

1 A dataset of vertical profiles of O₃ and HONO from the hyperspectral
2 vertical remote sensing network in China (2021-2024)

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22 **Abstract**

23 Photolysis of HONO and O₃ in the troposphere is one of the primary sources of
24 OH radical and a fundamental control on atmospheric oxidative capacity. Their
25 vertical distributions and diurnal evolution are therefore essential for elucidating
26 photochemical processes in the planetary boundary layer and the lower free
27 troposphere. Yet long-term, continuous observations of the vertical profiles of HONO,
28 O₃, their photolysis frequencies, and the resulting OH production rates remain
29 extremely limited, particularly at multi-regional and interannual scales. Here we
30 present vertical profile measurements of HONO and O₃ acquired by the Chinese
31 Hyperspectral Vertical Remote Sensing Network during 2021–2024. The dataset
32 comprises 22 representative sites spanning urban, suburban, plateau, and basin
33 environments, covering diverse surface and climatic regimes. Profiles extend from the
34 surface to 4 km with ~100 m vertical resolution and ~15 min temporal resolution.
35 Using the TUV model with co-retrieved aerosol and trace-gas profiles, we derive
36 photolysis frequencies of HONO and O₃ and the corresponding OH production rates,
37 P(OH)_{HONO} and P(OH)_{O₃}. The observations reveal robust regional patterns in the
38 diurnal and vertical structure of tropospheric photochemical activity. Photolysis
39 frequencies peak near local noon and generally increase with altitude from the surface
40 layer to the upper mixed layer and the lower free troposphere, whereas OH production
41 rates reach their maxima within the boundary layer and decrease with height.
42 Processed using a unified retrieval framework and rigorous quality control, this
43 dataset provides quantitative constraints on the contribution of HONO and O₃
44 photolysis to tropospheric OH, supports improved radical parameterizations in
45 chemical transport models, and enables synergistic multi-platform remote sensing
46 analyses. By delivering the first systematic, long-term vertical profiles of HONO, O₃,
47 and their OH production in China, this public dataset fills a critical observational gap
48 and offers a robust basis for investigating the spatiotemporal evolution of tropospheric
49 oxidative capacity across regions and altitude ranges, with substantial scientific
50 significance and long-term applicability. The dataset is available for free at Zenodo
51 (<https://doi.org/10.5281/zenodo.18489836>, Zou et al., 2026)

52

53 **1. Introduction**

54 Over the past decade, “the implementation of China’s Air Pollution Prevention
55 and Control Action Plan” (2013) and “the Three-Year Action Plan for Defending the
56 Blue Sky” (2018) has led to a marked reduction in fine particulate matter (PM_{2.5})
57 nationwide (Liu et al., 2023b; Wang et al., 2020). In contrast, ozone (O₃)—a
58 secondary pollutant and a major atmospheric oxidant—has continued to increase on
59 average in economically developed regions such as the Beijing–Tianjin–Hebei area,
60 the Yangtze River Delta, and the Pearl River Delta, where it has emerged as the most
61 intractable air-quality problem after PM_{2.5} (Guo et al., 2023; He et al., 2023a; Li et al.,
62 2020; Lyu et al., 2025; Zou et al., 2025). To address the observational gaps in these
63 and other key regions, we developed a comprehensive dataset. The dataset comprises

64 measurements from 22 ground-based sites across five major regions of China—North,
65 East, Southwest, South, and Central China. Photochemical air pollution is a dominant
66 driver of urban and regional air-quality degradation, characterized by the rapid
67 sunlight-driven accumulation of secondary species, most notably O₃ (Dewan and
68 Lakhani, 2022; Donzelli and Suarez-Varela, 2024; Sharma et al., 2025; Wang et al.,
69 2025b). Beyond being a typical secondary pollutant, O₃ is a powerful oxidant that
70 exerts substantial impacts on regional climate, ecosystems, and human health (Monks
71 et al., 2015; Sharma et al., 2025; Wang et al., 2025b; Xing et al., 2017). Nitrous acid
72 (HONO), a short-lived reactive nitrogen species, occurs at relatively low
73 concentrations but represents a major primary source of the OH radical, the key
74 “detergent” of the troposphere (Andersen et al., 2023; He et al., 2023c; Song et al.,
75 2023a; Zhang et al., 2023a). In polluted environments, photolysis of HONO can
76 account for 20–80% of total OH production, and its relative importance is particularly
77 pronounced during early morning and late afternoon, when solar elevation is low and
78 alternative OH sources are less efficient (Elshorbany et al., 2010; He et al., 2023c;
79 Zhang et al., 2023a). A quantitative understanding of the formation and transport of
80 both HONO and O₃ is therefore essential for elucidating the mechanisms of
81 tropospheric photochemical pollution and for designing effective mitigation strategies.

82 Despite extensive research on HONO and O₃, major gaps persist in observations
83 of their vertical structure and in the parameterization of key photochemical processes,
84 limiting a mechanistic understanding of photochemical air pollution (Liu et al., 2023a;
85 Wang et al., 2018, 2025c; Zhang et al., 2024; Zhu et al., 2025b). Vertical
86 measurements remain particularly sparse, and concurrent profiles of HONO and O₃
87 are largely unavailable (Garcia-Nieto et al., 2018a; Song et al., 2023a; Wang et al.,
88 2018, 2025c; Zhu et al., 2025b). China National Environmental Monitoring Center
89 (CNEMC), with more than 2,000 surface stations, provides routine measurements of
90 PM_{2.5}, NO₂, and SO₂, but lacks observations of key photochemical precursors such as
91 HONO and volatile organic compounds (VOCs) (Liu et al., 2023a; Qu et al., 2020;
92 Zhang et al., 2024; Zhu et al., 2025b). More fundamentally, surface observations
93 alone cannot resolve pollutant distributions within the planetary boundary layer or
94 capture variations in vertical atmospheric structure (Wang et al., 2018, 2019, 2025c;
95 Xuan et al., 2025a; Zhu et al., 2025b), and exclusive reliance on near-surface data
96 may therefore bias assessments of regional transport and accumulation (Liu et al.,
97 2023a; Wang et al., 2019, 2025c). Spaceborne sensors, including MODIS, CALIPSO,
98 TROPOMI, and OMI, provide global fields of aerosol optical depth (AOD) and
99 vertical column densities (VCDs) for selected trace gases. However, their limited
100 temporal sampling and spatial resolution preclude resolving the fine-scale diurnal
101 variability and fine vertical structure of O₃ and HONO (Itahashi et al., 2020; Johnson
102 et al., 2024; Torres et al., 2020a; Wang et al., 2025a). Chemical transport models
103 (CTMs) and regional climate models (RCMs) can reproduce the spatiotemporal
104 evolution of pollutants, but their performance depends critically on initial and
105 boundary conditions, and uncertainties in vertical parameterizations—such as

106 turbulent mixing and chemical mechanisms—often lead to substantial biases in
107 simulated profiles (Chambers et al., 2019; Kim et al., 2024; Li et al., 2021; Sekiya et
108 al., 2025; Thürkow et al., 2024). Current in situ and remote-sensing techniques also
109 have intrinsic limitations. Lidar systems provide high-resolution aerosol profiles but
110 are restricted in detectable gaseous species and spatial coverage (Anon, 2023; Johnson
111 et al., 2024; Torres et al., 2020b). Aircraft and balloon soundings yield detailed upper-
112 air observations but are expensive and unsuitable for sustained, long-term monitoring
113 (Johnson et al., 2024; Sekiya et al., 2025; Wang et al., 2025a; Yu et al., 2025). Tower
114 measurements, while valuable near the surface, are height-limited and cannot capture
115 the full vertical variability across the boundary layer (Chambers et al., 2019; Kim et
116 al., 2024; Thürkow et al., 2024).

117 To address the observational limitations and scientific questions outlined above,
118 we developed a comprehensive dataset of vertical profiles of HONO, O₃, and their
119 photolysis frequencies using the Chinese Hyperspectral Vertical Remote Sensing
120 Network. The primary objective is to resolve the vertical structure of HONO and O₃
121 and to quantify the altitude-resolved production of OH radicals from their photolysis.
122 This dataset fills a critical gap in vertical observations of key photochemical species
123 over China and provides a unique basis for assessing the contribution of HONO
124 photolysis to boundary-layer OH budget, the vertical characteristics of O₃ formation,
125 and the role of aerosols in modulating photolysis rates. The dataset comprises
126 measurements from 22 ground-based sites across five major regions of China—North,
127 East, Southwest, South, and Central China—collected during 2021–2024. Its core
128 products are high-temporal-resolution vertical profiles of HONO and O₃ spanning 0–4
129 km. Public release of this dataset will enable systematic investigations of the
130 unresolved sources of HONO in the boundary layer and the vertical variability in O₃
131 production sensitivity. When combined with numerical models, the high-resolution
132 vertical information can be used to evaluate and refine photochemical mechanisms,
133 quantify the contribution of HONO photolysis to the tropospheric OH budget, and
134 reduce uncertainties in vertical parameterizations. These advances will, in turn,
135 support robust source attribution of O₃ pollution and inform the development of
136 coordinated regional control strategies for PM_{2.5} and O₃. The following sections
137 describe the site distribution, observational and retrieval methods, and the seasonal
138 and diurnal features of the HONO and O₃ vertical structures revealed by this dataset.

