

A Dual-Perspective Analysis of Ozone Formation Sensitivity in the Yangtze River Delta: Satellite and Ground-Based Evidence

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Highlights

What are the main findings?

- A dual-perspective analysis integrating TROPOMI satellite and ground-based data reveals distinct ozone (O₃) formation regimes between megacities (G1) and smaller cities (G2) in the Yangtze River Delta, with G1 showing a narrower transitional regime.
- High O₃ pollution ($\geq 160 \mu\text{g}/\text{m}^3$) occurs under all chemical regimes (VOC-limited, transitional, NO_x-limited) in megacities, but primarily under transitional and NO_x-limited conditions in smaller cities.

What are the implications of the main findings?

- Effective O₃ mitigation under high-pollution scenarios requires simultaneous reduction of both VOCs and NO_x, as single-precursor control is insufficient, especially in megacities with high VOC backgrounds.
- The findings advocate for adaptive, city-specific co-control strategies: megacities need flexible VOC-NO_x co-reduction, while smaller cities can benefit from measures aligned with their dominant sensitivity regimes.

Abstract

Surface ozone (O₃) pollution persists in the Yangtze River Delta (YRD) of China, with frequent exceedances despite stringent controls, driven by complex NO_x-VOC interactions. Current monitoring struggles to accurately identify O₃ formation regimes (VOC- vs. NO_x-limited) at urban scales during daytime. By integrating TROPOMI satellite data with high-resolution ground-based joint observations, we reveal scale-dependent regime transitions: megacities (G1 cities, population > 5 million and GDP > 1 trillion CNY yr⁻¹) exhibit narrow HCHO-to-NO₂ ratio (FNR) transition ranges (1.54–2.86), and O₃ pollution ($\geq 160 \mu\text{g}/\text{m}^3$) may occur under all regimes (VOC-limited, transitional, and NO_x-limited). In contrast, smaller G2 cities show wider transition ranges (1.85–3.43), with O₃ pollution occurring primarily under transitional and NO_x-limited conditions. Observations from a typical G1 city demonstrate that VOC reductions effectively control O₃ under VOC-limited regimes, while NO_x mitigation alone is insufficient in NO_x-limited environments due to persistently high VOC backgrounds. Our findings suggest city-specific strategies: G1 cities require adaptive co-control of VOCs and NO_x to accommodate localized chemistry, whereas G2 cities benefit from targeted measures aligned with dominant sensitivity regimes. This study provides observational verification of environment-dependent O₃ formation mechanisms,



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offering a methodological framework for precision air quality management in rapidly developing regions.

Keywords: ozone formation sensitivity; nitrogen dioxide; formaldehyde; satellite; ground-based multi-platform observation

1. Introduction

Surface ozone (O_3) pollution has become a persistent environmental crisis in the Yangtze River Delta (YRD), one of the most populous and industrialized megaregions, with frequent exceedances of national air quality standards during the warm season (April–October) in China [1–3]. Despite stringent emission controls, O_3 levels continue to rise, driven by complex interactions between nitrogen oxides (NO_x) and volatile organic compounds (VOCs) [4–6]. Accurate characterization of O_3 formation regimes—whether VOC-limited or NO_x -limited—is critical for effective mitigation [6–8], yet remains highly uncertain due to methodological limitations in current monitoring frameworks. Satellite observations provide invaluable regional coverage but lack the temporal resolution to capture diurnal photochemical cycles [9–11], while ground-based networks suffer from sparse spatial coverage and frequent temporal misalignment between precursor measurements and O_3 observations [12–14]. Consequently, regional O_3 management strategies in the YRD often rely on oversimplified assumptions, leading to suboptimal policy outcomes and inconsistent reductions in O_3 pollution.

