Response to Reviewer 1

We thank the reviewers for the detailed comments and suggestions, also the time and effort that they invested into the review. We believe the revisions have strengthened the manuscript. Below we provide a detailed point-by-point response to all the comments. The reviewers' comments are shown in blue, and our responses follow in black. The changes in the manuscript are highlighted in red and underlined. Page and line numbers are indicated in the revised track-changed version.

General comments

Chi et al. (2025) used the high temporal and vertical resolution of ground-based remote sensing observations to study the hourly evolution of HCHO and NO2 columns (and near-surface concentrations) over several cities in Japan. They further classified the chemical conditions (e.g., NOx-sensitive, VOC-sensitive, and transitional) based on thresholds derived from a chemical transport model, which was vetted against in situ observations. The novelty of this work lies in its application of Pandora observations to ozone-sensitivity diagnosis. However, the paper overlooks several critical components that are necessary for reasonably attributing ozone sensitivities. Moreover, this reviewer is not entirely convinced that the model used to determine the thresholds has reasonable accuracy, or that some of its significant discrepancies with observations could result in a wide range of ambiguous possible outcomes. Therefore, I do not recommend the publication of this paper in the present form.

Response:

Thank you very much for the reviewer's comments and suggestions, which have helped us improve the manuscript. The authors have revised the manuscript accordingly and clarified the limitations of the current study. The HCHO-to-NO_2 ratio (FNR) has certain limitations in representing ozone (O_3) production; however, it remains a commonly used indicator because it can be readily obtained compared with more precise indicators that generally require extensive modeling efforts. Importantly, FNR can be derived with wide spatial coverage from both satellite and ground-based remote sensing networks, allowing quick assessment of the O_3 production on global or regional scale.

We would like to briefly explain the model framework used to determine the FNR thresholds. Our approach follows the method of Jin et al. (2017). Instead of using the LRO_x/LNO_x ratio of 0.35 derived from model simulations to identify the transition/ambitious range of FNR (Schroeder et al., 2017), the current method directly identifies the transition/ambiguous range through the response of O_3 to emission perturbations. Conceptually, the two methods are theoretically similar. We have also provided uncertainty estimates for the transition/ambiguous range obtained from the GEOS-Chem model in Section 3.3, in the revised version.

For better application of Pandora observations to O_3-sensitivity diagnosis, we have revised the manuscript to focus on O_3 exceedance events, where photochemical processes are generally strong. Although O_3 exceedances can be attributed to regional and vertical transport, sensitivity diagnosis is useful to reduce O_3 concentrations from local photochemical

production. Additionally, only Pandora observations obtained around local noon were considered to maximize the data accuracy for the analysis.

Major comments:

Light, light, light, A central but often under-emphasized point in tropospheric ozone research is the dominant role of photochemistry. Virtually all ozone-related studies begin by noting that ozone formation is a multifaceted problem driven by interactions among its precursors (primarily NOX and non-methane VOCs) under sunlight. Yet over the past two decades, many studies have disproportionately emphasized the roles of NOX and VOC while devoting relatively little attention to the availability of light (photolysis rates) itself, which is the fundamental driver of the chemical processes governing both ozone production and loss.

Beyond the well-known limitations that HCHO and NO2 do not fully represent VOCR and reactive nitrogen, it is crucial to recognize that both the magnitude and sensitivity of ozone production rates depend strongly on geophysical factors that are independent of FNR. Among these factors, photolysis rates and water vapor are dominant controls on atmospheric oxidative capacity, driving numerous reactions relevant to ozone formation (e.g., Kleinman et al., 2001). The use of FNR reduces this inherently multidimensional and nonlinear chemical system to only two dimensions, thereby concealing key variability of the light and water vapor.

To illustrate this limitation, I perturbed photolysis rates over polluted regions during the KORUS-AQ campaign using the observationally constrained F0AM model (Souri et al., 2023; 2025a; 2025b). Photolysis frequencies were multiplied by factors of 0.5 (dim), 1.0 (default), and 2.0 (bright), producing three corresponding sets of PO3 isopleths. Systematic increases in photolysis enhanced both net PO3 and its sensitivities to NOx and VOCs, as evidenced by more compact isopleths in the bright-light scenario (each contour shown corresponds to 3 ppbv hr-1). This demonstrates that identical FNR values can correspond to substantially different sensitivity regimes depending solely on available sunlight. Moreover, in situ observations show no meaningful correlation between FNR and photolysis rates, as the latter depend on solar zenith angle, altitude, surface albedo, column ozone, and particles.

Link to the figure:

https://drive.google.com/file/d/1055kbTHJ01fdjA2WcFbWokGeuExicxj4/view?usp=sharing

This is a fundamental point at which FNR falls apart.

Response:

The authors agree that the FNR has inherent limitations and does not fully represent the VOCR and reactive nitrogen. Additionally, the effects of solar radiation, along with other factors such as water vapor, further complicate the O_3 formation processes. Moreover, under certain environmental conditions, processes such as alkyl nitrate formation, peroxyacetyl nitrate formation (Kleinman, 2005), and aerosol uptake (Sakamoto et al., 2019) can contribute non-negligible influences on O_3 production via radical loss pathways. To better use FNR for analyzing O_3 formation sensitivity, we investigated the availability for altitude-dependent FNR and its improvement, and focused on strong photochemical conditions in the present study.

In the current manuscript, we attempted to improve the FNR by introducing FNR_{sec}. The figure below shows the variation of secondary and primary HCHO based on our model

simulations at the study locations. Secondary HCHO exhibited an inverted U-shaped diurnal variation, whereas primary HCHO showed a U-shape pattern. These diurnal variations are consistent with previous studies of secondary and primary HCHO (Xue et al., 2022; Yang et al., 2019).