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140 **2. Method**

141 **2.1 Description of the monitoring site**

142 The dataset is derived from 22 hyperspectral ground-based vertical remote
143 sensing stations distributed across five major regions of China—North, East,
144 Southwest, South, and Central China—forming an integrated network that samples a
145 wide range of representative atmospheric environments (Table 1). The sites span
146 urban cores, urban–suburban transition zones, regional background areas, coastal and
147 land–sea interaction regions, as well as plateau, mountain, and basin settings, thereby

148 providing a three-dimensional observational framework for key photochemical
 149 species. In North China, stations at the Chinese Academy of Meteorological Sciences
 150 (CAMS1, CAMS2) and the University of Chinese Academy of Sciences (UCAS),
 151 located within Beijing (~100–120 m a.s.l.), characterize the heavily urbanized and
 152 industrialized core of the Beijing–Tianjin–Hebei megacity cluster. The Wangdu (WD)
 153 site in suburban Baoding represents regional background conditions, whereas the
 154 Shijiazhuang Luancheng (SJZ_LC) site was included to better resolve pollution
 155 features specific to industrial cities. The Shanxi University (SXU) site in the Taihang
 156 Mountains (780 m a.s.l.) provides critical constraints on pollutant formation and
 157 transport between mountainous terrain and adjacent plains. In East China, stations are
 158 distributed across the Yangtze River Delta and its hinterland, covering topography
 159 from coastal lowlands to inland mountains. The summit of Mount Tai (TS; 1,500 m
 160 a.s.l.) offers vertical profiles under relatively clean, high-altitude background
 161 conditions. The Nanjing University of Information Science and Technology (NUIST)
 162 site represents a densely populated and economically developed urban environment,
 163 while sites at Huaibei Normal University (HNU), Anhui University (AHU), and
 164 Changfeng (CF) in Anhui Province (30–35 m a.s.l.) capture urban–suburban transition
 165 regimes. Southwest China is represented by the Chengdu Academy of Environmental
 166 Sciences (CDAES; 505 m a.s.l.) on the Chengdu Plain and the Chongqing (CQ; 332
 167 m a.s.l.) site within the Sichuan Basin. These stations are strategically located to
 168 investigate pollutant accumulation and transport under high-humidity conditions and
 169 strong topographic confinement, and to probe photochemical processes in complex
 170 terrain. In South China, a dense network was established over the Pearl River Delta
 171 megacity region. In addition to sites at the Guangzhou Institute of Geochemistry (GIG)
 172 and the Southern University of Science and Technology (SUST) in Shenzhen,
 173 multiple stations in Guangzhou (Zhuliao, Nansha, Timian, Gongyuan, and
 174 Daxuecheng; 15–155 m a.s.l.) form an intra-urban array. This configuration allows
 175 detailed examination of the combined influences of land–sea breezes, anthropogenic
 176 emissions, and local meteorology on the vertical distributions of HONO and O₃.
 177 Central China is represented by the Luoyang (LY) site, located in the middle reaches
 178 of the Yellow River within a mixed industrial–agricultural region, providing key
 179 constraints on regional transport and accumulation over the central plains. Together,
 180 the broad geographic coverage and pronounced contrasts in elevation and surface type
 181 make this network well suited to resolve the vertical distributions of aerosols, HONO,
 182 and O₃ across urban, suburban, coastal, mountainous, and basin environments. It thus
 183 offers a robust observational basis for investigating the dynamics of photochemical air
 184 pollution over major regions of China.

185 Table 1. Geographic information of the stations in the Chinese Hyperspectral Ground-
 186 Based Vertical Remote Sensing Network.

Region	Site (code)	Longitude (° E)	Latitude (° N)	Altitude (m)
North China	Chinese Academy of Meteorological Sciences (CAMS1)	116.32	39.94	100
	Chinese Academy of Meteorological	116.32	39.94	100

	Sciences (CAMS2)			
	University of Chinese Academy of Sciences (UCAS)	116.67	40.4	120
	Wangdu (WD)	115.15	38.17	35
	Shijiazhuang Luancheng (SJZ_LC)	114.61	37.91	70
	Shanxi University (SXU)	112.58	37.63	780
	Taishan (TS)	117.1	36.25	1500
	Nanjing University of Information Science and Technology (NUIST)	118.71	32.2	73
East China	Lin'an (LA)	119.75	30.3	140
	HuaiBei Normal University (HNU)	116.8	33.98	35
	Anhui University (AHU)	117.18	31.77	30
	Changfeng (CF)	117.18	32.21	30
Southwest China	Chengdu Academy of Environmental Sciences (CDAES)	104.04	30.65	505
	Chongqing (CQ)	106.5	29.6	332
	Guangzhou Institute of Geochemistry (GIG)	113.35	23.15	30
	Southern University of Science and Technology (SUST)	113.99	22.59	40
South China	Guangzhou Zhuliao (GZ_ZL)	113.34	23.36	20
	Guangzhou Nansha (GZ_NS)	113.61	22.77	15
	Guangzhou Timian (GZ_TM)	113.29	23.55	155
	Guangzhou Gongyuan (GZ_GY)	113.26	23.13	15
	Guangzhou Daxuecheng (GZ_DXC)	113.39	23.04	10
Central China	Luoyang (LY)	112.45	34.67	100

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2.2 Instrument setup

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2.3 Spectral retrieval

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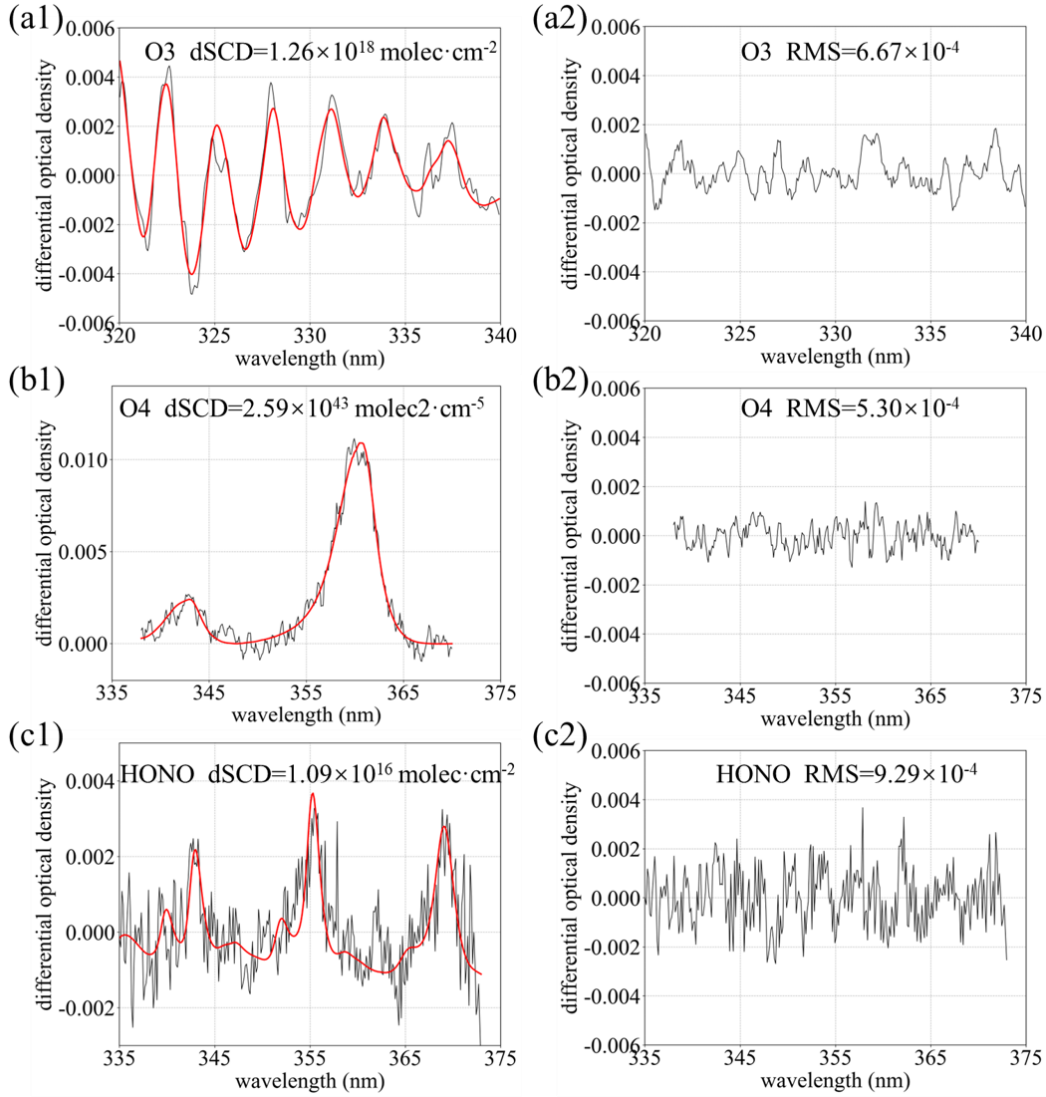
Ultraviolet–visible spectra measured by the ground-based instruments were analysed with the QDOAS software (version 3.2) developed by BIRA-IASB.

210 Differential optical absorption spectroscopy (DOAS) was applied to retrieve the
 211 differential slant column densities (DSCDs) of the oxygen dimer (O_4), O_3 , and HONO.
 212 For each elevation scan, the zenith spectrum (90° elevation) acquired within the same
 213 scanning sequence was used as the reference and subtracted from spectra at lower
 214 elevation angles, thereby isolating the narrow-band absorption features of trace gases
 215 from broadband structures and enabling robust retrieval of target species. The fitting
 216 settings follow Xing et al. (2021, 2024a, b) and are summarized in Table 2. To
 217 account for the Ring effect arising from rotational Raman scattering and Fraunhofer
 218 line filling-in, a Ring spectrum calculated with DOASIS was included in the fit.
 219 Broadband spectral structures were represented and removed using a fifth-order
 220 polynomial. This allowing accurate separation of narrow-band molecular absorption.
 221 Strict quality control was applied: only retrievals with a root-mean-square (RMS)
 222 fitting residual below 1×10^{-3} were retained, ensuring the robustness and stability of
 223 the dataset. Representative spectral fits and residuals for O_4 , O_3 , and HONO are
 224 shown in Figure 1.

225 Table 2. Detailed retrieval settings for O_4 , O_3 , and HONO.

Parameter	Data source	Fitting interval		
		O_4	O_3	HONO
Wavelength range		338–370 nm	320–340 nm	335–373 nm
NO_2	220 K, I_0^* correction (SCD of 10^{17} molec·cm $^{-2}$); (Vandaele et al., 1998)	√	×	√
NO_2	298 K, I_0 correction (SCD of 10^{17} molec·cm $^{-2}$); (Vandaele et al., 1998)	√	√	√
O_3	223 K, I_0 correction (SCD of 10^{18} molec·cm $^{-2}$); (Serdyuchenko et al., 2014)	√	√	√
O_3	243 K, I_0 correction (SCD of 10^{18} molec·cm $^{-2}$); (Serdyuchenko et al., 2014)	×	×	√
O_3	293 K, I_0 correction (SCD of 10^{18} molec·cm $^{-2}$); (Serdyuchenko et al., 2014)	√	√	×
O_4	293 K, I_0 correction (SCD of 3×10^{43} molec 2 ·cm $^{-5}$); (Thalman and Volkamer, 2013)	√	√	√
HCHO	293 K, I_0 correction (SCD of 5×10^{15} molec·cm $^{-2}$); (Orphal and Chance, 2003)	√	√	√
BrO	273 K, I_0 correction (SCD of 10^{13} molec·cm $^{-2}$); (Fleischmann et al., 2004)	√	×	√
Ring	Ring spectra calculated with DOASIS	√	√	√
HONO	I_0 correction (SCD of 10^{15} molec·cm $^{-2}$);(Stutz et al., 2000)	×	×	√
Polynomial degree		5	5	5
Intensity offset		Constant	Constant	No

226 * Solar I_0 correction, Aliwell et al.(2002).