Recent satellite-based studies have begun to assess regional O_3 sensitivity using tropospheric vertical column densities (TVCD) of HCHO and NO_2 as proxies for VOCs and NO_x , respectively [15–17]. Meanwhile, rapid advances in ground-based remote sensing techniques, including Multi-Axis Differential Optical Absorption Spectroscopy (MAX-DOAS) and differential absorption lidar, have considerably improved the capability to characterize the vertical distribution and temporal evolution of O_3 precursors [18–21]. These techniques provide valuable information on boundary-layer photochemistry and have enhanced our understanding of O_3 formation processes. Nevertheless, each observation platform has inherent limitations. Satellite observations provide broad spatial coverage but are restricted to a single daytime overpass, making them unable to capture continuous photochemical evolution. Ground-based remote sensing systems generally provide continuous temporal observations but remain spatially limited, while co-located measurements of ozone and precursor profiles are still scarce. As a result, integrated multi-scale observational validation remains limited, hindering a comprehensive understanding of how ozone formation regimes vary across urban environments and diurnal cycles, and ultimately constraining the development of effective emission-control strategies.

These limitations are particularly important in rapidly developing urban agglomerations such as the YRD, where O_3 formation regimes vary substantially among cities with different emission characteristics and urbanization levels. Although previous studies have investigated O_3 sensitivity using either satellite-derived FNR (the TVCD ratio of HCHO to NO_2) or local MAX-DOAS observations, the consistency between regional column-based indicators and local vertically resolved observations has not been systematically evaluated. Moreover, it remains unclear whether satellite-derived O_3 sensitivity thresholds are equally applicable across cities with different development intensities and whether they can adequately represent daytime photochemical conditions near the surface [22,23].

To address these challenges, this study presents a dual-perspective analysis that integrates high-resolution TROPospheric Monitoring Instrument (TROPOMI) satellite ob-

servations with ground-based multi-platform measurements to characterize O₃ formation sensitivity across urban gradients in the YRD. Unlike previous studies relying on either satellite retrievals or local observations alone, the proposed framework explicitly links regional satellite-derived indicators with continuous vertically resolved measurements. Our approach bridges the spatial–temporal divide by (1) mapping regional O₃ sensitivity patterns across cities stratified by population and economic scale, and (2) capturing continuous diurnal precursor dynamics through vertically resolved HCHO/NO₂ profiles and co-located O₃ measurements. This integrated methodology enables us to resolve previously uncharacterized differences in O₃ formation regimes between megacities and smaller urban centers, and to identify the critical role of diurnal VOC levels in sustaining high O₃ pollution. Our findings provide observational verification of environment-dependent O₃ formation mechanisms at urban scales, providing an observational framework for effective air quality management in rapidly developing regions where single-precursor control strategies have proven insufficient.

2. Materials and Methods

2.1. Satellite Data

TVCD of HCHO and NO₂ used in this study were retrieved from TROPOMI onboard the Copernicus Sentinel-5 Precursor satellite. The TROPOMI Level-2 offline data products for HCHO (<https://doi.org/10.5270/S5P-vg1i7t0>) and NO₂ (<https://doi.org/10.5270/S5P-9bnp8q8>) were obtained from the European Space Agency. The data cover overpass times around 12:00–14:00 local time (LT) from April to October 2021–2024, with a spatial resolution of approximately 5.5 km × 3.5 km. We applied a quality filter to retain pixels with a quality assurance value (qa_value) > 0.5 for HCHO and qa_value ≥ 0.75 for NO₂.

To align satellite observations with the surface O₃ measurements from the China National Environmental Monitoring Center (CNEMC), satellite HCHO and NO₂ TVCDs were spatially collocated with individual monitoring stations using ordinary Kriging interpolation. To preserve local spatial representativeness, only TROPOMI pixels located within the administrative boundary of the corresponding city were included in the interpolation, thereby minimizing the influence of neighboring cities with different emission characteristics. The interpolated TVCDs were subsequently paired with hourly O₃ observations acquired during the satellite overpass period (12:00–14:00 LT) for FNR analysis.