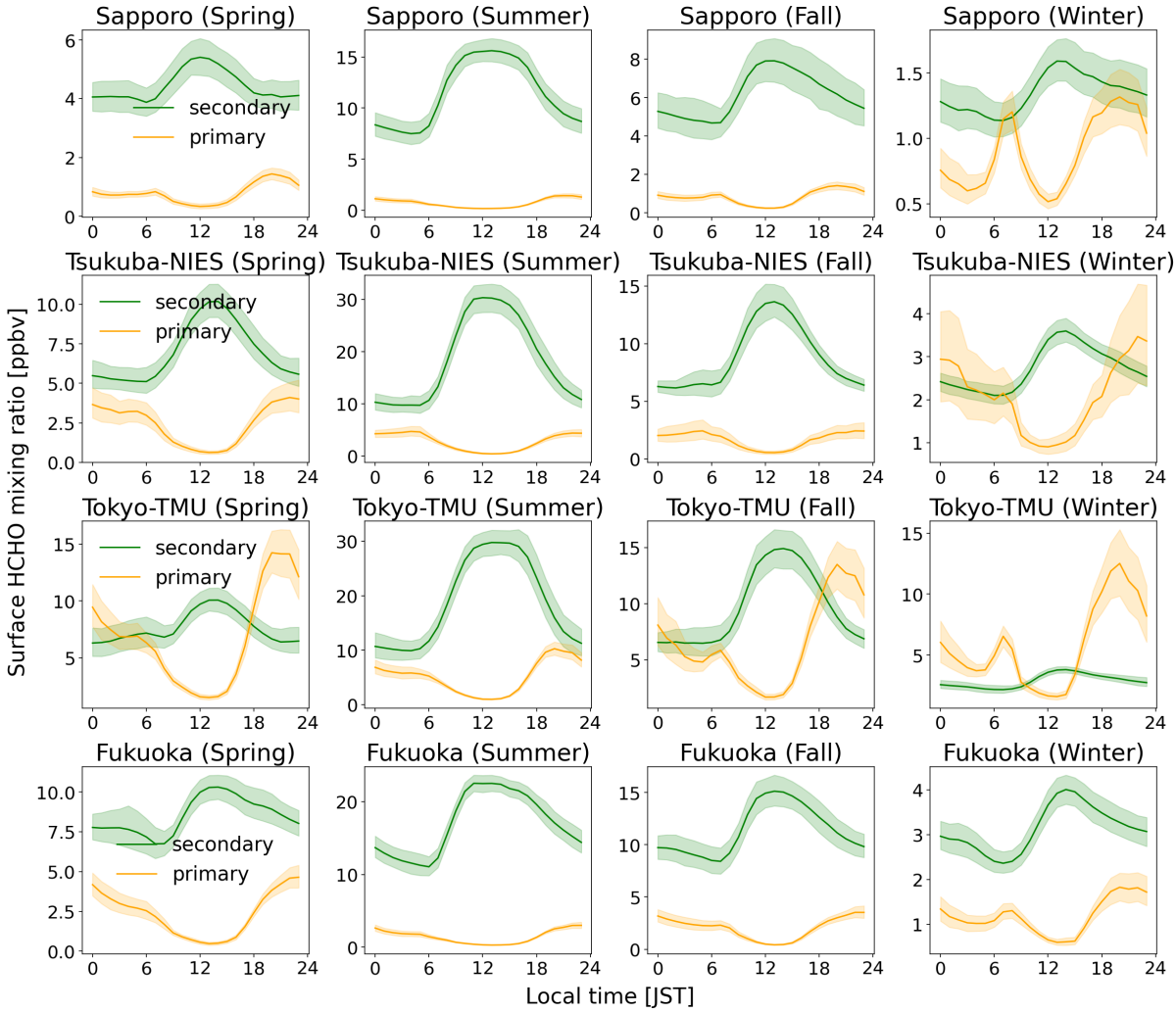

Figure: The distribution of secondary (green) and primary (orange) HCHO derived from the GEOS-Chem model simulations at the study location.

Secondary HCHO production depends on both VOC reactivity and light intensity, as can be seen between summer and winter and between Sapporo and the other study locations. Thus, FNR_{sec} can partially reflect the photolysis frequencies of sunlight. The term "VOC-limited" may be inadequate during low-light winter conditions; therefore, we adopted the term "RO_x-limited", as recommended by Schroder et al. (2017), for a more accurate description. In addition, we will focus our O_3-sensitivity diagnosis on Pandora observations around noon, when sufficiently similar and strong photochemical conditions can be assumed.

Primary HCHO also affects O_3 production by acting as a VOC and producing OH and HO_2 radical (Lei et al., 2009). However, primary HCHO is not a secondary product and thus does not directly serve the purpose of the FNR (Chang et al., 2025). As shown in the figure above, variations in primary HCHO are mainly associated with anthropogenic activities.

The manuscript attempts to classify air parcels into sensitivity regimes even during periods of weak photochemical activity (e.g., winter, early morning, late evening). Such classifications are utterly nonsensical. Under low-photolysis conditions (an entirely separate dimension of ozone chemistry, as discussed in Chatfield et al. (2010) and Souri et al. (2025)), ozone production becomes largely insensitive to perturbations in NOx and VOCs.

Response:

We have revised the manuscript to focus on O_3 diagnosis under strong photochemical conditions.

In the previous version of the manuscript, we classified air masses to examine their effects on the vertical profiles of NO_2 and HCHO. We did not investigate the O_3 sensitivity regimes of individual air parcels in detail because of other influencing factors such as solar intensity. We observed several NO_2 spikes at the Tsukuba-NIES site during summer; therefore, we revised the discussion to focus on summer air parcels (page 12, lines 422-440).

"The NO_2 distributions more rapidly decreased with increasing altitude compared to HCHO. Unlike HCHO, the bulk of NO_2 was generally concentrated below 1 km because NO_x emissions are mainly near the surface (i.e., traffic). Normally, the vertical NO_2 profile was enhanced in winter. However, we also observed several spikes of NO_2 during summer at Tsukuba-NIES. Generally, NO_2 concentrations are low in summer because of photochemical loss. The enhanced NO_2 profiles observed at Tsukuba-NIES during summer could be attributed to the transport of emissions from surrounding regions. Irie et al. (2021) reported that summertime air masses passing over Tokyo Bay area and reaching Tsukuba lead to a greater O_3 concentrations at this site.

To investigate whether any external transport affected the vertical profiles, we applied the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model for the Tsukuba-NIES case. The model, developed by the National Oceanic and Atmospheric Administration (NOAA) Air Resources Laboratory (ARL), has been widely used for atmospheric trajectory and dispersion calculations (Stein et al., 2015). We simulated 48-hour backward trajectories for the year 2022, with endpoints at 13:00 local time (JST) at Tsukuba-NIES, as a case study. The backward trajectories were assigned into four clusters: c1, c2, c3, and c4. The number of clusters was optimized using total spatial variance (TSV) analysis. The cluster frequencies were 15 %, 40 %, 36 %, and 9 % for c1, c2, c3, and c4, respectively (Fig. S7). Figure S8 shows the vertical profiles of NO_2 at Tsukuba-NIES during summer as a function of air mass clusters. During summer, cluster c1 occurred on only one day, clusters c2, c3, and c4 occurred on 34, 33, and 24 days, respectively. Thus, we observed elevated in NO_2 profiles during cluster c4, in which air masses passed through urban Tokyo and industrial areas, transporting anthropogenic pollutants to the Tsukuba-NIES site. This suggests that external pollution transport affected the vertical distributions of NO_2, and consequently, O_3 production."

