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 manuscript presents and publicly releases a valuable dataset derived from the Chinese Hyperspectral Vertical Remote Sensing Network, featuring vertical profiles of O_3 and HONO acquired from 22 representative sites between 2021 and 2024. This dataset fills a critical gap in the long-term, vertical observations of key photochemical species over China, holding significant scientific value for understanding the evolution of tropospheric oxidative capacity. Overall, it represents a high-quality data paper. However, several details in the manuscript still require further refinement to enhance the overall quality and readability of the paper.

1. Lines 85-86: The word "generally" in "but generally lacks observations of key photochemical precursors such as HONO and volatile organic compounds (VOCs)" could be ambiguous. It is suggested to change to "but lacks observations of key photochemical precursors such as HONO and volatile organic compounds (VOCs)".

Re: Thank you for your comments. We completely agree with your revision suggestion. The word "generally" is indeed ambiguous and might mislead readers into thinking that some sites have these observation capabilities, whereas in reality, current CNEMC sites generally do not include routine monitoring of HONO and VOCs. To make the expression more accurate, objective, and unambiguous, we have revised the original manuscript to: "…but lacks observations of key photochemical precursors such as HONO and volatile organic compounds (VOCs)…" This revision makes the meaning more definitive and aligns with the actual situation.

2. Lines 120-121: "the contribution of HONO photolysis to boundary-layer OH" could be changed to "the contribution of HONO photolysis to the boundary-layer OH budget" for greater precision.

Re: Thank you for your comments. In photochemical research, "OH budget" is a very rigorous and standard term, specifically referring to the quantitative balance between the sources and sinks of OH radicals. Using "boundary-layer OH budget" more accurately conveys our scientific objective of evaluating the proportion of HONO photolysis in the OH generation process compared to simply using "boundary-layer OH". We have modified it according to your suggestion. The original text has been modified to: "the contribution of

HONO photolysis to the boundary-layer OH budget".

3. Lines 186-187: The manuscript mentions a "standardized instrument configuration". Please explicitly state whether the spectrometers and telescopes used at all 22 sites are of the exact same brand and model.

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.

4. Line 194: The elevation scanning sequence includes 1°, 2°, …, 90°. Low elevation signals (e.g., 1°) are susceptible to obstructions, during the processing, were strict obstruction checks performed for low-elevation observations at all sites? If obstructions were present, were the relevant data excluded?

Re: Thank you for your comments. Thank you for raising this question, which shows rich practical experience. Low-elevation observations are indeed highly susceptible to obstruction by buildings, trees, or terrain, leading to abnormal optical paths and subsequently affecting retrieval results. We implemented a strict obstruction checking procedure in our data processing workflow: (1) Pre-deployment assessment: During the initial site construction, we used topographic maps and on-site inspections to ensure there were no obvious obstructions in the telescope's field of view (especially in low-elevation directions). (2) Software auto-screening: In routine data processing, we analyzed the variation in radiation intensity of low-elevation (especially 1° and 2°) spectra. If we found drastic jumps or abnormal attenuation in spectral intensity for non-meteorological reasons (usually manifesting as scattered light intensity lower than theoretically expected), the system would automatically flag it as "suspected obstruction." (3) Manual review and exclusion: For data flagged as such, we conducted manual review in conjunction with the site's surveillance photos and logs. Once an obstruction was confirmed, the low-elevation data from that scanning cycle were strictly excluded and did not participate in subsequent DOAS fitting and profile retrieval.

5. Line 195: "The integration time at each elevation was 1 min" refers to the integration time for observation at each elevation angle. It is suggested to change to "The integration time at each elevation angle was 1 min".

Re: Thank you for your comments. We agree with your suggestion. Adding the word "angle" eliminates any potential ambiguity between "elevation" and "azimuth" making the expression more rigorous. We have modified it to: "The integration time at each elevation angle was 1 min".

6. Line 197: "The instruments also operated at night to record dark current…". This phrasing might mislead readers into thinking the instruments were conducting routine atmospheric composition observations at night. It is recommended to explicitly state that nighttime operation was strictly for instrument calibration and did not produce scientific data.