2.2. Ground-Based Observation

Ground-based joint observations were conducted at the Binjiang Atmospheric Measurement Station of Nanjing University of Information Science and Technology (hereafter referred to as NUIST-BJ-AEMS, 32.08°N, 118.77°E) from 2022 to 2024. The station is located in northern Nanjing, bordered by residential areas, industry areas, and major expressways (Figure S1). Data were collected using (1) Multi-Axis Differential Optical Absorption Spectroscopy (MAX-DOAS) to retrieve vertical profiles and vertical column densities of HCHO and NO₂. The retrieval algorithm is a well-known optimal estimation method [24], and relevant DOAS fitting parameters are detailed in Table S1 [25–30]. (2) Ground-level NO_x and O₃ data were measured at the same location as the MAX-DOAS instrument, ensuring co-located measurements for direct comparison.

The vertical profiles were divided into 26 layers below 3 km: the first 10 layers on a 100 m grid (<2 km) and a 200 m grid (2–3 km). The vertical column density from the *i*th layer up to the *j*th layer ($VCD_{target, i \geq 2, j > i}$) was calculated as:

$$VCD_{target} = \sum_{i=2}^j (\overline{conc_{spec,i} + conc_{spec,i-1}}) \times (h_i - h_{i-1}) \quad (1)$$

where $conc_{spec,i}$ denotes the concentration of HCHO (or NO₂) in the i th layer (in molecules·cm⁻²), h_i is the altitude of the boundary of layer i , and the overbar indicates averaging between adjacent layers to approximate the trapezoidal integration of the concentration profile.

2.3. Ancillary Data

Hourly O₃ concentrations from the China National Environmental Monitoring Centre (CNEMC) were aligned with TROPOMI satellite overpasses (12:00–14:00 LT). O₃ measurements within this window were averaged.

2.4. FNR Analysis and Urban Classifications

To investigate urban differences in O₃ formation sensitivity, the cities in the YRD were classified into two categories according to their urban development level. Cities with both a permanent resident population exceeding 5 million and a gross domestic product (GDP) exceeding 1 trillion CNY in 2022 were classified as Group 1 cities (G1), whereas the remaining prefecture-level cities were classified as Group 2 (G2). These thresholds were selected to distinguish highly urbanized metropolitan areas from other prefecture-level cities while ensuring statistically meaningful sample sizes for subsequent comparisons. A complete list of cities and their corresponding socioeconomic statistics is provided in Table S2.

To characterize O₃ formation sensitivity, paired O₃ and FNR observations were ranked in ascending order of FNR and grouped into fixed bins with an interval of 0.1. The mean O₃ concentration was calculated for each bin, and bins containing fewer than 100 paired observations were excluded to ensure statistical robustness. The relationship between the binned mean O₃ concentrations and FNR was fitted using a third-degree polynomial to characterize the non-linear O₃–FNR relationship and to identify the FNR corresponding to the maximum fitted O₃ concentration (FNR[O₃-peak]). Following previous studies, the transitional regime was determined from the observed FNR values corresponding to the highest 10% of O₃ concentrations, whereas FNR values below and above this range were classified as VOC-limited and NO_x-limited regimes, respectively [31–33]. Following previous studies, the transitional regime was determined from the observed FNR values corresponding to the highest 10% of O₃ concentrations, whereas FNR values below and above this range were classified as VOC-limited and NO_x-limited regimes, respectively.

