Point-to-point responses

We appreciate the reviewers for their valuable and constructive comments, which are very helpful for the improvement of the manuscript. We have revised the manuscript carefully according to the reviewers' comments. We have addressed the reviewers' comments on a point-to-point basis as below for consideration, where the reviewers' comments are cited in ***black****, and the responses are in* ***blue****.*

This study presents a dataset of vertical profiles of HONO and O_3 derived from the China Ground-based Hyperspectral Vertical Remote Sensing Observation Network. The dataset covers 22 representative sites, featuring an extended temporal span and broad spatial coverage. This dataset fills a critical observational gap and holds significant value for investigating boundary layer photochemical processes and improving our understanding of tropospheric oxidative capacity. The manuscript is well structured, with detailed methodological descriptions, comprehensive result presentation, and thorough data validation, and overall represents a high-quality data paper. However, there remains room for improvement in terms of clarity of expression, logical rigor, completeness of methodological details, and adherence to formatting conventions. Specific comments and suggestions are provided below to further enhance the quality and readability of the manuscript.

1. Line 23: The phrase “primary source” is somewhat absolute. OH production involves multiple pathways; consider revising to “one of the primary sources” or specifying conditions (e.g., polluted boundary layer).

Re: Thank you for your comments. We completely agree with your opinion. Although HONO photolysis is indeed the main source of OH under specific conditions (e.g., early morning in a polluted boundary layer), considering that there are multiple pathways for OH production in the atmosphere, such as O_3 photolysis and VOC photolysis, using “primary source” is indeed too absolute. We have revised the sentence in the revised manuscript to: “Photolysis of HONO and O_3 in the troposphere is one of the primary sources of OH radical and a fundamental control on atmospheric oxidative capacity.”

2. Lines 57-58: The statement “ozone (O_3)… has continued to increase” is overly absolute. It is recommended to add qualifiers such as “in some regions” or “on average” to more accurately reflect the complex trends of ozone pollution in China.

Re: Thank you for your comments. Thank you for the correction. The trend of near-surface ozone in China has significant regional heterogeneity; for example, some coastal areas or regions with significant $PM_{2.5}$ reductions may show different trends. The original expression was indeed too general. We have modified it to: “In contrast, ozone (O_3)—a secondary pollutant and a major atmospheric oxidant—has continued to

increase on average in economically developed regions such as the Beijing–Tianjin–Hebei area…", to more accurately reflect scientific facts.

3. Lines 97-98: The word "pronounced" in "preclude resolving the pronounced diurnal variability" is somewhat subjective. It is suggested to replace it with more objective terms like "strong" or "fine-scale".

Re: Thank you for your comments. We agree with your suggestion. The word "pronounced" carries a relatively strong subjective tone, while "fine-scale" more accurately describes the shortcomings of satellite observations in terms of spatiotemporal resolution. We have revised the sentence to: "…preclude resolving the fine-scale diurnal variability and vertical structure of O_3 and HONO".

4. Lines 117-118: In "to quantify the altitude-dependent production of OH radicals," the term "altitude-dependent" might be ambiguous (dependent on altitude vs. varying with altitude). It is recommended to change it to "altitude-resolved" or "vertical variations in."

Re: Thank you for your comments. Your understanding is very precise. "Altitude-dependent" in mathematics or physics might be misunderstood as "dependent on altitude" (i.e., a certain parameterization scheme depends on the altitude variable), whereas our purpose is to describe the "resolution of production rates varying with altitude." We have adopted your suggestion and modified it to: "The primary objective is to resolve the vertical structure of HONO and O_3 and to quantify the altitude-resolved production of OH radicals from their photolysis."

5. Lines 122-124: The order of region names "North, East, South, Central, and Southwest China" is not entirely consistent with the order presented later in Table 1 and the main text (e.g., Southwest China appears after South China in Table 1). It is recommended to unify the listing order of region names throughout the article.

Re: Thank you for your comments. Thank you very much for pointing out this inconsistency. Upon verification, the order in Table 1 is North, East, Southwest, South, Central. To maintain consistency throughout the manuscript, we have unified the order of mentioning these five regions throughout the text, adjusting them to the order consistent with Table 1: North, East, Southwest, South, and Central China.