This figure from Souri et al. 2025 clearly shows this using five variables derived from TROPOMI and our PO3 parameterization across two seasons in Los Angeles. During December, January, and February, FNR values are low due to abundant NO2 relative to HCHO, implying a VOC-limited regime. However, the derivatives of PO3 with respect to HCHO and NO2 are substantially muted due to weak photochemistry, making PO3 largely unresponsive

to precursor concentrations. In contrast, summer conditions show markedly stronger sensitivities. The same logic extends to diurnal cycles, including differences between early-morning, late-evening, and afternoon Pandora observations.
The link to the figure: https://drive.google.com/file/d/1h2j8Wb0z9vVX-sb0FHVnEpe7-mAJaHns/view?usp=sharing
These findings underscore that discussions of PO3 magnitude or precursor sensitivity are of limited relevance during winter or under low-light conditions. This issue is not addressed in the current manuscript. Although the manuscript discusses diurnal variability in HCHO and NO2, which is interesting, its interpretation implicitly assumes that the sun remains constant throughout the day. Additional processes with intense diurnal cycles, such as HONO chemistry and water-vapor-dependent OH production, further complicate the picture. *Even without including these processes, the incompleteness of FNR with respect to photolysis rates alone is sufficient to undermine its interpretive value.*

Response:

The FNR has certain limitations; however, it is still a common photochemical indicator for diagnosing O_3 sensitivity. Determination of FNR is relatively simple and can be achieved from satellite, ground-based monitoring measurements, which have been well developed worldwide. In contrast, the more precise indicators such as H_2O_2/HNO_3 and LRO_X/LNO_X typically require extensive modeling efforts and specific measurements, which can be challenging for many research groups. To better account for photolysis effects, we investigate FNR_{sec} instead of the FNR_{total}.

We have replaced the term "VOC-limited" with "RO_x limited" to better interpret O_3 sensitivity regime under low-light conditions. Additionally, we have revised our manuscript to focus on interpreting of FNR and O_3 sensitivity during O_3 exceedance events using Pandora observations around noon. Under strong photochemical conditions, the differences between interpretation based on PO_3-LROx/LNOx and PO_3-FNR are minimized (Abdi-Oskouei et al., 2022).

Although FNR values under low-light conditions in the early morning and late afternoon may lead to significantly uncertain interpretation of O_3 production, it is still worthy to understand the development of FNR values. Therefore, we have moved the Figures 6 and 7 showing diurnal and seasonal variations of surface and column FNR in Section 3.4.1 and 3.4.2 in the previous preprint to the supplementary information (Figs. S9 and S10) and only discuss on the variation of FNR (page 14, lines 590-595):

"The diurnal and seasonal cycles of surface and column FNR are shown in Figs. S9 and S10. Surface FNR_{sec} developed in the early morning and decreased in the afternoon. During the 8-hour daytime period (8:00 to 16:00), the average surface FNR_{sec} was 0.99 ± 0.46 in July and 0.72 ± 0.64 in October. Using tropospheric vertical column densities derived from Pandora sky-scan observations, the column FNR_{sec} exhibited a higher value compared to those from surface measurements. Like surface FNR_{sec}, the tropospheric column FNR_{sec} increased during the morning. The highest column FNR_{sec} values were found during the summer months (JJA). In contrast, these values were normally below 1 in winter (DJF)."

If the authors wish to pursue publication in the current journal or another one, the following steps are highly recommended:
i) Restrict interpretations of FNR and ozone sensitivity to periods with active photochemistry, such as summertime and midday/afternoon hours.

Response:

We have revised our manuscript to focus on interpreting of FNR and O_3 sensitivity during O_3 exceedance events using Pandora observations around noon.

ii) Discuss the dependence of photolysis rates on altitude, emphasizing that photolysis frequencies can be at least 20–50% higher between 2–4 km altitude compared to the surface, substantially altering the magnitude of PO3 sensitivities.

Response:

We have added the dependence of photolysis rate for NO_2 on altitude obtained from the GEOS-Chem one-year benchmark. The vertical distribution of secondary HCHO which increases with altitude, also reflecting the photolysis rate. We have added this discussion to Section 3.1.2 (page 7, lines 275-280).

"For the tropospheric column, the seasonal contribution of secondary HCHO remained consistent. Additionally, the secondary HCHO contribution increased with altitude (Fig. S4a). The contribution at altitudes above 2 km was almost entirely attributed to secondary formation. This vertical distribution depends on primary emission sources, VOC oxidation, and photolysis rates. The higher photolysis frequency above 2 km compared to surface (Fig. S4b) therefore enhanced VOC oxidation as well as the magnitude of O_3 production."

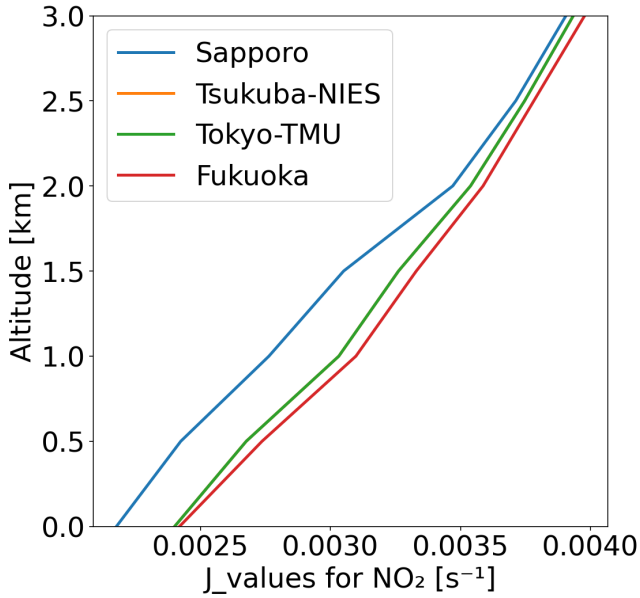

Figure: Photolysis rate (J_values) for NO_2 at the study locations

iii) Include a dedicated limitations section addressing uncertainties and assumptions inherent in FNR. Key sources of error have been quantified in Schroeder et al. (2017), Souri et al. (2020, 2023), and Jin et al. (2017). This section should explicitly note that FNR is insensitive to both

water vapor and photolysis rates, making FNR-based chemical classification incomplete and, in specific contexts, obsolete. Please quantify the errors of GEOS-Chem, the errors in Pandora, and other assumptions to find a set of threshold values. Will the errors be so large that the clear definition of the regimes becomes compromised?

Alternative approaches, including joint inversion/data assimilation frameworks (https://acp.copernicus.org/articles/20/9837/2020/) or data-driven parameterizations of PO3, such as those presented in Souri et al. (2025) (https://egusphere.copernicus.org/preprints/2025/egusphere-2025-1679/), offer more physically grounded paths forward.

The specific method is less important than the need for the community to recognize the intrinsic limitations of FNR and move toward more comprehensive representations.

Response:

We have added the limitations of FNR in the Introduction (page 2, lines 66-69).