3. Results and Discussion

3.1. Regional Ozone Sensitivity in YRD Cities

To characterize the regional O₃ formation regime in the Yangtze River Delta during the warm season, we constructed a satellite-derived function using HCHO and NO₂ TVCD as proxies for VOCs and NO_x, respectively (Figure 1a), which reveals a robust EKMA-like pattern. Each grid cell represents the mean surface O₃ concentration from all paired observations within a 1×10^{15} molecules·cm⁻² interval of TVCD[HCHO] and TVCD[NO₂]. The resulting O₃ distribution shows peak concentrations in the high-HCHO, high-NO₂ region, indicating that O₃ pollution in the YRD is jointly driven by both precursors. To accurately characterize the non-linear O₃–HCHO–NO₂ relationship between surface O₃ and the HCHO-to-NO₂ TVCD ratio (FNR), paired satellite–surface observations were stratified into bins with an FNR interval of 0.1. Statistical reliability was ensured by retaining only FNR bins containing at least 100 paired observations. Our analysis identifies a transitional regime across YRD cities when FNR = 1.67–3.11. VOC-limited conditions prevail when FNR < 1.67, whereas NO_x-limited chemistry dominates when FNR > 3.11. Notably, O₃ concentrations exceeding 160 µg·m⁻³ were observed across all three regimes, highlighting

that O_3 pollution in the YRD arises from heterogeneous photochemical regimes rather than a single dominant regime [34–36].