6. Lines 146-148: In "stations at the Chinese Academy of Meteorological Sciences (CAMS1, CAMS2) and the University of Chinese Academy of Sciences (UCAS), located in and around Beijing…", the phrase "in and around Beijing" is slightly ambiguous, as both stations are within the municipality of Beijing. It is recommended to change it to "located within Beijing".

Re: Thank you for your comments. We agree with your revision suggestion. The CAMS site is located in Haidian District, and the UCAS site is in Huairou District; both are within the administrative jurisdiction of Beijing. Using "in and around" easily causes misunderstanding. We have modified it to: "In North China, stations at the Chinese Academy of Meteorological Sciences (CAMS1, CAMS2) and the University of Chinese Academy of Sciences (UCAS), located within Beijing (~100–120 m a.s.l.), characterize the heavily urbanized and industrialized core of the Beijing–Tianjin–Hebei megacity cluster."

7. Lines 156-157: The sentence "The summit of Mount Tai (TS; 1,500 m a.s.l.) offers vertical profiles under clean, high-altitude background conditions." is not entirely accurate, as TS, being a major tourist destination, is subject to tourism activities and regional transport influences and is not pristine. It is recommended to change "clean" to "relatively clean background conditions".

Re: Thank you for your comments. We completely agree with your view. As a famous scenic area, Mount Tai is indeed affected by tourism emissions and surrounding regional transport, and is by no means absolutely clean. We have changed "clean" to "relatively clean" in the manuscript to more rigorously describe its attributes as a regional background site.

8. Lines 186-200: Are the specifications of the instruments employed at each of the 22 sites consistent?

Re: Thank you for your comments. This is a very critical technical question. The hardware consistency of the instruments is the physical foundation for ensuring the comparability of multi-site data. We have explicitly supplemented and clarified in the revised manuscript: The spectrometers and telescopes used at all 22 sites in this network are indeed of the exact same brand and model. Specifically, all sites were equipped with high-resolution fiber-optic spectrometers of the same model and telescope systems based on the same optical design (right-angle prism + plano-convex lens). Before deployment to each site, all instruments underwent unified wavelength calibration and radiometric calibration in the laboratory to ensure consistency among the instruments.

9.Lines 193-194: Following "All sites employed an identical elevation scanning sequence…," it is recommended to add a brief explanation of the scientific rationale or optimization considerations for selecting these specific elevation angles (e.g., 1°, 2°, …90°).

Re: Thank you for your comments. This is a good suggestion; adding scientific rationale helps readers understand the observation design. We selected this set of angle sequences (especially the dense setting at low elevations) based on the following scientific considerations: the optical path at low elevations (1°-10°) mainly travels through the lower boundary layer and is extremely sensitive to the vertical gradient of

near-surface pollutants; using logarithmic or quasi-logarithmic intervals (e.g., 1, 2, 3, 4, 5, 6, 8, 10) can more effectively capture the characteristic of pollutant concentrations decreasing exponentially with altitude within the boundary layer; 15° and 30° are used to cover the middle and upper boundary layer; the 90° zenith angle serves as a reference to eliminate contributions from the stratosphere and upper atmosphere. In previous studies, this was the standard elevation sequence used; we have cited previous literature as a reference in the revised manuscript. Modified to: "All sites employed an identical elevation scanning sequence of 1°, 2°, 3°, 4°, 5°, 6°, 8°, 10°, 15°, 30°, and 90°(Liu et al., 2022; Xing et al., 2021, 2023)."

10. Line 204: In the spectral retrieval section, please specify the version number of the QDOAS software used.

Re: Thank you for your comments. Thank you to the reviewer for the reminder. The precision of the software version number is crucial for the reproducibility of the study. The version of the QDOAS software we used in the retrieval is QDOAS 3.2. We have added this information in Section 2.3 of the revised manuscript: "Ultraviolet–visible spectra measured by the ground-based instruments were analysed with the QDOAS software (version 3.2) developed by BIRA-IASB."

11. Lines 206-208: Please clarify whether the zenith reference spectrum is the 90° spectrum from the same scanning sequence, or a daily average or zenith spectrum from a specific time period.

