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 utilizes ground-based hyperspectral stereoscopic remote sensing technology to conduct an in-depth investigation of the vertical structure and seasonal evolution of AOC over typical urban (AHU) and rural (CF) sites in the Yangtze-Huai River Basin during the spring and summer of 2023. By quantifying the vertical contributions of HONO, HCHO, and O_3 photolysis to OH radical production, the authors innovatively reveal a distinct vertical stratification of oxidation mechanisms within the boundary layer. The study also identifies an anomalous HONO heterogeneous conversion layer at approximately 2.4 km altitude during summer in the urban area and comprehensively elucidates the spatial decoupling of oxidation driving mechanisms between urban and rural environments. These findings challenge the conventional assumption that heterogeneous reactions are confined to the near-surface atmosphere, provide critical vertical observational constraints for three-dimensional atmospheric chemistry models, and offer a mechanistic explanation for the limited effectiveness of current surface-based NOx mitigation strategies. The research is rigorous, innovative, and of significant scientific value, making it highly suitable for publication in ACP.

1. Lines 117-120: The AHU and CF sites were selected with azimuth angles of 353° and 209°, respectively. Please clarify whether there are any physical obstructions at low elevation angles for these two azimuth directions, and briefly discuss the spatial representativeness of these two sites.

Re**:** Thank you for your comments. Regarding the setting of azimuth angles and the spatial representativeness of the observation sites, we have provided detailed supplementary explanations in the revised manuscript:

(1) Physical obstructions: During instrument installation, we surveyed the surrounding environment. The AHU instrument was deployed on the rooftop of a 25 m tall teaching building at Anhui University (azimuth angle 353°), while the CF instrument was installed on top of a 2 m high container in Changfeng County (azimuth angle 209°). Both azimuth directions point towards open areas, and there are no physical obstructions, such as tall buildings, mountains, or trees, within the field of view during low-elevation-angle observations (e.g., 1°, 2°). This ensures the authenticity of the atmospheric absorption signals acquired by the MAX-DOAS at low elevation angles.

(2) Spatial representativeness: The AHU site is located in a core commercial and traffic-intensive area of

Hefei, representing a typical complex urban environment. The CF site is approximately 49.2 km away from AHU in a straight line and is surrounded mainly by farmland and agricultural areas, representing a typical agricultural rural environment. The deployment of these urban-rural dual sites effectively captures the differences between anthropogenic sources (traffic and industrial emissions) and natural sources (biogenic emissions and soil release), demonstrating strong spatial representativeness for the urban-rural meteorological and chemical processes typical of urban agglomerations in the Yangtze-Huai River Basin.

2. Lines 118-119: For the descriptions "(AHU, 31.78°N, 117.20°E, a commercial and traffic intensive area)" and "(CF, 32.21°N, 117.18°E, a farm and agricultural area)", it is recommended to use semicolons to separate the parallel descriptive phrases within the parentheses for better readability.

***Re*:** Thank you for your comments. We appreciate the reviewer's careful reading and suggestion. In the revised manuscript, we have used semicolons to separate the parallel descriptive phrases within the parentheses, revising them to "(AHU, 31.78°N, 117.20°E; a commercial and traffic intensive area)" and "(CF, 32.21°N, 117.18°E; a farm and agricultural area)" to improve the readability of the article.

3. Lines 123-131: Differences in instrument models or configurations between the two sites could introduce systematic observational biases. Please briefly state in the methodology section whether the spectral resolutions (FWHM) of the instruments deployed at AHU and CF are identical.

***Re*:** Thank you for your comments. We deployed two ground-based hyperspectral remote sensing instruments of the exact same model at the AHU and CF sites. The core components (telescope and spectrometer) of both instruments are identically configured, and their spectral resolutions (FWHM) are identical. Prior to field observations, both instruments underwent strict laboratory mercury lamp wavelength calibration and radiometric calibration, ensuring consistent instrument performance between the two sites during the observation period, thereby excluding systematic observational biases caused by differences in instrument models or configurations. We have explicitly clarified this in Section "2.1 Vertical measurements" of the revised manuscript by adding the sentence: "The instrument configurations were identical at both sites".

4. Line 126: In the phrase "spectral range: UV: 296–408 nm, VIS: 420–565 nm", the repeated use of colons is somewhat redundant. It is recommended to revise this to "spectral range: UV (296–408 nm) and VIS (420–565 nm)".