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228 Figure 1. (a1) O₃, (b1) O₄, and (c1) HONO DOAS fitting examples; (a2) O₃, (b2) O₄,
 229 and (c2) HONO fitting residuals.

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231 2.4 Vertical profile retrieval algorithm

232 Vertical profiles of aerosols and trace gases (HONO and O₃) were retrieved using
 233 an inversion framework based on the optimal estimation method (OEM). The forward
 234 radiative transfer calculations were performed with the linearized pseudo-spherical
 235 vector discrete ordinate model VLIDORT (Spurr, 2006). The posterior state vector \mathbf{x}
 236 was obtained by minimizing the cost function χ^2 :

$$237 \quad \chi^2 = (\mathbf{y} - \mathbf{F}(\mathbf{x}, \mathbf{b}))^T \mathbf{S}_\varepsilon^{-1} (\mathbf{y} - \mathbf{F}(\mathbf{x}, \mathbf{b})) + (\mathbf{x} - \mathbf{x}_a)^T \mathbf{S}_a^{-1} (\mathbf{x} - \mathbf{x}_a) \quad (1)$$

238 where \mathbf{y} denotes the measured DSCDs, $\mathbf{F}(\mathbf{x}, \mathbf{b})$ is the forward model, \mathbf{b} represents
 239 ancillary meteorological parameters (e.g., temperature, pressure, single-scattering
 240 albedo, and asymmetry factor), \mathbf{x}_a is the a priori state vector, \mathbf{S}_ε is the measurement
 241 error covariance matrix, and \mathbf{S}_a is the a priori covariance matrix. For both aerosols
 242 and trace gases, the a priori vertical profiles were assumed to decrease exponentially

243 with altitude, reflecting the characteristic rapid decay of pollutant concentrations
244 within the planetary boundary layer. Because the absorption of the O₄ is strongly
245 linked to aerosol optical properties, aerosol vertical profiles were first retrieved from
246 multi-elevation O₄ DSCDs and subsequently used as inputs to the forward model for
247 the retrieval of O₃ and HONO profiles. The atmosphere from the surface to 4 km was
248 discretized into 20 layers with a vertical resolution of 200 m (Xing et al., 2024b).
249 Retrievals were subjected to strict quality control: profiles with degrees of freedom
250 (DOF) below 1.0, χ^2 values exceeding 200, or relative uncertainties greater than 50%
251 were excluded from further analysis.

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253 **2.5 TUV model**

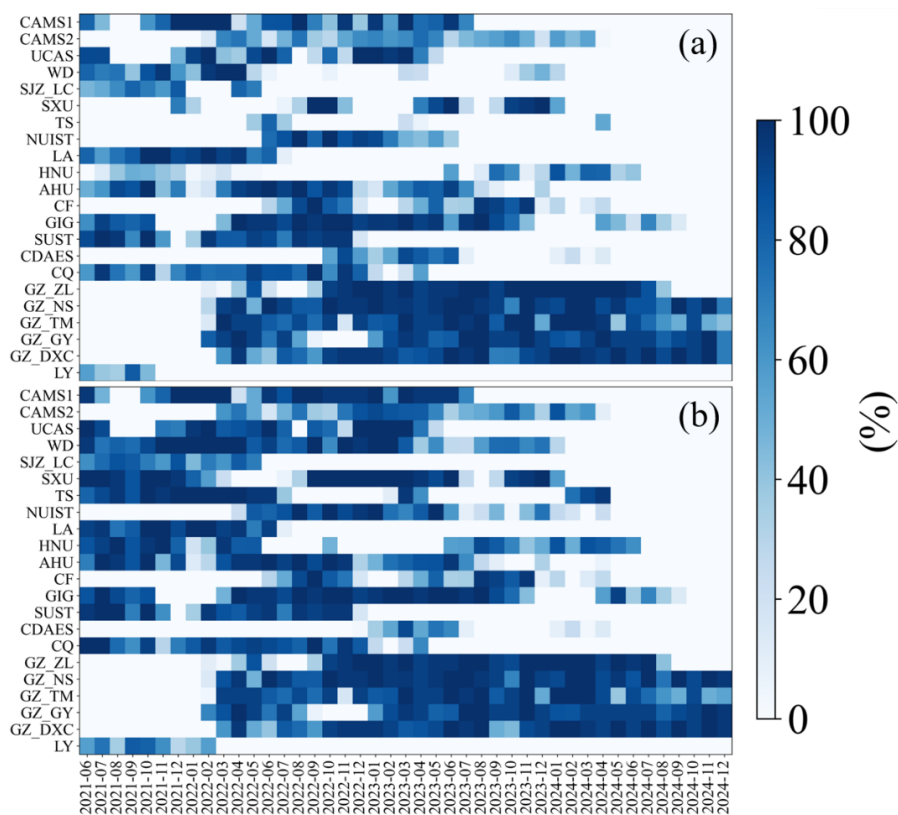
254 Photolysis rates of HONO and O₃ were computed with the Tropospheric
255 Ultraviolet and Visible (TUV) radiative transfer model developed by NCAR, which is
256 based on rigorous radiative transfer theory and implemented in FORTRAN
257 (<https://www2.acom.ucar.edu/modeling/tropospheric>, last access: 26 January 2026).
258 The TUV model simulates the propagation of solar radiation in the troposphere under
259 prescribed optical and chemical conditions and provides spectrally resolved
260 photolysis frequencies for key atmospheric reactions. These rates were used to
261 quantify the contributions of HONO and O₃ photolysis to OH production. Model
262 inputs included AOD at ~361 nm derived from MAX-DOAS-retrieved aerosol
263 extinction profiles, total ozone column from daily TROPOMI observations (typically
264 260–280 DU), and single-scattering albedo (SSA) constrained by regression analyses
265 of O₄ absorptions at 361 and 477 nm (Xing et al., 2019).

266

267 **3. Vertical profile observations of atmospheric composition**

268 Figure 2 summarizes the monthly completeness of O₃ and HONO vertical profile
269 measurements at 22 sites from 2021 to 2024. Shading denotes the fraction of valid
270 observations, with 100% indicating uninterrupted daytime measurements and
271 successful profile retrievals throughout the month. Because stations were
272 commissioned at different times and operated under varying maintenance and field
273 conditions, the available observation periods differ among sites. Most stations provide
274 long, continuous time series of both HONO and O₃. More than 85% of the sites had
275 operational histories spanning over one year, and 60% for more than two years,
276 although these periods may include intermittent data gaps due to maintenance,
277 weather, or technical issues, with only quality-controlled valid profiles retained in the
278 dataset, demonstrating the temporal stability of the network. This coverage enables
279 robust characterization of seasonal and diurnal variability under diverse climatic
280 regimes and emission backgrounds. Although a few sites had shorter operational
281 periods owing to instrument commissioning and field constraints, they still delivered
282 several months of continuous high-quality data, which are valuable for regional
283 intercomparison and support analyses of long-term trends and photochemical
284 processes. Isolated months with missing or incomplete data occur at some sites,

285 primarily because of unavoidable factors such as instrument maintenance, power
 286 interruptions, persistent cloud or precipitation, and quality-control filtering (e.g.,
 287 excessive fitting residuals or low DOF in the retrievals).

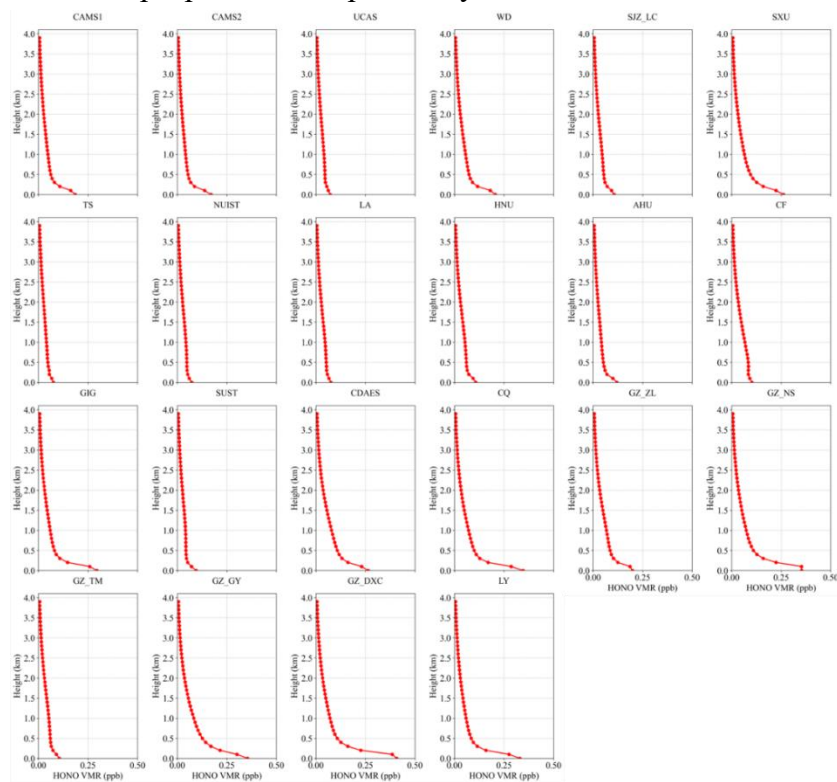


288
 289 Figure 2. Monthly data completeness of the vertical profiles of (a) O₃ and (b) HONO.
 290