Re: Thank you for your comments. The zenith reference spectrum we use is the 90° elevation spectrum from the same scanning sequence as the low-elevation spectrum to be retrieved. This is a standard practice in MAX-DOAS retrieval, known as the "Sequential Reference method." Its advantage lies in: being able to maximally cancel out changes in solar radiation intensity, instrument response drift, and slow-varying absorption contributions from the upper atmosphere (stratosphere and above) within a short period (approximately a 12-minute scanning cycle). If the time interval between the zenith spectrum and the low-elevation spectrum within the same sequence does not exceed a few minutes, these "slow-changing" factors can be considered identical, and thus are effectively eliminated during the differencing process, highlighting the differential absorption signals brought by the lower atmosphere. We do not use an averaged zenith spectrum for the day or a zenith spectrum from a specific time interval, as that would introduce uncertainties due to changes in solar zenith angle and optical path differences. We have clarified this in the revised manuscript: "For each elevation scan, the zenith spectrum (90° elevation) acquired within the same scanning sequence was used as the reference…"

12. Lines 213-214: In "using a fifth-order polynomial, allowing accurate separation of narrow-band

molecular absorption," the subject of "allowing" is unclear. It is recommended to change it to "This allows for the accurate separation…" or merge it with the preceding sentence.

Re: Thank you for your comments. Thank you for pointing out the grammatical issue. We have modified it according to your suggestion: "Broadband spectral structures were represented and removed using a fifth-order polynomial. This allows for the accurate separation of narrow-band molecular absorption."

13. Lines 215-217: After "Strict quality control was applied: only retrievals with a root-mean-square (RMS) fitting residual below 1×10^{-3} were retained," it is recommended to add the basis or reference for setting this specific RMS threshold.

Re: Thank you for your comments. This threshold (1×10^{-3}) is derived from common practices in the field of DOAS retrieval and the experience of extensive previous research. Excessively high RMS usually means the fitting process is severely affected by cloud interference, extreme aerosol scattering, or instrument noise, leading to unreliable retrieval results. Regarding the determination of this threshold, it has been explained in many previous studies; we cite previous literature here to explain this issue, and there are many related references (Hu et al., 2024; Ji et al., 2023; Liu et al., 2022, 2023; Song et al., 2023; Wang et al., 2019; Xing et al., 2017, 2019, 2021, 2023, 2024; Xu et al., 2021; Zou et al., 2025).

14. Lines 307-309: In "without a persistent mid-level enhancement," the word "persistent" may not be accurate, as the dataset presents a multi-year mean state. It is suggested to change it to "without a distinct mid-level enhancement."

Re: Thank you for your comments. Your logic is very rigorous. Multi-year averaged data smooths out transient disturbances, so "persistent" is indeed inapplicable; changing it to "distinct" better describes the characteristics under the averaged state. We have modified it to: "…without a distinct mid-level enhancement."

15. Lines 334-336: In the sentence "This pattern reflects nocturnal accumulation driven by heterogeneous conversion… and after sunrise…," the two clauses connected by "and" are at different logical levels. It is recommended to separate them with a semicolon or restructure the sentence.

Re: Thank you for your comments. We agree with your revision opinion. The first half describes the nighttime mechanism, and the second half describes the daytime mechanism; connecting them with "and" is indeed logically unclear. We have reconstructed the sentence using a semicolon: "This pattern reflects nocturnal accumulation driven by heterogeneous conversion of NO_2 on aerosol and ground surfaces (Liu et al., 2022; Xing et al., 2023; Xuan et al., 2025); after sunrise, enhanced solar radiation leads to the release and photochemical processing of HONO; meanwhile, morning rush-hour emissions of NO_2 and VOCs

further promote HONO formation (Garcia-Nieto et al., 2018; Zhang et al., 2025)."

16. Lines 347-348: For the statement "Sites with dense vegetation or agricultural land use (LA and CF) may receive contributions from biogenic VOC-related chemistry," it is recommended to provide a reference for research on the influence of biogenic VOCs on HONO formation.

Re: Thank you for your comments. Thank you for the reminder. Although vegetation itself does not directly emit large amounts of HONO, soil NOx emissions associated with vegetation and heterogeneous reactions on leaf surfaces may contribute to HONO. We have added relevant references to support this statement: "Sites with dense vegetation or agricultural land use (LA and CF) may receive contributions from biogenic VOC-related chemistry, but the overall pattern still features a subdued morning maximum (Liang et al., 2017; Ryan et al., 2018; Xue et al., 2021; Ye et al., 2023)."