***Re*:** Thank you for your comments. We have revised this expression in the revised manuscript to the

reviewer's suggested form, "spectral range: UV (296–408 nm) and VIS (420–565 nm)", making the text more concise and standard.

5. Line 146: The DOAS retrieval exclusively utilizes data with SZA < 75°. Please clarify the rationale behind selecting this specific solar zenith angle threshold.

***Re*:** Thank you for your comments. The selection of SZA < 75° as the data filtering threshold is primarily based on the following two scientific considerations:

(1) Stratospheric absorption interference: At large SZAs (especially > 75°), the optical path of sunlight through the atmosphere increases significantly, and the absorption by stratospheric ozone and other trace gases is drastically enhanced. This severely interferes with the differential absorption retrieval of tropospheric trace gases (such as HONO, HCHO, etc.).

(2) Signal-to-noise ratio and radiation intensity: When SZA > 75° (e.g., during early morning and late evening), solar radiation intensity weakens substantially, leading to a decrease in the spectral signal-to-noise ratio and affecting the stability of the DOAS fitting. Setting SZA < 75° effectively excludes data collected under long optical paths and low-light conditions in the early morning and late evening, ensuring the quality and reliability of tropospheric retrieval products. This threshold setting has been widely adopted as a standard quality control procedure in the MAX-DOAS remote sensing field (Hong et al., 2018; Song et al., 2023a; Tian et al., 2019; Xing et al., 2023).

6. Lines 147-150: The spectral fitting strictly filters out data with RMS > 1×10^{-3}. What is the justification for choosing this specific threshold? Please compare this criterion with similar MAX-DOAS studies to demonstrate the rationality of this data filtering standard.

***Re*:** Thank you for your comments. Setting the RMS of the spectral fit threshold to 1×10^{-3} is a widely accepted data quality control standard in MAX-DOAS retrievals. In the retrieval of weakly absorbing gases (especially HONO and HCHO), an RMS > 1×10^{-3} typically indicates the presence of large random noise in the spectrum, uncorrected instrument wavelength drift, or unidentified interfering absorption structures. Setting the RMS threshold to 1×10^{-3} can effectively filter out low-quality spectra and retain fitting data with a high signal-to-noise ratio. For example, in the CINDI-2 international intercomparison campaign and related literature, this threshold was used as a standard reference to evaluate the stability of retrieval algorithms (Kreher et al., 2020; Wang et al., 2020). In addition, our previous MAX-DOAS observation studies in the Yangtze River Delta and other regions also adopted the same standard and obtained reliable products (Xing et al., 2017, 2019, 2023, 2024).

7. Lines 136-150: Regarding the DOAS fitting process, please elaborate on how the reference spectrum was selected and detail the procedures for dark current and noise subtraction.

***Re*:** Thank you for your comments. Regarding the details of the DOAS retrieval process, we have provided more detailed supplements in Section "2.2 Data analysis" of the revised manuscript:

(1) Selection of the reference spectrum: The direct/scattered solar spectrum measured in the zenith direction at noon (e.g., 11:00–13:00, small SZA) with high atmospheric transparency was selected as the daily reference spectrum. By using the same-day reference spectrum, the influences of the spectral structure features of the instrument itself and the solar Fraunhofer lines can be minimized.

(2) Dark current and noise subtraction: Our instrument measured observation spectra during the day and dark background spectra at night. During the data processing stage, the measured radiance spectra were subtracted by the synchronously measured dark current spectra and intensity offset, thereby eliminating the baseline offset caused by detector thermal noise and the readout circuit. The above procedures comply with standard DOAS data processing protocols (Kreher et al., 2020; Wang et al., 2020).

8. Lines 169-171: The total error analysis indicates retrieval errors of 24%, 33%, and 22% for HONO, HCHO, and O_3, respectively. It is recommended to provide a breakdown of the contribution from each error source (e.g., smoothing error, noise error) to clearly identify the primary sources of uncertainty.

***Re*:** Thank you for your comments. We have detailed the breakdown contributions from each error source in Section S1 Error analysis of the Supplementary Information. We summarize them below and direct readers to the Supplementary Information in the main text:

(1) HONO: The total error is 24%, with smoothing and noise errors being the primary contributors at 16%; uncertainties related to aerosol retrieval account for 5%; absorption cross-section uncertainties and temperature-dependent errors account for the remaining ~3%.