291 3.1 HONO

292 Figure 3 presents the 2021–2024 mean vertical profiles of HONO across all sites.
 293 At every location, HONO is strongly enriched near the surface and decreases rapidly
 294 with height, following an approximately exponential decay. This structure is
 295 characteristic of a boundary-layer-dominated species controlled by ground-based
 296 sources (Li et al., 2025b; Meng et al., 2020; Xing et al., 2024c; Xu et al., 2021). Peak
 297 mixing ratios occur within the lowest 0–0.5 km, decline sharply between 0.5 and 1.5
 298 km, and generally fall to regional background or near the detection limit above 2 km
 299 (<0.05–0.1 ppb), becoming negligible by 4 km. Such steep gradients reflect dominant
 300 near-surface emissions and nocturnal heterogeneous formation of HONO from NO₂
 301 on ground and aerosol surfaces, combined with its short photochemical lifetime and
 302 rapid daytime photolysis, which preclude sustained accumulation in the free
 303 troposphere (Li et al., 2025b; Meng et al., 2020; Xing et al., 2024a). Pronounced
 304 regional contrasts are evident. Urban sites in North and East China (e.g., CAMS1,
 305 CAMS2, WD, SXU, AHU) exhibit the highest near-surface HONO (0.3–0.5 ppb
 306 below 0.3 km), followed by a rapid decrease to <0.1 ppb above 1 km. The sharp
 307 vertical gradients and absence of secondary maxima aloft indicate strong control by
 308 surface sources and nocturnal heterogeneous production, with efficient removal by

309 turbulent mixing and photolysis within the planetary boundary layer (Xu et al., 2021).
 310 In contrast, background or relatively clean sites (e.g., TS, LA) show much lower
 311 concentrations, with near-surface values typically <0.2 ppb and a monotonic decrease
 312 with altitude, consistent with weak local emissions and dominance of regional
 313 background (Garcia-Nieto et al., 2018b; Li et al., 2025b). Sites in South and
 314 Southwest China (e.g., GZ_ZL, GZ_NS, GZ_DXC, CQ, CDAES) display a similar
 315 monotonic decay: elevated HONO confined to the lowest 0–0.5 km and rapid
 316 attenuation to background levels above 1–2 km, without a distinct mid-level
 317 enhancement. Although near-surface mixing ratios at some locations (e.g., GZ_DXC,
 318 CQ) approach or slightly exceed 0.3 ppb, their vertical decay rates are comparable to
 319 those at northern and eastern urban sites. This indicates that, even under high
 320 humidity or complex topography, HONO remains largely restricted to the lower
 321 boundary layer, governed by its short lifetime, fast photolysis, and dilution by
 322 convective mixing, while large-scale vertical transport contributes little to its
 323 maintenance aloft (Li et al., 2025b; Xing et al., 2021b; Xu et al., 2021). Seasonal
 324 mean profiles are shown in Figures S1–S4. Taken together, the regionally averaged
 325 profiles consistently demonstrate strong near-surface accumulation and rapid vertical
 326 attenuation of HONO. This confirms that HONO is a short-lived, boundary-layer-
 327 derived reactive nitrogen species, tightly coupled to surface emissions and
 328 heterogeneous chemistry. It therefore plays a key role in initiating early-morning OH
 329 production and regulating boundary-layer oxidizing capacity, whereas its direct
 330 impact in the free troposphere is comparatively minor.

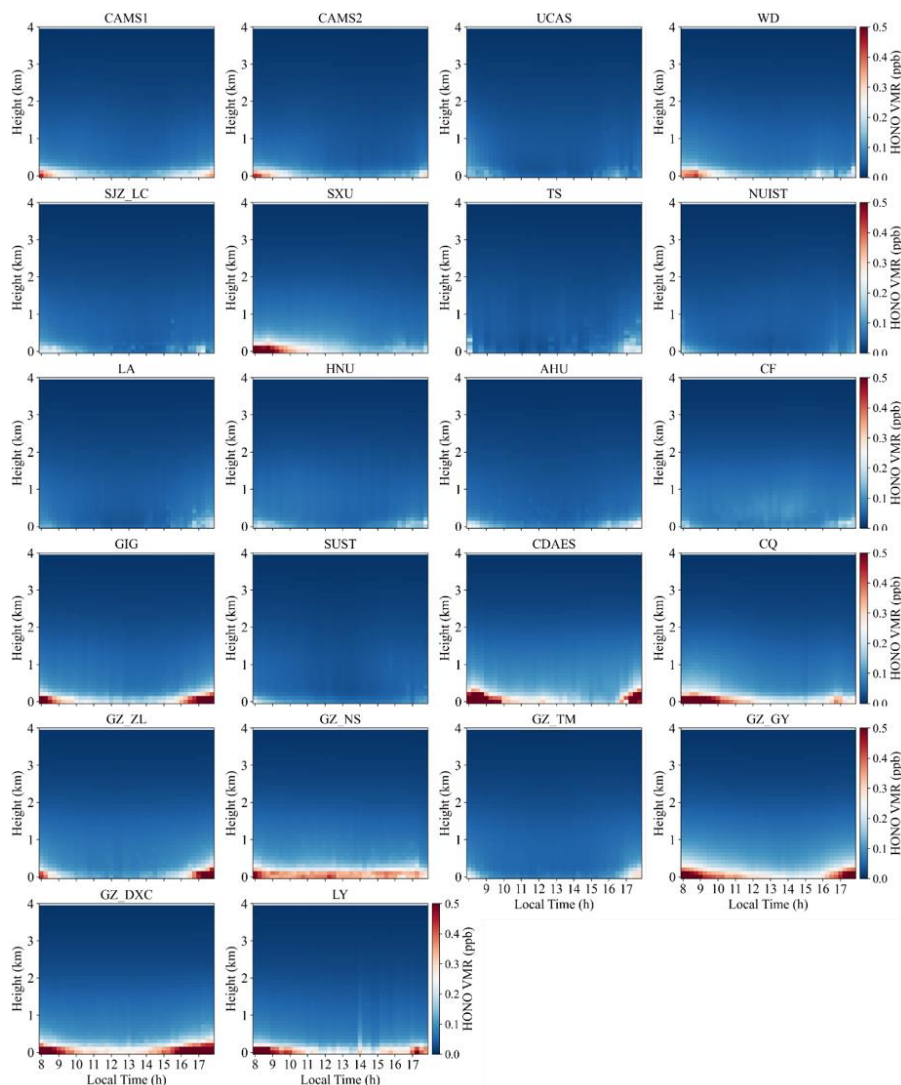


331 Figure 3. Mean vertical profiles of HONO averaged over 2021–2024.

332 Figure 4 illustrates the mean diurnal evolution of HONO. As a key reactive
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334 nitrogen species, HONO exhibits vertical and temporal patterns that integrate the
335 effects of surface emissions, heterogeneous and photochemical processes, and
336 boundary-layer dynamics. Based on HONO data obtained from the hyperspectral
337 vertical remote-sensing network, pronounced regional and site-specific patterns are
338 observed. Across North China (CAMS1, CAMS2, UCAS, WD, SJZ_LC and SXU),
339 HONO exhibits a clear near-surface morning maximum followed by an afternoon
340 minimum. At CAMS1 and CAMS2, the 0–1 km volume mixing ratio (VMR) peaks at
341 08:00–10:00 local time (0.3–0.4 ppb) and decreases to 0.1–0.2 ppb by 14:00–16:00.
342 This pattern reflects nocturnal accumulation driven by heterogeneous conversion of
343 NO₂ on aerosol and ground surfaces (Liu et al., 2022a; Xing et al., 2023; Xuan et al.,
344 2025b); after sunrise, enhanced solar radiation leads to the release and photochemical
345 processing of HONO; meanwhile, morning rush-hour emissions of NO₂ and VOCs
346 further promote HONO formation (Garcia-Nieto et al., 2018a; Zhang et al., 2025a). At
347 the mountain site SXU, topography-induced temperature inversions enhance
348 nighttime accumulation, yielding a more pronounced morning peak. In contrast,
349 UCAS and WD, characterized by weaker anthropogenic emissions, show lower near-
350 surface HONO levels and smaller diurnal amplitudes. In East China (TS, NUIST, LA,
351 HNU, AHU and CF), urban sites display modest morning enhancements (0.2–0.3 ppb
352 at 08:00–10:00) followed by afternoon decreases driven by boundary-layer growth
353 and photolysis. At the high-altitude TS site (1500 m), HONO remains below 0.1 ppb
354 with weak diurnal variability, reflecting clean background conditions and efficient
355 vertical mixing. Sites with dense vegetation or agricultural land use (LA and CF) may
356 receive contributions from biogenic VOC-related chemistry, but the overall pattern
357 still features a subdued morning maximum (Liang et al., 2017; Ryan et al., 2018a; Xue
358 et al., 2021; Ye et al., 2023a). At South China and Southwest China sites (GIG, SUST,
359 CDAES, CQ and the Guangzhou cluster: GZ_ZL, GZ_NS, GZ_TM, GZ_GY and
360 GZ_DXC), warm and humid conditions together with basin or coastal circulations
361 further modulate the diurnal cycle. Urban stations typically reach 0.3–0.5 ppb near the
362 surface in the morning and decline to 0.1–0.2 ppb in the afternoon. In the Sichuan
363 Basin (CQ), strong nocturnal inversions favour HONO accumulation, producing
364 slightly higher morning peaks (0.4–0.5 ppb). At coastal sites, land–sea breeze
365 circulation leads to a transient morning enhancement followed by dilution by cleaner
366 marine air masses. At the Central China site LY, the diurnal pattern resembles that in
367 North and South China, with a clear morning maximum and lower concentrations in
368 the afternoon associated with boundary-layer development. Seasonal mean diurnal
369 vertical profiles are shown in Figures S5–S8. Overall, the diurnal cycle of HONO is
370 governed by three coupled processes: (i) nocturnal heterogeneous production from
371 NO₂ on aerosol and surface substrates, which drives early-morning maxima (Li et al.,
372 2025b; Meng et al., 2020; Xuan et al., 2024); (ii) enhancement by morning
373 anthropogenic emissions of NO_x and VOCs from traffic and industrial activities (Hao
374 et al., 2020; Zhang et al., 2025a, 2023b); and (iii) rapid photolysis and boundary-layer
375 dilution in the afternoon (Xing et al., 2021b, 2024a; Zhang et al., 2023b). Regional

376 contrasts arise from the combined effects of emission intensity, topography (basin,
 377 mountain and coastal settings), and meteorological conditions, particularly
 378 temperature inversions and ventilation efficiency (Li et al., 2025b; Xuan et al., 2024;
 379 Zhang et al., 2025a).