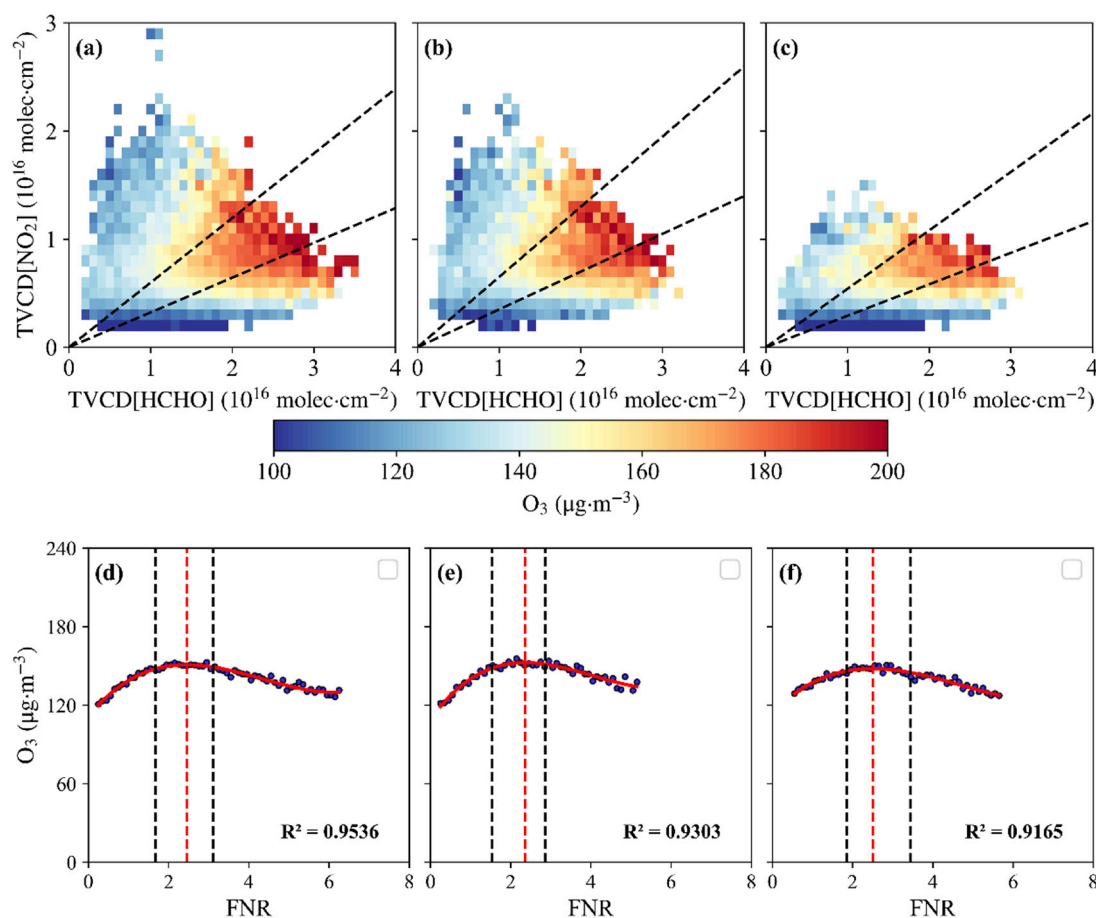


Figure 1. (a) Surface O_3 concentration as a function of TROPOMI TVCD[HCHO] and TVCD[NO₂] in YRD cities. All hourly O_3 concentrations from April to October during 2021–2024 were binned according to TVCD[HCHO] and TVCD[NO₂] (interval: 1×10^{15} molec·cm^{−2}) at corresponding overpass times (12:00–14:00 LT). The black dotted lines delineate the TROPOMI TVCD[HCHO]/TVCD[NO₂] (FNR) values of 1.67 and 3.11. (b) Same as (a), but for YRD G1 cities with FNR values of 1.54 and 2.86, and (c) YRD G2 cities with FNR values of 1.85 and 3.43. (d) Surface O_3 concentration as a function of FNR in all YRD cities, (e) YRD G1 cities, and (f) YRD G2 cities. The red solid line is the fitted third-degree polynomial curve. The red dotted lines indicate the FNR values at peak O_3 concentrations (FNR[O_3 -peak]), and the black dotted lines represent the FNR thresholds for the VOC-limited, transitional, and NO_x-limited regimes (FNR ranges from (left) to (right)).

To account for heterogeneity in urban scale and economic development, YRD cities were divided into two groups based on thresholds of population greater than 5 million and GDP exceeding 1 trillion CNY per year. Results confirm that the non-linear O_3 –HCHO–NO₂ relationship persists across all urban classification schemes. G1 exhibited a narrower transitional regime (FNR = 1.54–2.86), with O_3 concentration peaking at FNR = 2.20 (FNR[O_3 -peak], Figure 1b,d). G2 cities have a broader transitional regime (FNR = 1.85–3.43) and a higher FNR[O_3 -peak] value (2.64, Figure 1e,f). The fitted O_3 –FNR curves consistently show a leftward shift for both the transitional regime and O_3 peak in G1 relative to G2, suggesting that highly urbanized cities reach maximum ozone production under comparatively lower FNR conditions. This shift is observed over the overall fitted relationship rather than being controlled by a limited number of FNR bins, indicating that the differences between the two urban groups are systematic rather than arising from isolated observations.

Unlike G1 cities, where O₃ exceedances are observed across all three regimes, O₃ exceedances in G2 cities are primarily concentrated in the transitional and NO_x-limited regimes. This difference suggests that intense NO_x emissions in G1 cities may lead to atmospheric NO_x saturation, where further NO_x reductions could paradoxically elevate O₃ levels, while VOC mitigation would consistently suppress O₃ [37–39]. G2 cities remain primarily sensitive to NO_x or to the synergistic effects of NO_x and VOCs [39,40]. Besides differences in urban development level, variations in regional emission characteristics may also contribute to the observed contrast between G1 and G2 cities. Differences in industrial composition, traffic intensity, solvent use, and petrochemical activities can modify VOC abundance and precursor ratios, thereby influencing ozone formation sensitivity. Because the present classification is based on socioeconomic indicators rather than emission inventories, these factors are proposed as plausible contributors rather than direct evidence for the observed regional differences.

Collectively, these findings demonstrate that TROPOMI captures not only the qualitative EKMA structure but also its quantitative variation across urban scales, enabling targeted air quality management based on city-specific pollution profiles. Although uncertainties associated with satellite retrievals, cloud screening, and spatial interpolation may affect individual observations, the large number of paired satellite–surface measurements retained in each FNR bin improves the statistical robustness of the derived regional O₃–FNR relationships.