(2) HCHO: The total error is 33%, mainly originating from smoothing and noise errors (27%), aerosol retrieval errors (5%), and absorption cross-section-related errors (1%).

(3) O_3: The total error is 22%, of which temperature-dependent absorption cross-section errors contribute the most (13%), smoothing and noise errors account for 8%, and other algorithmic and aerosol errors account for 1%. The above data indicate that for weakly absorbing pollutants (HONO, HCHO), smoothing and noise errors caused by limited detection sensitivity are the dominant uncertainties; whereas for O_3, temperature-dependent cross-section errors account for a larger proportion.

9. Lines 277-280: The explanation for the high near-surface HONO/NO_2 ratio at the rural site in summer attributes the phenomenon to agricultural fertilization and soil emissions. Please provide records of

agricultural activities (e.g., fertilization dates and types) around the CF site during the observation period to substantiate this conclusion.

***Re*:** Thank you for your comments. During this observation period (March to August 2023), the surroundings of the CF site were a typical rice-wheat rotation and facility agricultural area. According to local agrometeorological records and communication with local farmers during the observation period, there were concentrated fertilization activities during the wheat jointing and rice seedling stages in spring (April–May), and heavy application of base fertilizers for rice in summer (June–August), primarily urea and compound fertilizers. The abundant precipitation and high temperatures in summer led to a sharp increase in soil moisture and microbial activity, significantly promoting the release of soil nitrite and HONO emissions. This is well corroborated by the phenomenon that the near-surface $C_{(HONO)}$ at the CF site exhibited extreme peaks in April, June, and August. Numerous field observations and laboratory studies have confirmed that soil release after agricultural fertilization is an important unknown source of atmospheric HONO (Song et al., 2023b; Xue et al., 2021). We have added the above explanation of the agricultural activity background in the revised manuscript.

10. Lines 343-346: The ratio of secondary HCHO to NO_2 (FNR) is used to determine O_3 formation sensitivity. Please explain the methodology used to effectively distinguish between primary and secondary HCHO.

***Re*:** Thank you for your comments. Regarding the separation of primary and secondary sources of formaldehyde (HCHO), we adopted a well-established multiple linear regression method in Section S3 "Separation of Primary and Secondary Sources of HCHO" of the Supplementary Information (Hua et al., 2023; Li et al., 2024a; Lin et al., 2022). The specific methodology is as follows:

Ground-level CO and Ox (O_3+NO_2) serve as tracers to isolate primary and secondary HCHO sources via a multiple linear regression framework (Hua et al., 2023; Li et al., 2024a; Lin et al., 2022)。

$$[HCHO] = \beta_0 + \beta_1 \times [CO] + \beta_2 \times [O_X] \quad \text{(S1)}$$

In this equation, [HCHO], [CO], and [O_X] denote the measured concentrations of HCHO, CO, and O_X, respectively. HCHO data are acquired from ground-based hyperspectral observations, whereas CO and Ox data originate from national monitoring networks. The coefficients β_0, β_1, and β_2 are derived from the regression fit: β_0 represents the background HCHO concentration, β_1 quantifies the emission ratio of HCHO to CO (primary source), and β_2 reflects the photochemically generated fraction of HCHO (secondary source).

The relative contributions of HCHO's background (P_{bg}), primary (P_{pri}), and secondary (P_{sec}) sources are then calculated as:

$$P_{bg} = \frac{\beta_0}{\beta_0 + \beta_1 \times [CO] + \beta_2 \times [O_X]} \times 100\% \quad \text{(S2)}$$

$$P_{pri} = \frac{\beta_1 \times [CO]}{\beta_0 + \beta_1 \times [CO] + \beta_2 \times [O_X]} \times 100\% \quad \text{(S3)}$$

$$P_{sec} = \frac{\beta_2 \times [O_X]}{\beta_0 + \beta_1 \times [CO] + \beta_2 \times [O_X]} \times 100\% \quad \text{(S4)}$$

11. Lines 347-348: In the O_3 formation sensitivity analysis, a transition zone is defined based on the intersection of the S_{HCHO} and S_{NO2} slopes. Please discuss the advantages of using this slope-intersection method from linear fitting when processing highly scattered vertical observation data.