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Figure 4. Mean diurnal vertical profiles of HONO for 2021–2024.

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383 3.2 O₃

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Figure 5 presents the mean vertical profiles of O₃ averaged over 2021–2024 for all sites. A consistent “low near the surface–high aloft” structure is observed, characterized by a monotonic increase or a weak S-shaped pattern. O₃ VMR are lowest in the lowest 0–0.5 km (20–60 ppb), rise rapidly between 1 and 2 km, and reach daytime maxima at 3–4 km (60–100 ppb). This vertical gradient agrees well with MAX-DOAS and ozone-sonde observations over eastern China and other regions worldwide (Couillard et al., 2021; Ji et al., 2023; Liao et al., 2024; Su et al., 2017; Wang et al., 2018; Zeng et al., 2023), and reflects the combined effects of strong near-surface NO₂ titration, dry deposition, and boundary-layer mixing that

393 suppress O₃, together with photochemical production and regional transport that
394 enhance O₃ aloft (Couillard et al., 2021; Donzelli and Suarez-Varela, 2024; Liao et al.,
395 2024; Zeng et al., 2023; Zhu et al., 2025b). Within the boundary layer (0–1 km), O₃
396 generally increases sharply with height, and a weak local maximum or inflection is
397 often found at 0.2–0.5 km. This contrasts with the vertical distributions of NO₂ and
398 HCHO at the same sites, which show high near-surface concentrations dominated by
399 emissions (Couillard et al., 2021; Hu et al., 2024; Hong et al., 2022; Jiao et al., 2025;
400 Liu et al., 2023b). In contrast, O₃ is efficiently removed near the ground by nocturnal
401 NO₂ titration and daytime surface deposition (Liao et al., 2024; Xing et al., 2022). At
402 urban and suburban stations (e.g., UCAS and CF), O₃ in the lowest 0–0.3 km can
403 decrease to 20–40 ppb, indicating strong titration by traffic and industry related NO₂
404 (Hu et al., 2024). Between 1 and 3 km, O₃ increases nearly monotonically at most
405 sites, with the largest vertical gradient typically occurring around 2–3 km. This layer
406 often corresponds to the daytime boundary-layer top or the nocturnal residual layer
407 and represents a key altitude for regional photochemical accumulation and downward
408 transport (He et al., 2023b; Liao et al., 2024; Zhu et al., 2025a). Numerous studies
409 have shown that O₃-rich air in the upper boundary layer and residual layer can be
410 mixed downward during boundary-layer growth, and that O₃ stored aloft at night is re-
411 entrained to the surface the following morning, making an important contribution to
412 surface O₃ levels (Ancellet et al., 2024; Donzelli and Suarez-Varela, 2024; Liu et al.,
413 2022b; Shi et al., 2022; Song et al., 2024; Wang et al., 2024b). At 3–4 km, O₃ VMR
414 further increase and tend to level off, with some sites exhibiting distinct maxima. At
415 these altitudes, the influence of surface NO₂ titration becomes negligible, whereas
416 long-range transport and possible stratosphere–troposphere exchange start to play a
417 role. Previous studies have shown that enhanced O₃ at 3–5 km over East Asia in
418 spring and summer can partly arise from stratospheric intrusions and westerly long-
419 range transport (Li et al., 2025a; Liao et al., 2024, 2025; Park et al., 2020). The
420 pronounced O₃ enhancements observed at 3–4 km at sites such as CQ, GZ_TM and
421 SUST are therefore likely linked to free-tropospheric background O₃ and regional-
422 scale transport processes. Seasonal mean O₃ vertical profiles are shown in Figures
423 S9–S12.

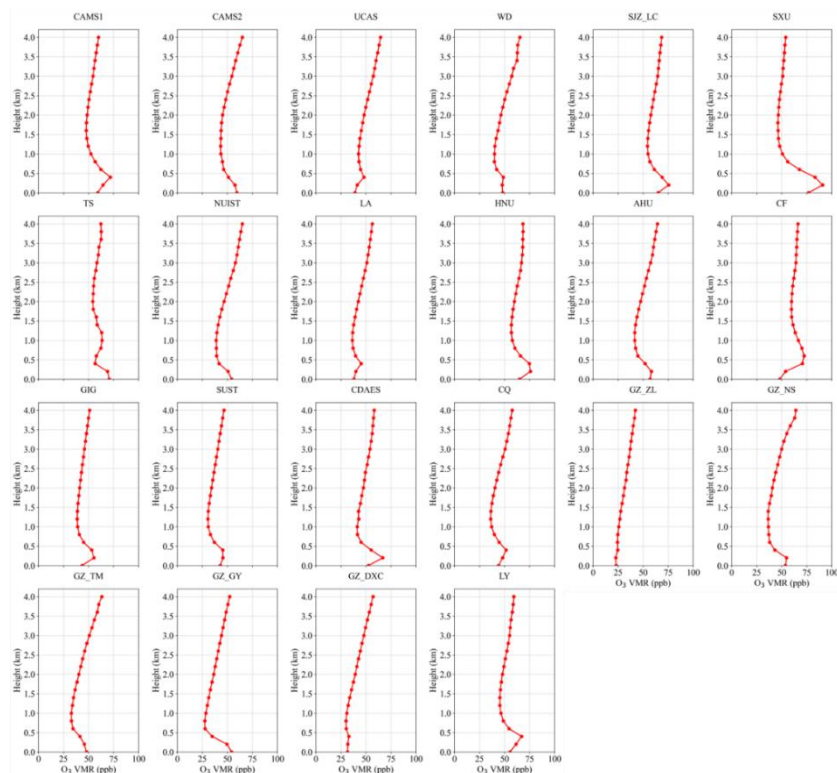


Figure 5. Mean vertical profiles of O₃ averaged over 2021–2024.

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Figure 6 presents the mean diurnal evolution of O₃. All sites exhibit pronounced daily cycles with clear regional contrasts. O₃ typically peaks in the morning (08:00–10:00) or in the early afternoon (12:00–15:00), in phase with the diurnal variation of solar irradiance and photochemical reaction rates (Xia et al., 2021; Yang et al., 2020). This behaviour is most evident in North and East China, whereas the cycle is weaker in South China, likely owing to persistently high temperature and humidity that modulate boundary-layer development and photochemistry (Zhou et al., 2022). At several sites (e.g., TS, CDAES and CQ), enhanced O₃ at 1–2 km during 12:00–15:00 points to the influence of local meteorology and emission distributions (Chen et al., 2023; Li et al., 2025a). In contrast, morning maxima at CAMS1, CAMS2, UCAS, NUIST and AHU reflect the rapid re-entrainment and photochemical processing of O₃ accumulated overnight after sunrise (David and Nair, 2011; Liao et al., 2023). The vertical structure of the diurnal cycle also differs markedly among regions. North China sites show strong near-surface variability, whereas peak O₃ in South China is generally lower, consistent with regional differences in pollution levels and meteorological conditions. Relatively high near-surface O₃ at GZ_ZL and GZ_NS is likely linked to local emissions combined with weak dispersion (Yang et al., 2020; Zhou et al., 2022). North China stations (CAMS1, CAMS2, UCAS, WD, SJZ_LC and SXU) display a typical urban O₃ diurnal pattern. At CAMS1 and CAMS2, O₃ in the 0–1 km layer reaches 80–120 ppb in the morning (08:00–10:00) and decreases markedly in the early afternoon, reflecting rapid boundary-layer growth and photochemical loss after sunrise (David and Nair, 2011; Liao et al., 2023). UCAS and WD show similar morning maxima, whereas SJZ_LC is characterized by lower and

449 more stable O₃, indicative of relatively clean background conditions. At SXU, high
450 morning O₃ (80–100 ppb) is followed by even higher afternoon levels (>100 ppb),
451 pointing to strong in situ secondary production under intense photochemical activity
452 (Wang et al., 2017; Xia et al., 2021). East China sites (TS, NUIST, LA, HNU, AHU
453 and CF) exhibit more complex diurnal behaviour. At TS, O₃ peaks at 1–2 km during
454 12:00–15:00 (80–100 ppb), suggesting an important role of vertical transport and
455 local emissions. NUIST and AHU show morning maxima similar to those in North
456 China, whereas LA maintains low and weakly varying O₃, consistent with relatively
457 clean conditions (Chen et al., 2024). At HNU, near-surface O₃ increases in the early
458 afternoon (60–80 ppb), reflecting active photochemistry (Wang et al., 2025c). CF
459 shows a pronounced afternoon peak (13:00–17:00, 80–120 ppb), indicating a strong
460 influence of local sources (Xia et al., 2021; Yang et al., 2020). South China sites (GIG,
461 SUST, GZ_ZL, GZ_NS, GZ_TM, GZ_GY and GZ_DXC) differ substantially from
462 those in the north and east. GIG exhibits low and weakly varying O₃, representative of
463 background conditions (Chen et al., 2024; Lin et al., 2022). The other sites show
464 morning near-surface maxima (80–100 ppb at 08:00–10:00), followed by decreases
465 associated with rapid boundary-layer development after sunrise (David and Nair, 2011;
466 Liao et al., 2023), and enhanced O₃ at 3–4 km in the afternoon (13:00–17:00),
467 highlighting the pronounced vertical structure of O₃ pollution in this region.
468 Southwestern sites (CDAES and CQ) display distinct afternoon enhancements at 1–2
469 km. At CDAES, O₃ reaches 80–120 ppb during 15:00–18:00, likely favored by high
470 temperature and humidity that accelerate photochemical production (Yang et al., 2020;
471 Zhang et al., 2022), while CQ shows a similar but weaker enhancement (60–80 ppb).
472 The central China site LY exhibits morning near-surface maxima (60–80 ppb) and
473 elevated O₃ at 2–4 km in the afternoon, characteristic of a typical urban diurnal cycle.
474 Seasonal mean diurnal vertical profiles are shown in Figures S13–S16. These regional
475 contrasts underline the differing controls on O₃ across China, with strong local
476 photochemistry in North China, combined regional transport and sustained
477 photochemical production in South China, and mixed influences of emissions and
478 meteorology in East China.