"The FNR has inherent limitations in representing O_3 chemistry (Souri et al., 2023a). A wide transition/ambiguous range of FNR values has been reported compared with the more precise LNO_x/LRO_x ratio (Schroeder et al., 2017). Consequently, under certain conditions, FNR may misclassify O_3 formation sensitivity. Nonetheless, FNR remains a commonly used indicator because it can be readily obtained and does not require extensive modeling."

We already provided the errors in Pandora measurements, in Section 2.2 (page 3, lines 140-144):

"The direct-sun mode measures total NO_2 column with high precision (2.7×10^{14} molecules cm^{-2}) and accuracy (2.7×10^{15} molecules cm^{-2}). For total HCHO column, a statistical error of 6 % and a systematic error of 26 % have been reported (Spinei et al., 2018). The bias of the sky-scan measurement is approximately -0.02×10^{16} molecules cm^{-2} for NO_2 and -0.05×10^{16} molecules cm^{-2} for HCHO (Tirpitz et al., 2021; Verhoelst et al., 2021)."

We have added interpretive discussion explaining the method uncertainty-based model in Section 3.3 (pages 13, lines 563-569).

"For the column, the transition range for Sapporo was defined as 0.86–1.52 where the cumulative probability of NO_x-limited and RO_x-limited condition reached 95%, corresponding to a 5% probability of misclassification. Consequently, 15% of NO_x-limited and 15% of RO_x-limited conditions were incorrectly classified as transitional (Fig. 7). Similarly, the probabilities of misclassifying NO_x-limited and RO_x-limited conditions as transitional were 20% and 25%, respectively, for Tsukuba-NIES, 35% and 25% for Tokyo-TMU, and 15% and 60% for Fukuoka. Notably, the probability of misclassification of RO_x-limited condition at Fukuoka was higher than at the other locations. Because we simulated for the year 2022, this uncertainty could be reduced by extending the model simulations to longer time periods."

Because the model simulations were performed for only one year (2022). The uncertainty could be improved by extending the simulations to a longer period. Jin et al. (2017) reported approximately 10% misclassification of O_3 sensitivity regimes based on model simulations covering 2006–2012.

Why exclude the primary source of HCHO? I found it puzzling that the authors are so focused on separating primary from secondary formaldehyde. The title of the manuscript (which I think is unclear) appears to suggest that the goal is to study secondary formation pathways. However, the manuscript does not provide a strong justification for why this focus is necessary. Primary HCHO also affects both net ozone production rates and their sensitivities to NOx. Because HCHO is reactive, even directly emitted HCHO can be an important source of HO2, accelerating the ROx–HOx cycle and enhancing ozone formation per molecule of NOx. In many regions, the primary source of HCHO, which is a regulatory concern, can substantially contribute to ozone production. Ignoring it weakens the motivation of the study.

Response:

We have briefly explained the reason to focus on secondary HCHO. Secondary HCHO production results from both VOC reactivity and light intensity. Thus, FNR_{sec} could reflect the photolysis frequencies of sunlight. The authors agree that primary HCHO also affects O_3 production because it acts as VOC and produces HO_2 radical. However, primary HCHO is directly connected with anthropogenic activities. Additionally, primary HCHO is not a secondary product and thus does not directly serve the purpose of the FNR (Chang et al., 2025).

*Ozone titration by NO is not the issue***.** The manuscript repeatedly brings up the influence of NOx on ozone through titration in NOx-saturated environments, but this is not the central point of defining chemical sensitivity regimes. Classification of ozone chemistry is about determining whether radical termination occurs through the loss of ROx or through the loss of NOx, and how the relative amounts of NOx and VOCs shift that balance. When NOx is extremely high relative to VOCs, the radical chain that drives ozone formation is suppressed. As a result, the efficiency of producing ozone per molecule of NOx becomes very low, meaning low P(O3)/NOx. Ozone titration itself is not particularly relevant to chemical sensitivity or to regulatory interpretation.
Titration is rapid, local, and temporary, and it is easily reversed during the day through photolysis. What matters are the processes that are not easily reversible, such as the formation of H2O2, HNO3, or organic nitrates. In NOx-saturated regimes, adding more NOx removes OH and reduces the radical pool through HNO3 formation. This is the central mechanism that determines chemical sensitivity, not the short-term titration of ozone by NO.

Response:

We have revised the explanation based on the LRO_X and LNO_X theory.

What do we learn from validating the model? The authors validated GEOS-Chem against in situ and ground-based remote sensing data, but found substantial biases (especially for ozone). But I found it to be highly disjointed from the rest of the paper. What do they imply for the rest of the manuscript? Are these errors so significant that we cannot confidently determine the chemical conditions? Are they indicative of a missing source or mechanisms? What is the connection of the analysis to the rest? If the goal is to build confidence in the model, it needs to be compared with a large set of modeling efforts to understand how far it falls short.

Response:

The purpose of comparing model simulations with in-situ and ground-based remote sensing measurements is to build confidence in the model. The model simulations could not well generate the nighttime variation of O_3 may be attributed to insufficient mechanism of O_3 deposition. However, during daytime model simulations could reproduce the O_3 production at the study sites. We compared one year dataset with model simulations to cover seasonal conditions.

The model simulations show good agreement with the Pandora HCHO and NO_2, thereby reducing biases in the simulated column FNR. Therefore, we focus on applying the column FNR threshold values determined from the model for O_3-sensitivity diagnosis.

Storytelling. Another issue is that the authors pick and choose what to show in the primary draft versus the supplementary material. I think all stations should be included in the primary draft. There should be more emphasis on the evolution of NO2 and HCHO, and less on the implications of ozone sensitivities arising from the factors above. The study also ends a bit prematurely. What do we learn from Pandora that we cannot do using satellites? How important is it to look at the vertical components for regulators? Are we expecting to see changes in emissions or composition at higher altitudes (changes in aviation)?

Response:

We have revised the manuscript and present results from all stations in the main text (Sections 3.2 and 3.4).

Satellite observations have limitations in accuracy, particularly for HCHO retrievals. Temporal averaging (e.g., monthly means) can conceal daily information in O_3 production. In addition, satellite retrievals are more sensitive to higher atmospheric layers, which also hinders the interpretation of near-surface O_3 production.

Pandora provides higher accuracy of both NO_2 and HCHO and is more sensitive to near-surface layers, making it appropriate for investigate O_3 production within the PBL. Vertical profile obtained from Pandora sky-scan also essential for assessing the vertical sensitivity relevant to O_3 formation. We investigated the dependence of FNR on altitude and its improvements for assessing O_3 formation sensitivity. Every O_3 productions within the PBL markedly contribute to tropospheric O_3 pollutions.

We have added a details sensitivity analysis on specific O_3 exceedance events at the study location to better support policy purposes (Section 3.4).

Specific comments:

Line 27: In the presence of sunlight. That's a critical knob missing from the analysis.