***Re*:** Thank you for your comments. Using the slope intersection method of S_{HCHO} and S_{NO2} to define the transition regime of O_3 production sensitivity has significant advantages when processing vertical observation data with high dispersion: First, due to the high complexity of physicochemical processes such as meteorological conditions (e.g., temperature, humidity, and pressure), mixing layer dynamics, and precursor transport at various vertical layers, directly using single-point FNR ratios causes the thresholds to fluctuate drastically and unstably with altitude. Adopting the linear fitting slope method is essentially a macroscopic statistical regression of multi-day observational data within a certain vertical layer, which can effectively filter out short-term local noise and extract the steady-state characteristics of the regional photochemical system. Second, the slope intersection method adaptively identifies the chemical inflection point of sensitivity based on statistical characteristics, avoiding misjudgments caused by the subjective and empirical setting of uniform thresholds. The advantages and reliability of this method have been fully validated in our previous study on vertical ozone sensitivity diagnostics (Li et al., 2024b).

12. Lines 397-399: The calculation formulas for the OH production rate are consistent with mainstream methods. However, it is recommended to supplement an estimation of the contribution from the HO_2 + NO → OH reaction to evaluate its potential impact on total OH production.

***Re*:** Thank you for your comments. We fully agree that HO_2+NO → OH is an important daytime OH regeneration pathway in the atmosphere, which may have a certain contribution, particularly in boundary layer environments with active VOC oxidation and high NO concentrations. However, limited by the experimental design of this observation, the ground-based hyperspectral remote sensing system deployed in this study cannot directly quantify the vertical concentration profile of HO_2. HO_2 is a short-lived radical with an extremely weak absorption cross-section, and current ground-based DOAS technology cannot yet achieve its direct retrieval (Mao et al., 2010; Tan et al., 2019). If a photochemical box model is used to indirectly

estimate HO_2 concentrations, it would require reliance on high-precision input parameters such as VOC species, OH radicals, and J values, which also lack vertical resolution observation constraints in this study. Forcibly estimating them would introduce large and difficult-to-quantify uncertainties. Therefore, to avoid introducing unreliable speculative results, this paper does not include the HO_2+NO pathway in the quantitative calculation of AOC. It should be noted that similar urban boundary layer observation studies have shown that under sufficient daytime light conditions, the contribution of the HO_2+NO pathway to total OH production is typically less than 10% (Dai et al., 2023). Thus, neglecting this pathway has a limited impact on the core conclusion of this paper, which reveals the "vertically stratified oxidation mechanism." In the future, we will combine techniques such as airborne or long-path DOAS to further supplement direct observations of HOx radicals to improve the vertical budget assessment of AOC.

13. Lines 451-455: The four columns in Figure 6 represent different sites and seasons. Due to significant magnitude differences, a unified color scale might obscure the diurnal variation characteristics in low-value regions. Please verify the appropriateness of the color scale settings; consider using a logarithmic scale or independent color scales if necessary.

***Re*:** Thank you for your comments. We have re-examined the color scale settings of Figure 6. The current color scales are not identical; a distinct color scale is used for each of the three components. This not only preserves the clear presentation of their respective vertical profiles and diurnal characteristics but also facilitates the readers in comparing the relative changes across different seasons at the same site.

14. Lines 457-468: Regarding the quantitative AOC metrics in formulas (6) and (7), only the photolysis pathways of HCHO, HONO, and O_3 are calculated. It must be explicitly stated that this does not represent the strictly defined total AOC (as it excludes pathways such as ozone reactions with alkenes and NO_3 radicals).

***Re*:** Thank you for your comments. The quantitative metric defined in our text indeed covers exclusively the photolysis processes of HCHO, HONO, and O_3. Strictly speaking, AOC should encompass the sum of the oxidation capacity of all oxidants (e.g., OH, O_3, NO_3, etc.) towards all reduced species (e.g., SO_2, NOx, VOCs, etc.) (Dai et al., 2023). As OH radicals contribute over 90% of the oxidation capacity during the daytime due to their extremely high activity, this paper adopts the aforementioned three major OH production pathways as a surrogate quantitative metric for daytime AOC. We have explicitly stated in Section "3.5 The Role of OH Radicals in Combined Air Pollution" of the revised manuscript: "We note that the quantitative AOC metric employed here is based exclusively on photolytic OH production from HCHO, HONO, and O_3. By excluding secondary pathways (e.g., alkene-O_3 oxidation and NO_3 radical reactions),

this metric constitutes a core representation of daytime photochemical oxidation potential, rather than a strictly defined total AOC." This qualification clarifies the scope of application of the conclusions in this study.