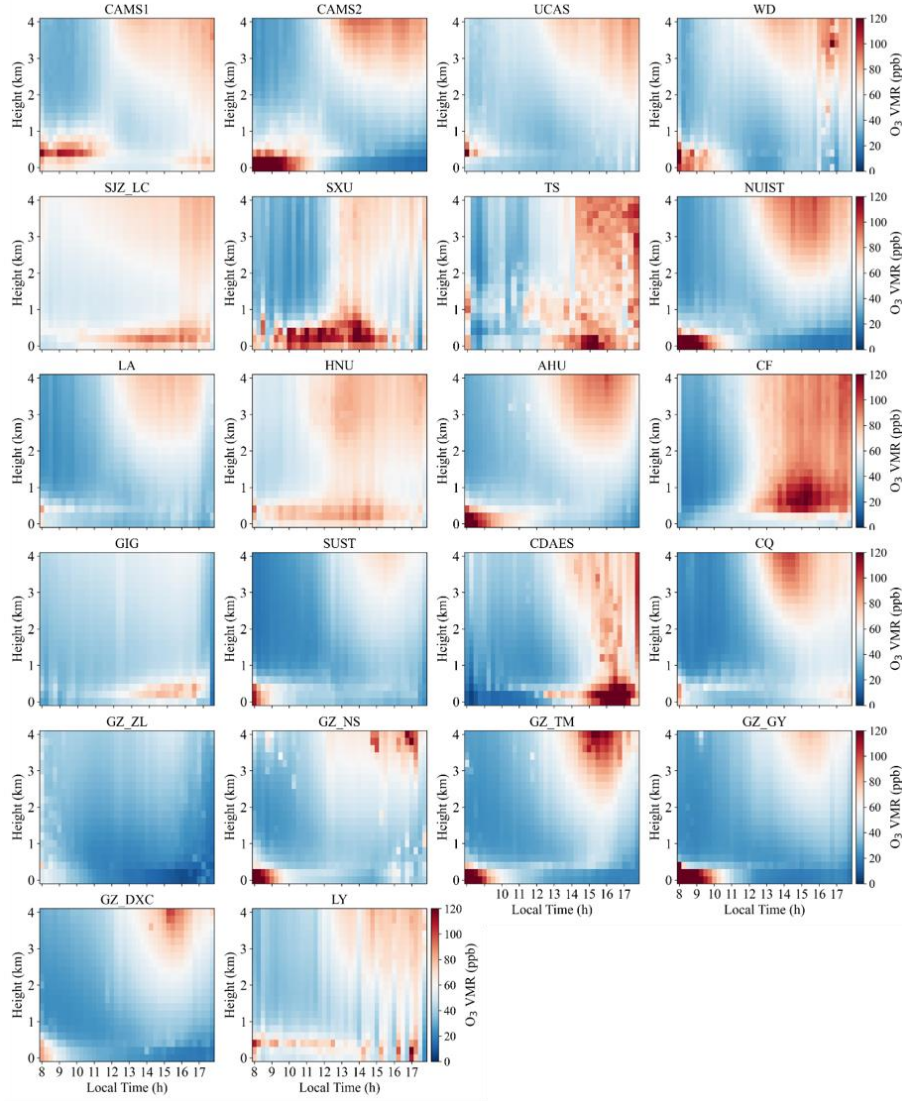


Figure 6. Mean diurnal vertical profiles of O₃ for 2021–2024.

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482 3.3 OH production

483 Photolysis of HONO and O₃ constitutes a primary source of OH radicals and
 484 therefore controls the atmospheric oxidation capacity (AOC). To quantify the AOC at
 485 each site, we evaluated altitude-resolved OH production from HONO and O₃ using
 486 retrieved profiles combined with photolysis frequencies calculated by the TUV model.
 487 OH production from HONO and O₃ was computed from the following expressions.

$$488 \quad P(OH)_{HONO} = J(HONO) \times [HONO] \quad (2)$$

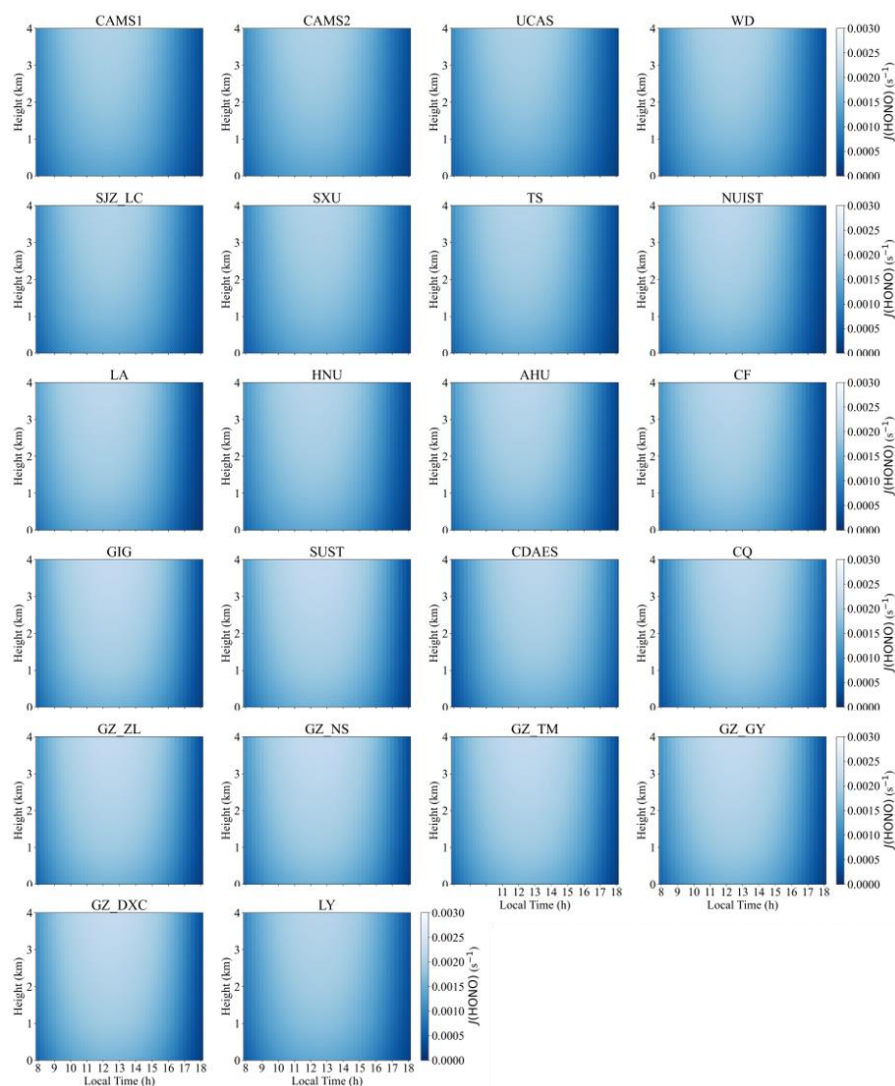
$$489 \quad P(OH)_{O_3} = 2 \times f \times J(O(^1D)) \times [O_3] \quad (3)$$

490 Here, J(HONO) and J(O(¹D)) are the photolysis rate coefficients of HONO and O₃,
 491 respectively, obtained from the TUV model. O(¹D) denotes electronically excited
 492 atomic oxygen produced by O₃ photodissociation, and *f* represents the branching
 493 fraction of the reaction O(¹D) + H₂O → 2OH. [HONO] and [O₃] are the

494 concentrations of HONO and O₃ at each altitude level.

495 Figure 7 presents the mean diurnal vertical profiles of the HONO photolysis
496 frequency, J(HONO), at 22 sites during 2021–2024; the corresponding seasonal mean
497 diurnal variations are presented in Figures S17–S20. All sites exhibit a canonical
498 photochemical pattern: J(HONO) increases rapidly after sunrise, reaches a maximum
499 around local noon, and then gradually decreases with increasing solar zenith angle.
500 Elevated values persist between 10:00 and 14:00 local time, with peak J(HONO)
501 typically occurring near 12:00–13:00, indicating that HONO photolysis is primarily
502 controlled by solar irradiance, in agreement with observations in Beijing and
503 Guangzhou (He et al., 2023c; Ryan et al., 2018b). Vertically, J(HONO) increases
504 systematically with altitude. Photolysis rates are relatively low in the near-surface
505 layer (0–0.5 km), increase markedly in the upper mixed layer and lower free
506 troposphere (approximately 1–3 km), and reach maxima between 2 and 4 km, with
507 peak values around $2.5 \times 10^{-3} \text{ s}^{-1}$. This “weaker near the surface and stronger aloft”
508 structure is highly consistent with the J(HONO) profiles reported by Xing et al.
509 (2024a) and reflects the combined effects of aerosol attenuation of ultraviolet
510 radiation in the lower atmosphere and enhanced shortwave actinic flux at higher
511 altitudes (He et al., 2023c; Ryan et al., 2018b; Spataro and Ianniello, 2014). At the
512 North China sites (CAMS1, CAMS2, UCAS, WD, SJZ_LC, SXU), J(HONO) exhibits
513 a pronounced diurnal cycle, increasing after sunrise with rising solar radiation,
514 peaking at midday (12:00–14:00) at 0.0020–0.0025 s⁻¹, and decreasing in the
515 afternoon (14:00–18:00). The urban Beijing sites CAMS1 and CAMS2 show peak
516 values of $\sim 0.0025 \text{ s}^{-1}$, comparable to those at other North China Plain stations (e.g.,
517 UCAS and WD), reflecting strong photolysis under high HONO loading and
518 favourable radiation conditions. At SJZ_LC, located at the foothills of the Taihang
519 Mountains, morning J(HONO) is slightly enhanced, likely owing to temperature
520 inversions that modulate the vertical distribution of aerosols and actinic flux. The
521 elevated site SXU (780 m a.s.l.) exhibits systematically higher J(HONO) than lowland
522 stations, with a peak of $\sim 0.0022 \text{ s}^{-1}$, consistent with reduced aerosol extinction and
523 stronger solar radiation at higher altitude. East China sites (TS, NUIST, LA, HNU,
524 AHU, CF) display similar peak timing to North China but slightly lower magnitudes
525 (0.0015–0.0025 s⁻¹). For example, J(HONO) at NUIST peaks at $\sim 0.0020 \text{ s}^{-1}$, whereas
526 the high-altitude background site TS (1500 m a.s.l.) reaches $\sim 0.0021 \text{ s}^{-1}$, consistent
527 with enhanced actinic flux under cleaner atmospheric conditions. In South China
528 (GIG, SUST, GZ_ZL, GZ_NS, GZ_TM, GZ_GY, GZ_DXC), the maximum J(HONO)
529 occurs slightly later in the day (13:00–15:00) and attains higher values (0.0020–
530 0.0030 s⁻¹). The highest peak is observed at GZ_DXC ($\sim 0.0030 \text{ s}^{-1}$), likely reflecting
531 elevated HONO concentrations promoted by warm and humid conditions that favor
532 heterogeneous formation. The southwestern basin site CQ shows a comparable peak
533 ($\sim 0.0025 \text{ s}^{-1}$), while the Central China site LY reaches $\sim 0.0020 \text{ s}^{-1}$, similar to values in
534 North and East China. Overall, urban sites exhibit larger diurnal amplitudes and 20–
535 40% higher J(HONO) maxima than mountain or clean-background sites, owing to

536 higher HONO abundances and aerosol loading that modulate the effective actinic flux.
 537 This behaviour is fully consistent with previous findings from Beijing, Shanghai and
 538 other megacities, which reported pronounced daytime enhancement of $J(\text{HONO})$
 539 under high- NO_2 and high-HONO conditions (He et al., 2023c; Spataro and Ianniello,
 540 2014; Ye et al., 2023b).