3.2. Local Ozone Sensitivity with Diurnal Coverage

Satellite observations (TROPOMI in this study) provide regional-scale insights but have critical temporal limitations. The single overpass (typically 12:00–14:00 LT) captures only a snapshot of photochemical activity, missing the diurnal variability in O₃ formation [9,16]. To address this, we deployed a multi-platform observational system at NUIST-BJ-AEMS comprising (1) MAX-DOAS for vertical profiles and VCD of HCHO and NO₂, and (2) co-located ground NO_x (for validation, R = 0.87, Figure S2) and O₃ observations. This configuration ensures spatiotemporally matched data (9:00–16:00, coinciding with peak photochemical activity), resolving a problem in previous studies where MAX-DOAS-derived HCHO/NO₂ ratios were often paired with distant, mismatched O₃ measurements [12,21,41]. This integrated data set uniquely enables us to quantify ozone sensitivity without spatial misalignment or temporal averaging artifacts.

Based on tropospheric FNR observed by NUIST-BJ-AEMS, the transitional regime is 1.50–2.78, similar to the regional FNR threshold (1.54–2.86). O₃ exceedances occur predominantly when HCHO TVCD exceeds 1.8×10^{16} molecules·cm^{−2} within this FNR range (Figure S3). Numerous studies have established that photochemical production within the boundary layer is a primary driver of O₃ pollution episodes, and anthropogenic emissions strongly modulate the concentrations of key O₃ precursors within this shallow atmospheric layer [42–44]. Figure 2 shows the diurnal evolution of HCHO and NO₂ vertical distributions. Both species exhibit strong enrichment in lower troposphere (LTropo, below 500 m, Figure 2a–c) and large relative diurnal variability rates (RDVR, Figure 2d), highlighting the dominant role of the lower troposphere in regulating daytime ozone production.

Figure 3 shows a transitional regime characterized by an FNR range in LTropo (FNR_{LTropo}) of 2.28–4.23, where O₃ formation shifts from VOC-limited (FNR_{LTropo} < 2.28) to NO_x-limited (FNR_{LTropo} > 4.23). The peak O₃ concentration occurs at FNR_{LTropo} = 4.02, near the upper boundary of the transitional regime. O₃ concentrations exceeding 160 µg m^{−3} predominantly occur when FNR_{LTropo} > 2.28 (Figure 3b), highlighting the dominant role of the lower troposphere in regulating daytime O₃ production.