15. Lines 462-464: AOC is defined here as the daytime total OH production rate. It is recommended to add a brief discussion on nighttime AOC (e.g., the contribution of NO_3 radicals) to clearly define the scope of applicability for this study's conclusions.

***Re*:** Thank you for your comments. Nighttime atmospheric oxidation processes are indeed equally important, especially the oxidative degradation of volatile organic compounds (particularly alkenes and aromatics) by NO_3 radicals, as well as the heterogeneous hydrolysis of N_2O_5 to generate HNO_3. However, because the instruments deployed in this study rely on scattered solar spectra for retrieval, our observations are strictly limited to daytime periods (SZA < 75°) and cannot directly provide vertical profile constraints for nighttime NO_3 radicals and related photochemical precursors. Therefore, the conclusions of this paper are mainly applicable to explaining the stratified mechanism of photochemical oxidation within the daytime boundary layer. We have explicitly defined in lines 550-551 of the "4. Conclusion" section that this study primarily focuses on the daytime boundary layer, and that the assessment of nighttime oxidation capacity needs to rely on other observation methods, which constitutes an external limitation of this study.

16. Lines 495-500: Figure 8 reveals a strong correlation between AOC and aerosol loading (R = 0.88–0.93). Although the authors note that AOC promotes secondary aerosol formation, high aerosol concentrations simultaneously alter the radiation field, thereby inhibiting photolysis rates and AOC. It is suggested to add a qualitative description of this "feedback inhibition" effect of aerosols on AOC to provide a more comprehensive discussion of the causal relationship.

***Re*:** Thank you for your comments. There indeed exists not merely a unidirectional promotional relationship but a complex bidirectional feedback mechanism between AOC and aerosols. High aerosol concentrations significantly alter the boundary layer radiation field through scattering and absorption, leading to a decrease in photolysis frequencies (e.g., J[HONO], J[O_3]), thereby inhibiting further photochemical OH production. This "aerosol-AOC feedback inhibition effect" is crucial for the oxidative evolution during pollution accumulation periods (Ivatt et al., 2022). We have added a qualitative discussion of this mechanism in Section "3.5" of the revised manuscript, explicitly stating: "While strong AOC promotes secondary aerosol formation, leading to a robust positive correlation between AOC and aerosol loading, it is critical not to overlook that high aerosol concentrations can attenuate surface-reaching UV radiation. This attenuation reduces photolysis frequencies, thereby exerting a feedback inhibition on AOC. Such bidirectional feedback

represents a key factor driving the nonlinearity of boundary layer photochemistry during pollution episodes." This addition makes the discussion of causal relationships more comprehensive.

17. Lines 551-552: Regarding the limitations in spatial representativeness, it is recommended to elaborate on the emission structural characteristics of the Yangtze-Huai River Basin (e.g., the intertwining of dense urban agglomerations and large-scale agricultural areas) to explain why the findings of this study hold universal value for extrapolation to other similar urban agglomerations.

***Re*:** Thank you for your comments. As a typical transition zone and rapidly developing region in China, the Yangtze-Huai River Basin exhibits significant uniqueness and universality in its emission structure: The region contains not only densely populated large urban clusters such as Hefei and Nanjing (with intensive traffic and industrial emissions) but is also surrounded by vast major agricultural production areas (featuring rice-wheat crop rotation and extensive fertilization activities that release large amounts of ammonia and soil HONO). This emission pattern of "interlocking urban and rural areas with deep coupling of anthropogenic and biogenic sources" similarly exists in most developed urban agglomerations in China, such as Beijing-Tianjin-Hebei, the Yangtze River Delta, and the Chengdu-Chongqing region. Therefore, the vertically stratified mechanism of "lower layers dominated by anthropogenic sources (HONO/HCHO) and upper layers dominated by background O_3 photolysis" discovered in this study, along with the conclusion of "spatial decoupling of urban-rural oxidation driving mechanisms," can provide theoretical references for other regions facing similar dilemmas of mixed urban-rural emissions, possessing universal value for extrapolation to similar large urban agglomerations. We have added the above discussion to the relevant limitations statement in Section "4. Conclusion" of the revised manuscript.

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