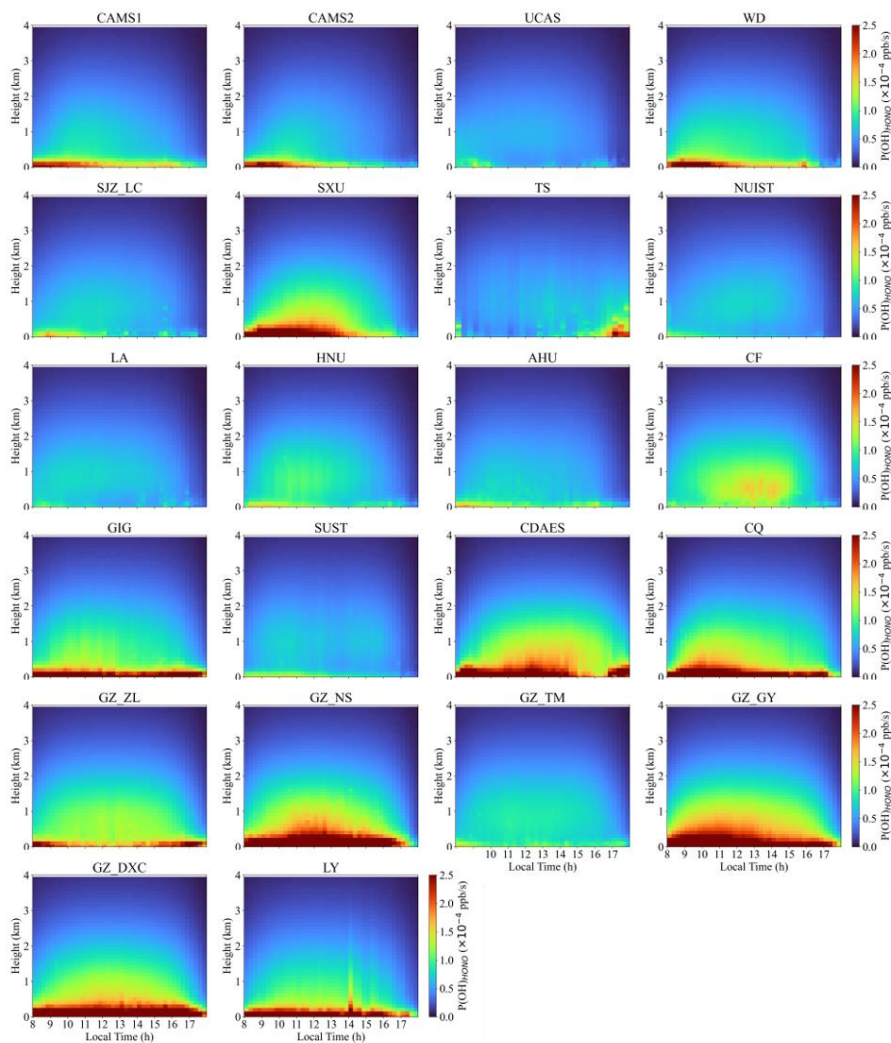


541

542 Figure 7. Mean diurnal vertical profiles of the HONO photolysis rate, $J(\text{HONO})$.

543 Figure 8 presents the mean diurnal vertical profiles of OH production from
 544 HONO photolysis, $P(\text{OH})_{\text{HONO}}$, at 22 sites; the corresponding seasonal mean profiles
 545 are shown in Figures S21–S24. At all sites, $P(\text{OH})_{\text{HONO}}$ displays a pronounced
 546 unimodal diurnal cycle, increasing rapidly after sunrise, peaking between 10:00 and
 547 14:00, and declining thereafter. The peak timing closely follows the maximum of the
 548 $J(\text{HONO})$, whereas the peak altitude remains confined to the near-surface layer,
 549 reflecting the strong surface enhancement of HONO. In the lower boundary layer (0–
 550 0.5 km), $P(\text{OH})_{\text{HONO}}$ attains its column maximum, with most sites peaking between
 551 11:00 and 13:00 and reaching 1.0×10^{-4} – 5.5×10^{-4} $\text{ppb} \cdot \text{s}^{-1}$. For all stations, $P(\text{OH})_{\text{HONO}}$
 552 is largest within 0–1 km and decreases monotonically with height, consistent with the

553 preferential accumulation of HONO near the surface and the resulting localization of
554 photochemically produced OH (He et al., 2023c; Li et al., 2025b; Xing et al., 2021b;
555 Zhang et al., 2025a). Several sites, including SXU, CDAES, CQ, GZ_NS, GZ_GY,
556 and GZ_DXC, exhibit particularly strong OH production, with peak $P(\text{OH})_{\text{HONO}}$
557 commonly exceeding $3.0 \times 10^{-4} \text{ ppb} \cdot \text{s}^{-1}$. This reflects the combined effects of elevated
558 HONO levels and intense solar radiation. At these locations, the high- $P(\text{OH})_{\text{HONO}}$
559 layer can extend to 1–2 km, indicating a deeper photochemically active region. This
560 feature is consistent with earlier reports highlighting the substantial contribution of
561 HONO to OH in the lower free troposphere (Aumont et al., 2003; R. Crilley et al.,
562 2016; Xue et al., 2025; Zhang et al., 2025a). Vertically, $P(\text{OH})_{\text{HONO}}$ decreases rapidly
563 with altitude at all sites and is reduced to 20–40% of its surface value above 2 km,
564 demonstrating that the impact of HONO photolysis on OH is largely confined to the
565 boundary layer. In agreement with previous observations in Beijing and Guangzhou
566 (Gu et al., 2022; Meng et al., 2020; Yu et al., 2022), HONO photolysis represents one
567 of the dominant OH sources during the morning and around local noon, accounting
568 for 30–60% of the daytime OH production near the surface (Song et al., 2023a; Tang
569 et al., 2015). In the present study, several plateau sites show even larger relative
570 contributions at midday, indicating that under conditions of low NO_2 and strong solar
571 irradiance, HONO photolysis becomes an especially efficient radical source,
572 consistent with findings at Nam Co (Xing et al., 2024b). Regionally, North China sites
573 (CAMS1, CAMS2, UCAS, WD, SJZ_LC, and SXU) exhibit near-surface (0–1 km)
574 $P(\text{OH})_{\text{HONO}}$ maxima between 12:00 and 14:00 local time, with values of 1.0×10^{-4} –
575 $3.0 \times 10^{-4} \text{ ppb} \cdot \text{s}^{-1}$. At CAMS1, the peak reaches $\sim 1.5 \times 10^{-4} \text{ ppb} \cdot \text{s}^{-1}$, whereas at SJZ_LC,
576 although nocturnal temperature inversions near the Taihang Mountains may favour
577 HONO accumulation, the peak remains modest ($\sim 1.2 \times 10^{-4} \text{ ppb} \cdot \text{s}^{-1}$) owing to weaker
578 local emissions, comparable to UCAS and WD (1.0×10^{-4} – $2.5 \times 10^{-4} \text{ ppb} \cdot \text{s}^{-1}$). At these
579 sites, $P(\text{OH})_{\text{HONO}}$ declines sharply with height and is substantially reduced above 1
580 km, underscoring the near-surface confinement of both HONO and OH production
581 from its photolysis. East China stations (TS, NUIST, LA, HNU, AHU, and CF) show
582 similar peak times (12:00–14:00) but slightly lower magnitudes (1.0×10^{-4} – 1.5×10^{-4}
583 $\text{ppb} \cdot \text{s}^{-1}$). In South China (GIG, SUST, GZ_ZL, GZ_NS, GZ_TM, GZ_GY, and
584 GZ_DXC), peaks occur later (13:00–15:00) and are substantially higher (2.5×10^{-4} –
585 $5.5 \times 10^{-4} \text{ ppb} \cdot \text{s}^{-1}$), consistent with enhanced heterogeneous HONO formation under
586 warm and humid conditions. Southwest China sites (CDAES and CQ) reach peak
587 values of $\sim 3.5 \times 10^{-4} \text{ ppb} \cdot \text{s}^{-1}$, comparable to those in South China, likely owing to
588 basin-induced HONO accumulation and vigorous photochemistry. In Central China
589 (LY), the peak ($\sim 2.7 \times 10^{-4} \text{ ppb} \cdot \text{s}^{-1}$) is similar to that in North and East China,
590 indicating broadly comparable HONO sources and photolysis efficiencies across these
591 regions.