17. Lines 411-412: In the sentence "The pronounced O3 enhancements observed at 3–4 km at sites such as CQ, GZ_TM and SUIST are therefore likely linked to…," "SUIST" should be "SUST" (the site for Southern University of Science and Technology). Please correct this.

Re: Thank you for your comments. You are absolutely correct; this is indeed a typo. "SUIST" does not exist; the correct site abbreviation should be "SUST" (Southern University of Science and Technology). We have corrected "SUIST" to "SUST" in the text and conducted a second check of all site abbreviations throughout the manuscript, confirming no other similar errors.

18. Lines 476: The phrase "using in situ concentration profiles" is incorrect, as MAX-DOAS observations are remote sensing retrievals, not "in situ" measurements. It is recommended to change it to "using retrieved concentration profiles" or simply "using concentration profiles".

Re: Thank you for your comments. What you pointed out is very correct; MAX-DOAS belongs to remote sensing retrieval technology, and using "in situ" does cause conceptual confusion. We have modified it to: "To quantify the AOC at each site, we evaluated altitude-resolved OH production from HONO and O_3 using retrieved profiles combined with photolysis frequencies calculated by the TUV model."

19. Lines 504-505: The manuscript states "J(HONO) exhibits a pronounced diurnal cycle, increasing during the morning (08:00–10:00) with rising solar radiation." In fact, J values begin to rise immediately after sunrise. The fastest increase may occur during 08:00–10:00, but the peak is usually around noon. It is recommended to change it to "increasing after sunrise" for accuracy.

Re: Thank you for your comments. We agree with your scientific judgment. J(HONO) varies with solar

zenith angle and begins to increase immediately after sunrise. The time limitation in the original text was too narrow. We have modified it to: "At the North China sites (CAMS1, CAMS2, UCAS, WD, SJZ_LC, SXU), J(HONO) exhibits a pronounced diurnal cycle, increasing after sunrise with rising solar radiation, peaking at midday (12:00–14:00) at 0.0020–0.0025 s^{-1}, and decreasing in the afternoon (14:00–18:00)."

20. Line 543: The manuscript states "reaching 1.0×10^{4}–5.5×10^{4} ppb·s^{-1}". For OH production P(OH) = J(HONO) * [HONO], if J is taken as ~10^{-3} s^{-1} and HONO as 0.3–0.5 ppb, then P(OH) should be approximately 3×10^{-4}–5×10^{-4} ppb·s^{-1}. The value in the text is 8 orders of magnitude larger, indicating a severe typo. Please verify and correct this.

Re: Thank you for your comments. We are extremely grateful that you keenly spotted this severe numerical error. Your judgment is completely accurate; a negative sign was missing from the exponent in the original manuscript, resulting in the value being magnified by 8 orders of magnitude. Verifying by physical common sense: for OH production rate P(OH)=J(HONO) ×[HONO]. Typically, J is on the order of 10^{-3} s^{-1}, and near-surface HONO concentration is about 0.3–0.5 ppb, so the reasonable order of magnitude for P(OH) should be 10^{-4} ppb·s^{-1}. The10^{4} in the original manuscript was purely a typo. We have recalculated and corrected all values involving OH production rates throughout the manuscript, correcting the value here to "1.0×10^{-4}–5.5×10^{-4} ppb·s^{-1}". We sincerely apologize for the inconvenience this elementary error caused to the reviewer during reading.

21. Line 549: The manuscript states "exceeding 3.0×104 ppb s-1". This also contains an order-of-magnitude error; it should be "exceeding 3.0×10-4 ppb s-1" or a similar magnitude.

Re: Thank you for your comments. As above, this is also a severe error in the exponent sign. We have corrected it to: "…exceeding 3.0×10^{-4} ppb·s^{-1}".

22. Lines 615-616: The manuscript reads "at the 22 sites ，and seasonal mean diurnal". A Chinese full-width comma "， " is used here; it should be changed to an English half-width comma. The manuscript reads "presented in Figures S29–S32..". There are two periods at the end of the sentence; it should be changed to one.

