of OVOCs, as inferred from Fig. 3, was overestimated by 42.6% (observed OVOCs: 204.6 μg m^{-3}, i.e., 42.9% × 476.9 μg m^{-3}; reconstructed primary emitted OVOCs: 143.4 μg m^{-3}, i.e., 33.2% × 432.0 μg m^{-3};), which is a bias largely attributable to secondary production."

3.18 *Line 322: I would make clear that it refers here to the reconstructed VOCs and the corresponding OFP.*

Response: Clarified, which reads (Line 369-370): "The top-ten reconstructed primary emitted VOCs collectively accounted for 55.3% of initial TOFP, despite representing only 31.2% of the initial TVOCs,"

3.19 *Line 327 (Figure 4): It should be added that it is the contribution to the TOFP. Are only anthropogenic primary emissions shown? This is unclear.*

Response: The caption of Fig. 4 has been rewritten to make these points explicit. Both anthropogenic and biogenic contributions are now shown, which reads (Caption of Fig. 4, Lines 375-376): "Reconstructed initial concentrations and contributions to the initial TOFP of the top-ten contributors during the daytime. Bar colors indicate source attribution (blue: anthropogenic sources; green: biogenic sources)."

3.20 *Line 328: The OFP should be added.*

Response: Added.

3.21 *Line 350: The number (14.5%) should be checked. According to Fig. 2b it should rather be 12.6%.*

Response: Both numbers are correct but refer to different quantities: 12.6% is the contribution of unmeasured VOCs to the initial TVOC concentrations (Fig. 2b), whereas 14.5% is their contribution to the initial TOFP (Fig. 3b). We have rewritten the sentence to remove the ambiguity, which reads (Lines 420-423): "Moreover, the substantial contribution of unmeasured VOCs (14.5% of the initial TOFP, despite accounting for only 12.6% of the initial TVOC concentrations) underscores the limitations of current monitoring networks, particularly for biogenic and oxygenated species; expanding measurement capabilities is therefore critical for capturing the true O_3 formation capacity of the atmosphere."

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