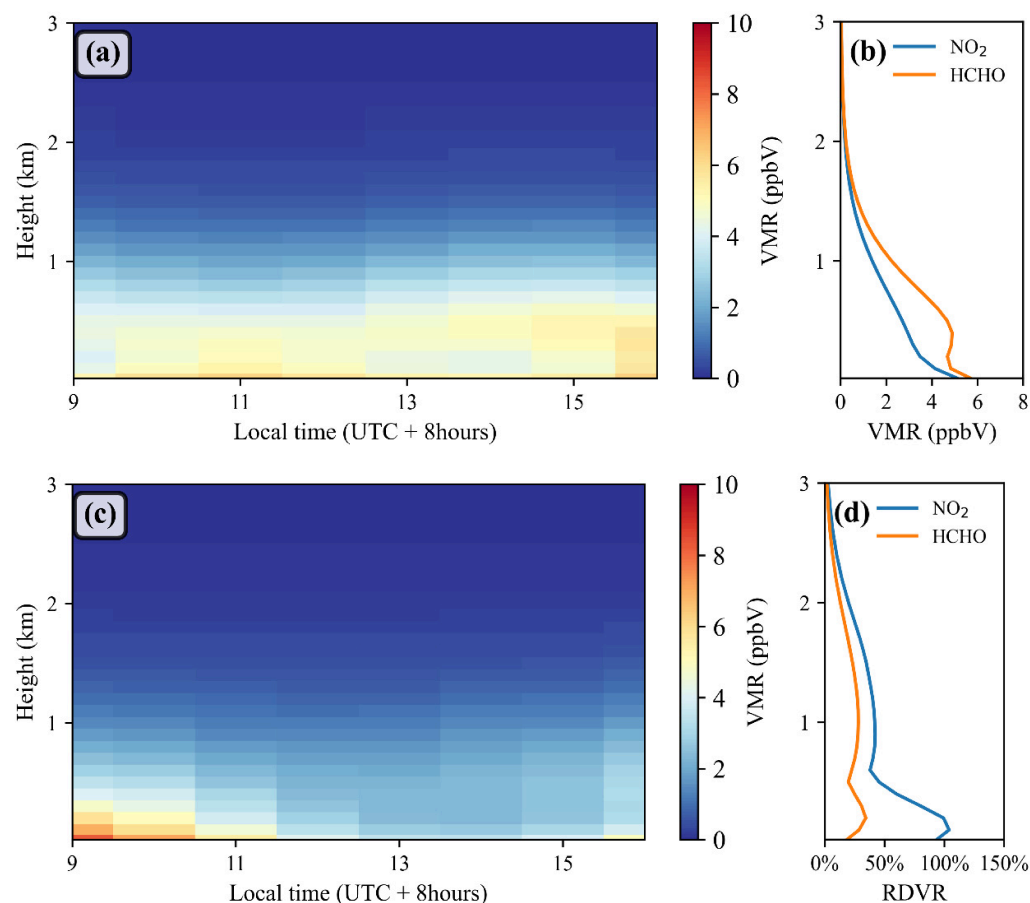


Figure 2. (a) Diurnal evolution of HCHO vertical profiles. (b) Average vertical profiles of HCHO and NO₂. (c) Same as (a), but for NO₂. (d) Relative diurnal variation rate (RDVR) profiles of HCHO and NO₂, where the RDVR is calculated by subtracting the minimum diurnal concentration from the maximum diurnal concentration and dividing by the mean diurnal concentration.

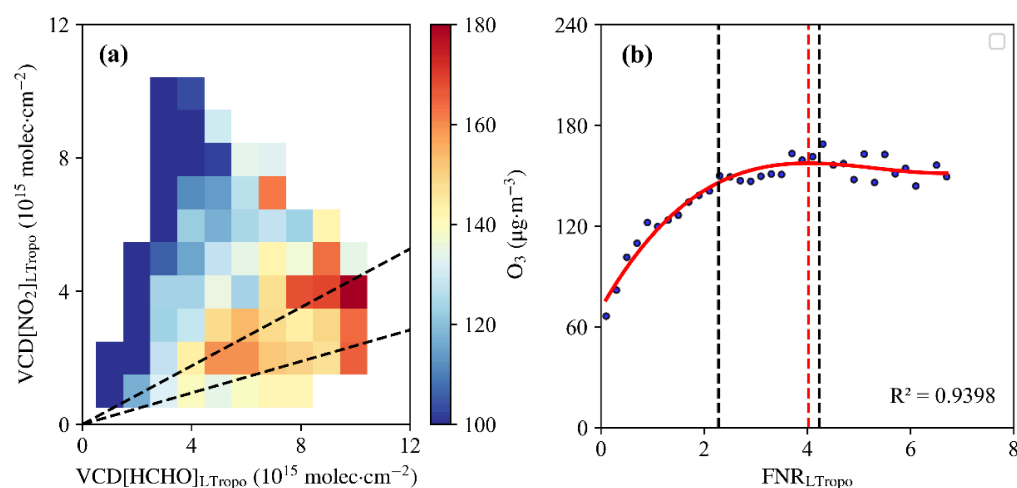


Figure 3. (a) Surface O₃ concentration as a function of VCD[HCHO]_{LTropo} and VCD[NO₂]_{LTropo} in NUIST-BJ-AEMS. Diurnal hourly O₃ concentrations from April to October during 2022–2024 were binned according to VCD[HCHO]_{LTropo} and VCD[NO₂]_{LTropo} (interval: 1×10^{14} molec·cm⁻²). The black dotted lines indicate the VCD[HCHO]_{LTropo}/[NO₂]_{LTropo} (FNR_{LTropo}) values of 2.28 and 4.23. (b) Surface O₃ concentration as a function of FNR_{LTropo}. The red solid line is the fitted third degree polynomial curve. The red dotted line represents FNR[O₃-peak], and the black dotted lines represent the FNR thresholds for the VOC-limited, transitional, and NO_x-limited regimes (FNR ranges from (left) to (right)).