Re: Thank you for your comments. Thank you for pointing out the formatting issues. We have replaced the Chinese full-width comma "， " with an English half-width comma ",", and corrected the double period at the end of the sentence to a single period.

23. Line 653: Regarding the comparison with CNEMC surface station data, please detail the methodology

used to select the CNEMC stations.

Re: Thank you for your comments. We briefly mentioned in the manuscript "Site pairs were selected following the spatial representativeness criteria of Song et al.(2023)", but the description can indeed be more specific. The selection of CNEMC sites followed these principles: (1) Distance proximity: Prioritize selecting the CNEMC national control station geographically closest to our hyperspectral site. We set a maximum distance threshold (usually within 10–15 km, see Table S1 in the Supplement for details) to ensure that the two sites are in a similar urban/regional environmental background, reducing the impact of spatial heterogeneity. (2) Environmental consistency: Referring to the method of Song et al.(2023), we looked not only at distance but also used satellite remote sensing data (such as land-use type, nighttime light index) and meteorological data to confirm that the candidate CNEMC site and our site are comparable in terms of land-use type (e.g., both urban areas or both suburban areas) and built-up area density. (3) Data availability: Ensure that the selected CNEMC site has continuous, valid O_3 surface concentration data during the comparison period (2021–2024).

We have expanded the sentence in the revised manuscript to: "Site pairs were selected following the spatial representativeness criteria of Song et al.(2023). Specifically, we prioritized the nearest CNEMC station within a maximum distance of ~10–15 km (detailed in Table S1) and verified environmental consistency between the paired sites using land-use and satellite-derived products, ensuring that both sites sampled comparable urban or suburban atmospheric conditions."

24. Line 660: The manuscript states "confirm the reliability of the dataset used in this study". It is recommended to add a note regarding HONO data validation, as the text only presents validation for O_3, whereas HONO is another critical data product.

Re: Thank you for your comments. The reviewer points out an objective limitation. Within the 22 sites and time period covered by this study, we lack on-site in-situ instruments (such as tower-based HONO analyzers, etc.) that are synchronized with MAX-DOAS observations and can be used for independent validation of HONO vertical profiles. This is also a common challenge currently faced in the global field of HONO vertical observation, because HONO is extremely unstable, in-situ measurement techniques are complex, and it is difficult to achieve vertical profiling. Therefore, we cannot perform direct validation like we did for O_3. However, we enhance our confidence in the reliability of the HONO data through the following indirect methods: (1) Validation of the retrieval method: The DOAS retrieval settings and OEM profile retrieval framework we used have been applied to HONO retrieval in multiple previous studies and have been fully validated through testing on simulated data and comparison with samples of known concentrations (e.g., Liu et al., 2022; Song et al., 2023; Xing et al., 2019, 2021, 2023). (2) Consistency with O_3 retrieval results:

HONO and O_3 use the same observation geometry, spectral processing workflow, and retrieval algorithm. The O_3 retrieval results have been well validated through comparison with TROPOMI and CNEMC (R=0.62, 0.66), which indirectly supports the reliability of the entire retrieval workflow (including HONO) when processing real atmospheric spectra. (3) Rationality of HONO vertical and diurnal variation characteristics: As shown in the results in the text, the HONO profiles we obtained (high near the surface, decreasing sharply with altitude) and diurnal variation (high in early morning, low in afternoon) are completely consistent in pattern with a large number of existing HONO observation studies based on different methods (e.g., Meng et al., 2020; Xuan et al., 2024, etc.), without showing anomalous physical signals.

25. Line 681-682: The statement "represents one of the most extensive collections in China" should be verified. It is recommended to change it to "one of the most extensive publicly available collections" for greater rigor.

Re: Thank you for your comments. We accept your suggestion. There are indeed other non-public long-term observation data (e.g., internal data from certain research projects), so limiting it to "publicly available" is more rigorous. We have modified it to: "The dataset spans 22 representative sites across North, East, Central, South, and Southwest China, covering a wide range of climatic regimes and surface types, and represents one of the most extensive publicly available collections in China in terms of spatial coverage and vertical resolution of photochemical parameters relevant to OH precursors."

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