Response:

We revised the sentence (page 1, lines 27-28):

"Tropospheric ozone (O_3) is a central secondary pollutant formed through photochemical reactions involving its main precursors: nitrogen oxides (NO_x, including NO and NO_2) and volatile organic compounds (VOCs) in the presence of sunlight"

Line 40. FNR is only a subset of significant (and more robust) indicators such as H2O2/HNO3 or Ox/NOz. You may want to say that FNR gained popularity because it can be accessed from remote sensing satellites. However, if H2O2/HNO3 were not limited to limb sounding, it would be preferred over FNR, as it directly explains the chemical loss of peroxy radicals over the chemical loss of NOX.

Response:

We have introduced the H_2O_2/HNO_3 and LRO_x/LNO_x ration (pages 2, lines 53-57).
"Sensitivity analyses have been conducted using various approaches, including model-based methods (Thorp et al., 2021), absolute sensitivity analysis (Sakamoto et al., 2019), and photochemical indicators such as the formaldehyde-to-nitrogen dioxide ratio (FNR) (Jung et al., 2022; Qian et al., 2024; Souri et al., 2023b), the robust H_2O_2-to-HNO_3 ratio (Hammer, et al., 2002; Souri et al., 2023a), and the LRO_x/LNO_x ratio (Abdi-Oskouei et al., 2022; Schroeder et al., 2017)."

Line 40. NMHC/NOx is a very crude ratio. It must be reactivity-weighted VOCs over NOx.

Response:

We have removed the $NMHC/NO_x$ ratio (page 2, line 57).

Line 42. FNR is not the most precise indicator. Please introduce the HOx-ROx cycle notion and the robustness of H2O2/HNO3.

Response:

Thank you for the correction. We revised the sentence (page 2, lines 68-69):
"Nonetheless, FNR remains a commonly used indicator because it can be readily obtained and does not require extensive modeling."

Line 45. A proxy for VOCs reactivity, not VOCs.

Response:

We have revised it (page 2, lines 59-60).
"Meanwhile, HCHO reflects the VOC oxidation strength and is widely used as a proxy for VOCs reactivity (Irie et al., 2021)."

Line 48. I don't understand how the primary source of HCHO can be misleading. HCHO is reactive so it has an influence on PO3 by itself.

Response:

We revised the statement (page 2, lines 61-63):
"Primary HCHO is directly emitted from anthropogenic activities; therefore, considering primary HCHO may be misleading in the assessment of VOCs reactivity."

Line 50. More recently? HCHO/NO2 was introduced 25 years ago.

Response:

We have removed "More recently" (page 2, line 64).

The whole paragraph from L35 onward should be significantly improved. It is unclear what message is conveyed. Please begin by discussing the controlling factors of the HOX-ROX cycle and where FNR fits into this picture. Unfortunately, Sillman's and Kleinman's pioneering works have not been recognized here:
Sillman, S. and He, D.: Some theoretical results concerning O3-NOx-VOC chemistry and NOx-VOC indicators, J. Geophys. Res., 107, 4659, https://doi.org/10.1029/2001JD001123, 2002.
Sillman, S., Logan, J. A., and Wofsy, S. C.: The sensitivity of ozone to nitrogen oxides and hydrocarbons in regional ozone episodes, J. Geophys. Res., 95, 1837–1851, https://doi.org/10.1029/JD095iD02p01837, 1990.
Kleinman, L. I., Daum, P. H., Lee, Y.-N., Nunnermacker, L. J., Springston, S. R., Weinstein-Lloyd, J., and Rudolph, J.: Sensitivity of ozone production rate to ozone precursors, Geophys. Res. Lett., 28, 2903–2906, https://doi.org/10.1029/2000GL012597, 2001.

Response:

We introduced the HO_x-RO_x cycle in the first paragraph of the Introduction (page 1, lines 34-38).
"Internally, O_3 production is initiated by the formation of peroxyl radical (RO_x), generated through reactions between VOCs and hydroxyl radical (OH). O_3 is subsequently produced when RO_x radicals (HO_2 or RO_2) react with NO, leading to radical propagation (Kleinman et al., 2001; Sillman and He, 2002). The radical termination proceeds control the O_3 production. These proceeds occur either through the reaction of OH with NO_x to form nitric acid (referred as LNO_x) or through radical-radical reactions (referred as LRO_x) (Schroeder et al., 2017)."

Line 54. Please remove "some".

Response:

We have removed "some" (page 2, line 76).

Line 59. Some issues have prevented people from using Pandora. Aside from the lack of high spatial coverage, there are lingering issues with Pandora HCHO retrievals that may need to be disclosed here. In addition, in very high SZA, the airmass Pandora observes may not be fully representative of the overhead columns (the homogeneity assumption used in the application of geometric AMF gets violated).

Response:

Pandora direct-sun and sky-scan modes provide accurate total column amount and lower tropospheric column of NO_2, respectively. Pandora direct-sun HCHO have been reported

with significant error budgets (Spinei et al., 2018). We already mentioned this in Section 2.2 (page 3, lines 140-144):

"The direct-sun mode measures total NO_2 column with high precision (2.7×10^{14} molecules cm^{-2}) and accuracy (2.7×10^{15} molecules cm^{-2}). For total HCHO column, a statistical error of 6 % and a systematic error of 26 % have been reported (Spinei et al., 2018). The bias of the sky-scan measurement is approximately -0.02×10^{16} molecules cm^{-2} for NO_2 and -0.05×10^{16} molecules cm^{-2} for HCHO (Tirpitz et al., 2021; Verhoelst et al., 2021)."

Sky-scan HCHO retrievals have been shown to have better quality (Zhao et al., 2025). Previous studies have deployed Pandora sky-scan derived vertical profiles of NO_2 and HCHO for satellite validation, model comparison, and investigations of their vertical variability (Alwarda et al., 2025; Mouat et al., 2024; Zhao et al., 2025). Therefore, we have revised the manuscript to focus on the application of Pandora sky-scan observations around noon during high-O_3 episodes to maximize data accuracy.

L79. What do you mean by oxidant levels? Ox? Ox is immune to NO titration because O3+NO2 won't change under titration. When we label a regime as NOx-saturated, it means NO2+OH is taking place. Titration is not a concern because once the sun is out or the air mass ages outside of super emitters, the titration effects go away, and NO2 will contribute to O3. In other words, Ox will remain the same in both conditions, but the partitioning between NO2- and NO-O3 changes. This isn't important for regulations. What matters for regulations is the formation of H2O2 and HNO3, which can be mostly lost in the system (not always).

Response:

The increase in average ambient concentrations of oxidants (O_x, a collective term, of which the major components are O_3, peroxyl acetyl nitrate, hydrogen peroxide, and organic hydroperoxides) in Japan during the 2000s has been reported by Akimoto, (2017). We meant that oxidant levels may change under different emission scenarios. We have revised this statement for clarification (page 3, lines 112-114).