The observed O_3 –FNR relationships suggest that reducing NO_x alone may yield only limited O_3 reductions under severe ozone pollution conditions, likely because abundant VOCs continue to sustain rapid photochemical production. Likewise, within the transitional regime ($2.28 < FNR_{LTropo} < 4.23$), reducing either precursor alone may not be sufficient to substantially suppress O_3 formation. Our observations are therefore consistent with the potential benefit of coordinated VOC and NO_x reductions during severe ozone pollution episodes. Because the present study is based on observational relationships rather than chemical sensitivity simulations, this inference should be regarded as qualitative rather than quantitative.

These findings have significant implications for air quality management and help resolve apparent contradictions in the literature regarding the effectiveness of NO_x controls in high- O_3 environments [45–47]. While traditional EKMA-based strategies recommend VOC-focused controls in VOC-limited regimes and NO_x -focused controls in NO_x -limited regimes, our observations suggest that coordinated VOC– NO_x reductions may provide greater benefits during persistently elevated O_3 episodes. This may explain why cities implementing single-precursor control strategies sometimes experience only modest O_3 reductions despite substantial emission reductions. The consistency between our observations and regional satellite-derived thresholds (Section 3.1) further supports this control paradigm across spatial scales.

Meteorological conditions, including boundary-layer evolution, temperature, and solar radiation, may also influence ozone formation sensitivity and deserve further investigation using process-based modeling [48,49]. Furthermore, because vertically resolved observations were only available for a typical G1 city, the proposed framework should be further evaluated using observations from representative G2 cities.

4. Conclusions

This study presents a dual-perspective analysis integrating TROPOMI satellite observations with high-resolution ground-based measurements to characterize O_3 formation sensitivity in the YRD cities. Our findings reveal significant scale-dependent differences in O_3 formation regimes between G1 and G2 cities. G1 cities exhibit a narrower transitional regime ($FNR = 1.54$ – 2.86) and experience O_3 pollution under all chemical regimes (VOC-limited, transitional, and NO_x -limited), whereas G2 cities show a broader transition range ($FNR = 1.85$ – 3.43) with pollution primarily occurring in transitional and NO_x -limited conditions.

Ground-based diurnal observations further suggest that coordinated reductions in both VOCs and NO_x may provide greater benefits during severe ozone pollution episodes than single-precursor control strategies. Because these findings are derived from observational relationships rather than chemical sensitivity simulations, they should be interpreted as qualitative observational evidence.

Our work provides observational evidence for environment-dependent O_3 formation mechanisms at urban scales and establishes a robust methodological framework that bridges satellite and ground-based platforms. This integrated approach offers actionable insights for precision air-quality management and supports adaptive, city-specific precursor control strategies tailored to local photochemical environments. Future validation using vertically resolved observations from representative G2 cities, together with process-based modeling, will further improve the applicability of the proposed framework.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/rs18142336/s1>. Table S1: DOAS fitting configuration for the DSCDs of O_4 , NO_2 , and HCHO; Table S2: Population, GDP, and urban classification of the 41 cities in the Yangtze River Delta; Figure S1: Geographical characteristics, location, and surrounding functional

areas of NUIST-BJ-AEMS; Figure S2: Linear correlation analysis between the Ground-level NO₂ concentration of MAX-DOAS and NUIST-BJ-AEMS; Figure S3: (a) Surface O₃ concentration as a function of VCD[HCHO] and VCD[NO₂] in NUIST-BJ-AEMS. (b) Surface O₃ concentration as a function of FNR.

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Data Availability Statement: The data presented in this study are available on request from the corresponding author.

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Conflicts of Interest: The authors declare no conflicts of interest.

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