Re: Thank you for your comments. Thank you very much for pointing out this potential pitfall in the wording. MAX-DOAS technology relies on scattered sunlight; it cannot retrieve atmospheric compositions at night, and the original phrasing might indeed mislead non-specialist readers. We have restructured the original sentence to explicitly distinguish between "observation" and "calibration": "For instrument calibration purposes only, the instruments operated at night to record dark current and electronic offsets. No scientific atmospheric data were derived from nighttime measurements." With this revision, the logic is very clear and avoids misleading readers.

7. Line 207: The manuscript mentioned that the zenith spectrum (90°) was used as the reference. Please clarify whether the reference spectrum is from the same scanning cycle or if a fixed reference was used.

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.

8. Line 268: In "More than ~85% of the sites operated for over one year", "~" indicates an approximation, which is slightly redundant when used with "more than". It is suggested to change to "Over 85% of the sites operated for more than one year".

Re: Thank you for your comments. Your logic is very rigorous. "More than" itself already implies a certain degree of approximation or a lower limit; superimposing the tilde "~" indeed appears semantically redundant and unprofessional. We have adopted your suggestion and modified it to: "Over 85% of the sites operated for more than one year…", making the manuscript more concise and crisp.

9. Line 335: Please pay attention to the subscripts for NO2; it should be written as "NO_2". Please check the full manuscript to ensure all instances of NO_2 are written correctly.

Re: Thank you for your comments. We apologize for this formatting oversight. Standardized writing of chemical formulas is a basic requirement for scientific papers. We have conducted multiple thorough searches and replacements throughout the manuscript to ensure that all instances of "NO2" (including in the main text, figure captions, tables, etc.) have been correctly replaced with the subscripted format "NO_2". At the same time, we took this opportunity to check the formats of other chemical formulas (such as O_3, HONO, O_4, etc.) to ensure consistent and standardized formatting throughout the manuscript.

10. Line 412: "the pronounced O_3 enhancements observed at 3–4 km at sites such as CQ, GZ_TM and SUIST" – here "SUIST" should be "SUST". Please verify.

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.

11. Line 543: The manuscript reads "reaching 1.0×10^{4}–5.5×10^{4} ppb·s^{-1}". This should likely be 10^{-4}. It appears to be a typographical error. Please verify and correct.

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.

12. Line 572-573: "underscoring the near-surface confinement of both HONO and its photolytic OH production." "its photolytic OH production" could be changed to "OH production from its photolysis" for clarity.

Re: Thank you for your comments. We agree with your suggestion. The original phrasing "its photolytic OH production," where "photolytic" acts as an adjective modifying "OH production," is somewhat stiff and prone to ambiguity. Modifying it to "OH production from its photolysis" treats "photolysis" as a process, making the logical relationship clearer and the language more consistent with authentic scientific English expression habits. We have made this change.

13. Line 616: There are two periods in "Figures S29–S32..". One should be removed.

Re: Thank you for your comments. Thank you for carefully proofreading the punctuation. This is obviously a mistake operation during typesetting. We have corrected the double period at the end of the sentence to a single period.

14. Line 643: The validation section only presents the validation of O_3 data and does not cover the validation of HONO data. Is this because there is no verifiable data for HONO, or is it due to other reasons?

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.

15. Lines 652-655: “Song et al. (2023b)” is cited as the criterion for site selection. For readers unfamiliar with this work, it is recommended to briefly summarize the main content of the selection criteria so that readers can understand the spatial representativeness of the validation data.

Re: Thank you for your comments. This is a very good suggestion, helping to enhance the self-consistency and readability of the paper. Simply citing a literature without explaining the criteria would indeed cause confusion for readers when assessing the reliability of the validation results. The spatial representativeness matching criteria proposed in Song et al.(2023)mainly include the following core dimensions: (1) Distance threshold: Prioritize selecting the CNEMC national control station nearest to the MAX-DOAS site, usually requiring a distance within a few to a dozen kilometers (the specific threshold depends on the site environment, avoiding crossing large pollution source blocks). (2) Underlying surface consistency: Use satellite remote sensing data (such as land-use type maps, nighttime light data) to confirm that the candidate CNEMC station and the MAX-DOAS site are in the same type of underlying surface environment (e.g., both urban core areas, or both suburban transition zones), avoiding erroneous matching like “remote sensing in the city, ground station in the village.” (3) Exclusion of interference sources: Ensure there are no large point source emissions (such as chemical plants, thermal power plants) between the two sites, to guarantee that the air masses observed by both are homologous.

References

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