"Despite the emission reductions, O_3 level remain unpredictable due to non-linear photochemical processes and an increase in transboundary transport (Akimoto, 2017; Irie et al., 2021)"

Line 135. "a sensitivity run" is ambiguous.

Response:

We have removed the run's name to avoid confusion (page 4, line 172).

Figure S1 is confusing because the signs are the opposite. It suggests that reducing VOCs should lead to higher ozone levels.

Response:

The authors would like to thank the reviewer for the opportunity to clarify Figure S1 in the SI. Figure S1 presents change in O_3 concentration due to emission perturbations. Change in O_3 is defined by subtracting Run-3 or Run-4 from Run-2. Positive O_3 change means emission

reduction improve O_3 pollution levels and vice versa negative O_3 change implies that emission reduction leads to an increase in O_3 concentration. Therefore, positive O_3 change corresponded to VOCs reductions (Run-2 minus Run-4) implies lower O_3 levels. We have added the definition of O_3 change in the Figure S1 caption.

"Figure S1: GEOS-Chem model simulations of changes in the surface ozone concentration at 13:00 in response to NO_x emission reduction (a, c) and VOC emission reduction (b, d). Change in O_3 is defined by subtracting Run-3 or Run-4 from Run-2. The top panels present changes in summer, while the bottom panels present changes in winter. Red and blue colors indicate positive (decrease) and negative change (increase) in the surface ozone concentration, respectively."

Section 3.2.1. Supplementary figures should be used only for supplementary information. This section is deliberately designed to discuss model comparisons vs. surface observations, necessitating the inclusion of figures in the primary draft.

Response:

We have moved the scatterplots comparing model simulations with in situ and Pandora measurements from the supplementary information to the main text (page 6).

Section 3.2.1. It's interesting to see HCHO being higher in Oct than in July. What is contributing to it, given that the model couldn't see this tendency? Could this be caused by different air masses or primary anthropogenic HCHO levels? It's also interesting that the observed columns show an opposite trend, given that most of HCHO is confined in the first few kms. Are we confident in the accuracy of the HCHO measurements?

Response:

We found negligible differences in surface HCHO concentrations between July and October (t-test, p-value > 0.05). The mean HCHO concentrations in July and October were 2.77 ± 1.73 and 3.06 ± 2.10 ppbv, respectively. Surface HCHO is driven more by anthropogenic activities, in particular at Tokyo-TMU location. A higher PBLH and stronger photolysis in summer may dilute anthropogenic HCHO, whereas a lower PBLH in fall may lead to its accumulation and increase concentrations.

Surface HCHO was measured using a SIFT-MS instrument. In the previous preprint, mixing ratios for HCHO calculated from rate constants and branching ratios in Syft Technologies database were used. Recently, we obtained a correction factor of 1.52 for HCHO by comparing ambient air measurements from the SIFT-MS with those from a calibrated Picarro HCHO monitor. We have corrected the surface HCHO data in the revised manuscript.

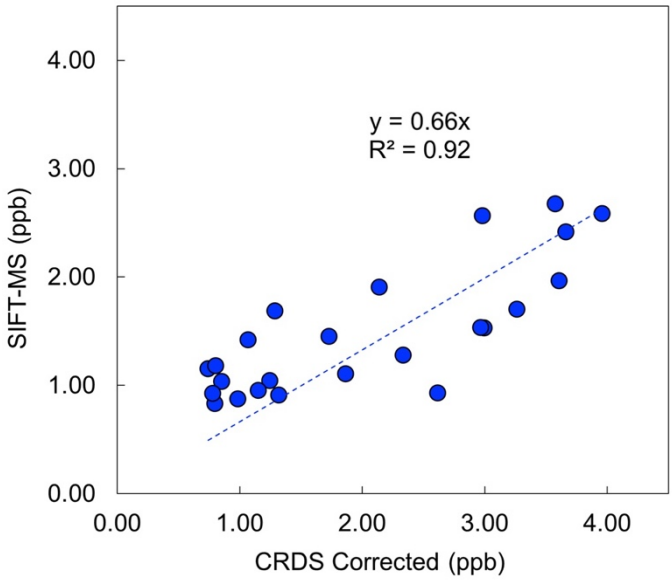

Figure: Scatter plots of SIFT-MS HCHO (y-axis) versus Picarro HCHO (x-axis). The data are fitted by y = ax.

Column HCHO was retrieved from Pandora measurements using the L2 (Level 2) Air-Ratio Sky Algorithm developed by Elena Spinei and the Pqandora Global Network (Cede et al., 2021). We have carefully screened the data to ensure the quality. We have plotted the TROPOMI monthly HCHO data for July and October. Higher HCHO column densities in July were also observed by TROPOMI, consistent with the Pandora measurements.

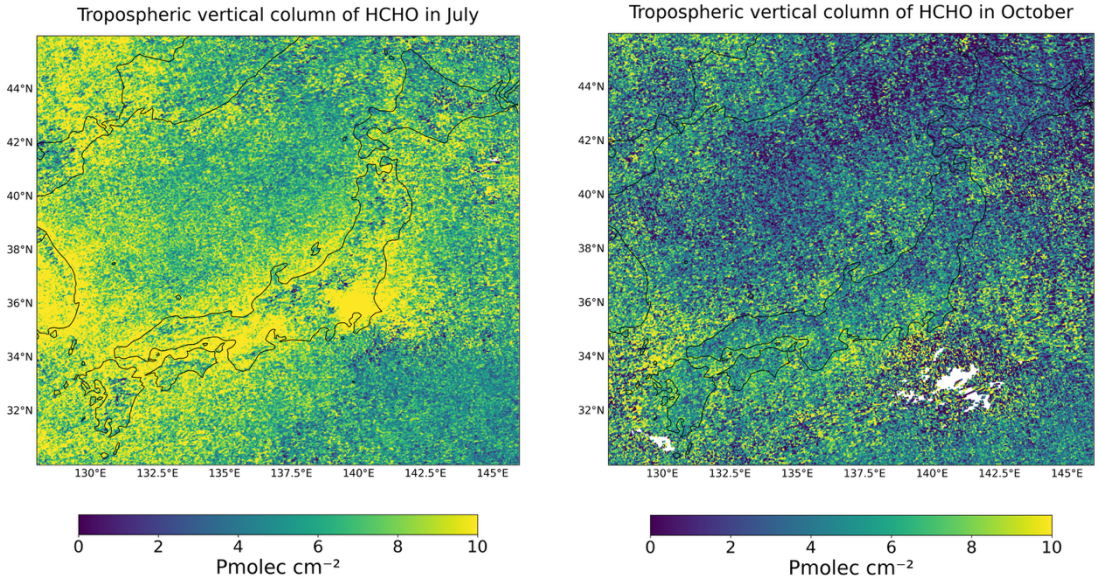

Figure: Monthly tropospheric vertical column of HCHO derived from oversampling TROPOMI data in July (left) and October 2022 (right).

Line 253. I disagree with this statement. Surface measurement is designed to monitor the surface, and there is nothing inherently wrong about it. Columnar data represents something

else. I don't think the discrepancies indicate that each domain is inferior. They represent different regions.

Response:

We meant column information complement to surface HCHO measurements to better understand atmospheric HCHO. We have revised the statement for clarification (page 8, lines 315-316):

"This suggests that column information is essential and complement to surface measurements for fully representing atmospheric HCHO."

Line 260. You have cited this paper elsewhere, and it may be highly relevant. We examined the individual tendencies shaping HCHO concentrations (Fig. 7): https://www.sciencedirect.com/science/article/pii/S1352231023003552

Response:

We have added citation as suggested by the reviewer.

Figure 3. Interestingly, PBLH did not change much throughout the seasons. Is it expected over Japan?

Response:

The PBLH data discussed in this study derived from GOES-Chem simulations driven by MERRA2 meteorological reanalysis. Below, we also compared these results with ERA5 PBLH data at the grid boxes nearest to the Pandora stations. ERA5 is obtained from Climate Data Store (C3S, 2018). ERA5 PBLH are slight lower than those from MERRA2. However, the PBLH at the study locations is similar between the two dataset and shows little seasonal variability.

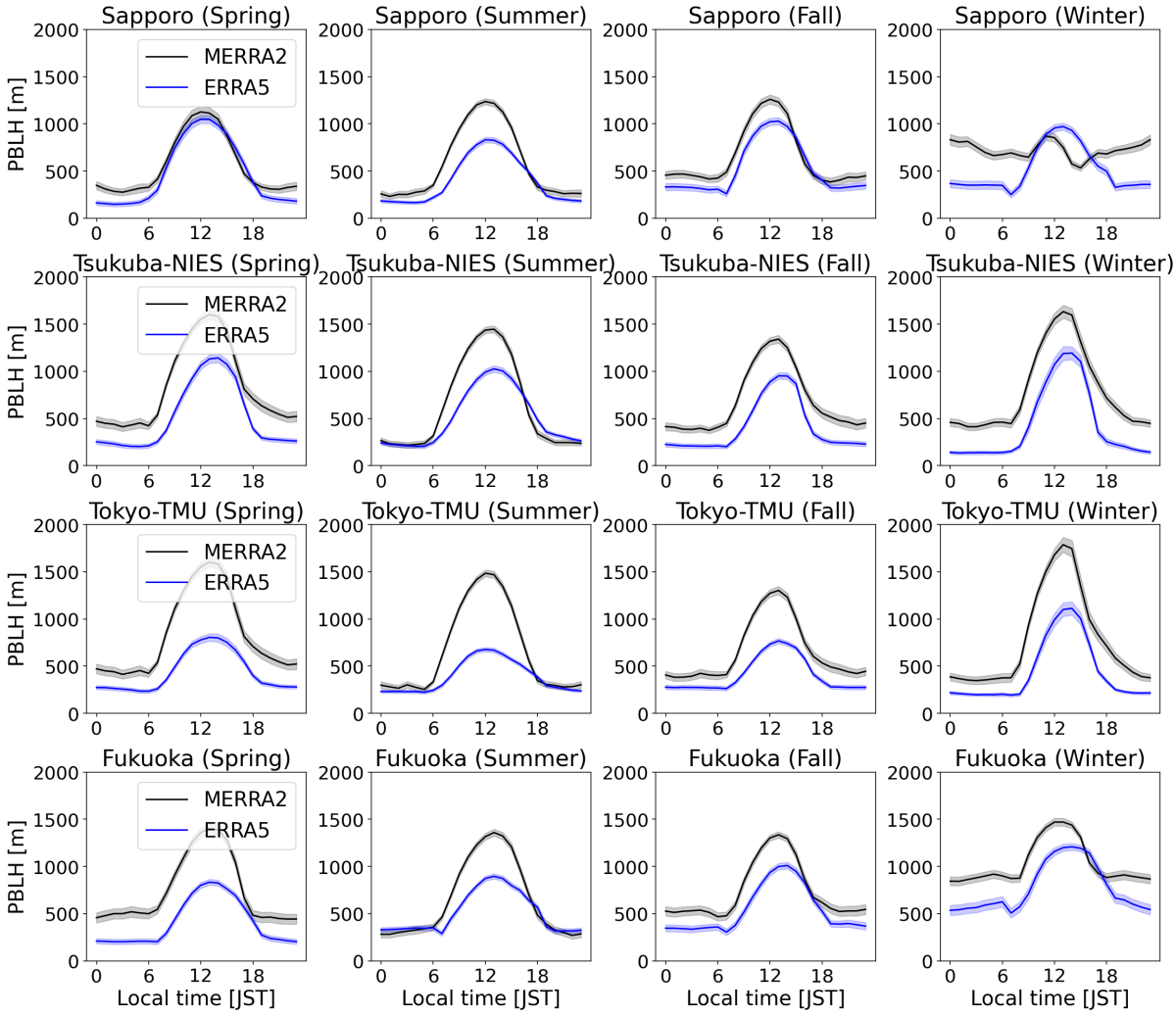

Figure: Planetary boundary layer height (PBLH) at the study locations.

Line 295. Could anthropogenic VOC oxidation also contribute to the enhancement?

Response:

We have revised the manuscript to focus on strong photochemical conditions using Pandora observations around noon. Therefore, we have removed this sentence from line 295 "In the current study, higher amounts of HCHO commonly occurred in the late afternoon in spring and fall". We have also added a discussion on anthropogenic VOC oxidation during summer. We separated the biogenic and anthropogenic VOCs from model diagnosis and estimated their contributions to O_3 formation potential (OFP). OFP is estimated by multiplying VOC with maximum incremental reactivity coefficient (Carter, 1994). OFP indicates VOC oxidation.

In summer, the increase emission from biogenic sources primarily contribute to OFP. Anthropogenic VOCs emissions do not change much, may be slightly increase due to evaporation in summer (Wu et al., 2024). Normally, that increase is negligible compared to biogenic sources. Typically, biogenic VOCs may still account for the majority VOC reactivity and therefore of HCHO production even in urban areas and can blur the role of anthropogenic VOCs (Zhao et al., 2025). However, for Tsukuba-NIES, and Tokyo-TMU, the anthropogenic

VOC reactivity is important even in summer. This anthropogenic contribution also increases in fall and spring. We have revised the statement (pages 11-12, lines 407-415):

"The higher HCHO production at Tsukuba-NIES and Tokyo-TMU compared to Fukuoka suggests that, in addition to biogenic VOCs, anthropogenic VOCs play an important role in HCHO production (Fig. S6). Anthropogenic VOC accounted for 20% of the total VOC oxidation potential at the two locations. The summertime increase in anthropogenic VOCs emissions is likely driven by enhanced evaporation processes (Wu et al., 2024)."

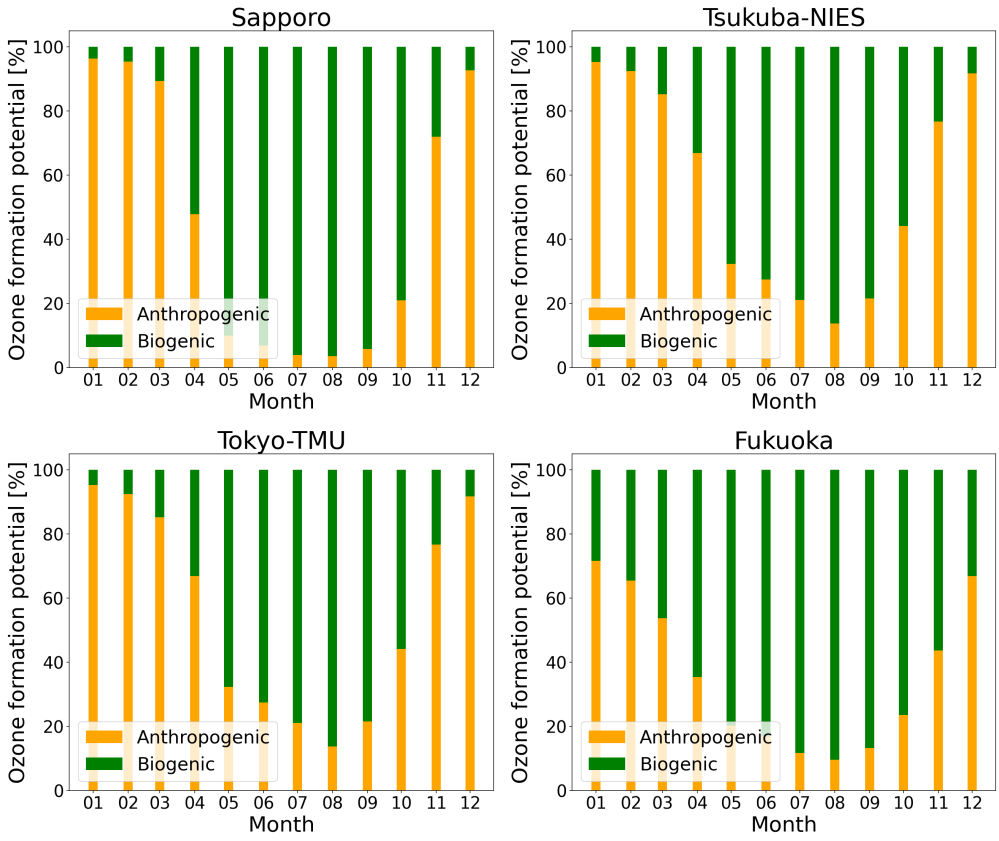

Figure: Biogenic and anthropogenic VOCs contributions to O_3 formation potential.

Line 300. This isn't a strong argument. If by "exceedances" you meant high ozone episodes, those could have been caused by factors such as regional background, vertical contributions, and other precursors. I agree that high ozone exceedance usually is associated with high HCHO levels because large HCHO concentrations are indicative of favorable meteorological conditions to form ozone, but it is not the only factor.

Response:

We intended to emphasize the importance of the vertical profile for O_3 sensitivity analysis. We have revised this argument from the manuscript for clarification (page 12, line 419-421). "HCHO extended to higher altitudes could be attributed to vertical and horizontal transport and chemistry (Souri et al., 2023c). For a more accurate FNR analysis, it is essential to consider not only surface but also elevated measurements."

L302. This is precisely why I think it is not interesting to study surface ozone sensitivity. Every parcel within the PBL is important for ozone production rates. This is why Souri et al. (2023; 2025a, 2025b) focused on the PBL region rather than the surface.

Response:

We have cited the references for this statement.

Section 3.2.3. Unfortunately, only one station is shown here. Also, it's essential to include the trajectory characteristics somewhere in the primary draft. This could be a major comment, but I mention it here: the location of trajectories is one thing, and when it occurs more frequently is another. This part needs to be elaborated to understand how often they happen before delving into where they are coming from.

Response:

Since we observed several enhancements in NO_2 during summer at Tsukuba-NIES, we wanted to investigate the air mass at this site. We have described the frequently of the air masses (page 12, lines 434-440):
"The cluster frequencies were 15 %, 40 %, 36 %, and 9 % for c1, c2, c3, and c4, respectively (Fig. S7). Figure S8 shows the vertical profiles of NO_2 at Tsukuba-NIES during summer as a function of air mass clusters. During summer, cluster c1 occurred on only one day, clusters c2, c3, and c4 occurred on 34, 33, and 24 days, respectively. Thus, we observed elevated in NO_2 profiles during cluster c4, in which air masses passed through urban Tokyo and industrial areas, transporting anthropogenic pollutants to the Tsukuba-NIES site. This suggests that external pollution transport affected the vertical distributions of NO_2, and consequently, O_3 production."
We have added more figures in the main text; therefore, we would like to keep the trajectory figure in the SI.

Line 343. This tendency was observed in Jin et al. 2017 and Figure 6 in https://acp.copernicus.org/articles/23/1963/2023/

Response:

We have added the citation for this result (page 12, lines 452-454):
"The surface FNR_{sec} thresholds were generally lower than the column FNR_{sec}. The finding of higher column FNR is consistent with previous studies (Jin et al., 2017; Souri et al., 2023a)."

Figure 7. What caused a significant drop in June in Sapporo?

Response:

The authors thank the reviewer for pointing this out. Figure 7 in the previous version shows the tropospheric column FNR derived from Pandora direct-sun measurements. The HCHO direct-sun data have significant retrieval error budget. Sky-scan observations provide a more robust HCHO signal (Zhao et al., 2025). Therefore, we have revised the column FNR using sky-scan measurements instead of direct-sun data (Fig. S10 in the SI).

On MAX-DOAS plots: it may be worth adding AKs to show where the sensitivity of the retrieval to the absorber is the largest.

Response:

We directly used the L2 (Level 2) Air-Ratio Sky Algorithm developed by Elena Spinei and the Pandora Global Network (Cede et al., 2021). The data were obtained from the PGN (https://www.pandonia-global-network.org/home/documents/pgn-data/). Unfortunately, we cannot assess to the averaging kernels from the Pandora sky-scan.

According to the averaging kernel using HeiPro retrieval algorithm developed by Alwarda et al. (2025). The vertical sensitivity of the retrievals depends on the elevation viewing angle of Pandora and vertical distributions of atmospheric components. Viewing angle of 1 degree is sensitive within 100 m near the surface, 3 degree is sensitive to layer of approximately 500 m.

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