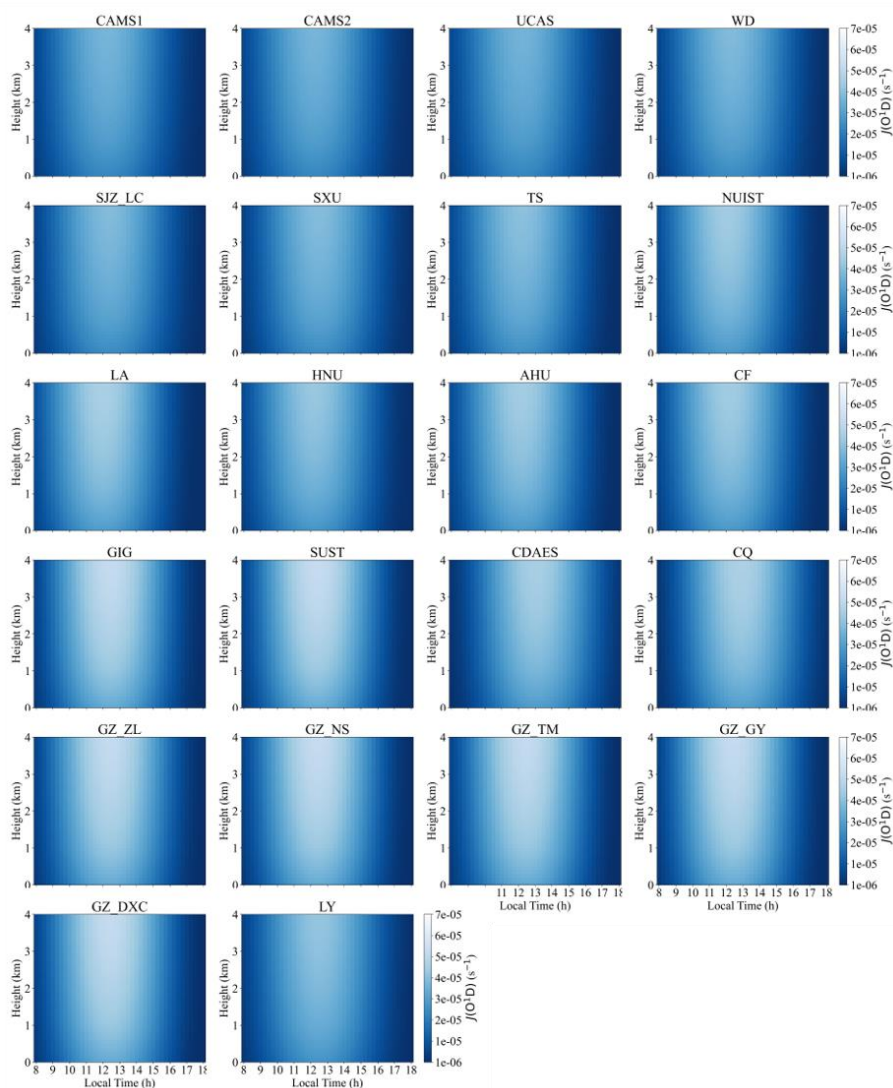


592

593 Figure 8. Mean vertical profiles of OH radicals generated by HONO photolysis.

594 Figure 9 presents the O_3 photolysis frequency, $J(O(^1D))$, at all 22 sites follows a
 595 pronounced diurnal cycle, with maxima consistently occurring between 12:00 and
 596 14:00 local time. Seasonal mean diurnal variations are presented in Figures S25–S28.
 597 In North China (CAMS1, CAMS2, UCAS, WD, SJZ_LC, and SXU), near-surface
 598 peak $J(O(^1D))$ ranges from $\sim 5 \times 10^{-5}$ to $7 \times 10^{-5} \text{ s}^{-1}$. The urban sites CAMS1, CAMS2,
 599 UCAS, and WD reach the highest values ($\sim 6 \times 10^{-5}$ – $7 \times 10^{-5} \text{ s}^{-1}$) at midday. At SJZ_LC,
 600 located at the foothills of the Taihang Mountains, nocturnal temperature inversions
 601 can favor O_3 accumulation (Guo et al., 2024b; He et al., 2021), but the peak remains
 602 slightly lower ($\sim 5 \times 10^{-5}$ – $6 \times 10^{-5} \text{ s}^{-1}$), likely constrained by local emissions. At the
 603 higher-altitude SXU site (780 m a.s.l.), near-surface $J(O(^1D))$ is reduced ($\sim 4 \times 10^{-5}$ –
 604 $5 \times 10^{-5} \text{ s}^{-1}$). Overall, urban stations exhibit larger $J(O(^1D))$ than suburban and rural
 605 sites, reflecting higher O_3 levels driven by anthropogenic precursors and consistent
 606 with reported regional contrasts (Fardilah et al., 2023; Guo et al., 2024a; Qiu et al.,
 607 2025). In East China (TS, NUIST, LA, HNU, AHU, and CF), near-surface $J(O(^1D))$
 608 peaks at $\sim 4 \times 10^{-5}$ – $6 \times 10^{-5} \text{ s}^{-1}$, with urban sites such as NUIST and AHU reaching
 609 $\sim 5 \times 10^{-5}$ – $6 \times 10^{-5} \text{ s}^{-1}$ at noon. In contrast, the high-altitude TS site (1500 m a.s.l.)

610 shows lower values ($\sim 3 \times 10^{-5}$ – $4 \times 10^{-5} \text{ s}^{-1}$), consistent with its lower O₃ burden and
 611 cleaner background conditions. South China stations (GIG, SUST, GZ_ZL, GZ_NS,
 612 GZ_TM, GZ_GY, and GZ_DXC) display slightly higher peak J(O(¹D))) ($\sim 6 \times 10^{-5}$ –
 613 $7 \times 10^{-5} \text{ s}^{-1}$), in line with enhanced O₃ production under warm and humid subtropical
 614 conditions (Lu et al., 2025; Song et al., 2026; Zhang et al., 2025b). In Southwest
 615 China (CDAES and CQ), peak values ($\sim 5 \times 10^{-5}$ – $6 \times 10^{-5} \text{ s}^{-1}$) are comparable to those in
 616 South China, likely driven by basin topography that favors O₃ accumulation and
 617 vigorous photochemistry (Qiao et al., 2019; Shu et al., 2023; Wang et al., 2024a). The
 618 Central China site LY exhibits slightly lower peaks ($\sim 4 \times 10^{-5}$ – $5 \times 10^{-5} \text{ s}^{-1}$), similar to
 619 North and East China, indicating broadly comparable O₃ sources and photolysis
 620 efficiencies across these regions.



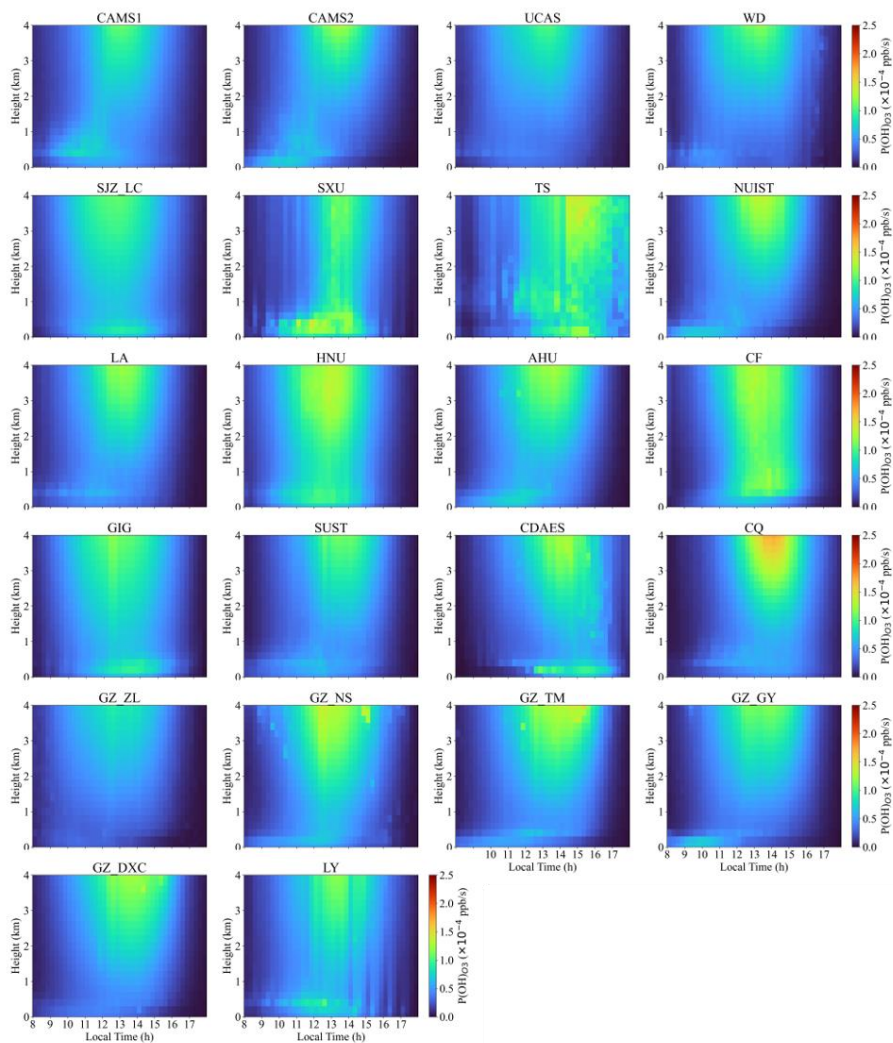
621

622 Figure 9. Mean diurnal vertical profiles of the O₃ photolysis rate, J(O(¹D))).

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624 Figure 10 presents the mean diurnal vertical profiles of the OH production rate
 625 from ozone photolysis, P(OH)_{O₃}, at the 22 sites, and seasonal mean diurnal variations
 626 are presented in Figures S29–S32. All sites exhibit a pronounced unimodal diurnal
 cycle, with P(OH)_{O₃} increasing rapidly after sunrise, peaking between 12:00 and

627 14:00 local time, and declining thereafter. The vertical location of the maxima varies
628 markedly among sites: at some, enhanced production is confined to the near-surface
629 layer (0–0.5 km), whereas at others distinct maxima occur at 3–4 km, indicating
630 substantial regional differences in photochemical regimes. Peak $P(OH)O_3$ spans
631 0.5×10^{-4} – 2.0×10^{-4} $ppb \cdot s^{-1}$ across the network. In North China (CAMS1, CAMS2,
632 UCAS, WD, SJZ_LC, and SXU), near-surface $P(OH)O_3$ during 12:00–14:00 reaches
633 1.0×10^{-4} – 2.0×10^{-4} $ppb \cdot s^{-1}$. The urban sites CAMS1, CAMS2, UCAS, and WD show
634 the highest values (1.5×10^{-4} – 2.0×10^{-4} $ppb \cdot s^{-1}$), whereas SJZ_LC, likely influenced by
635 local emissions and complex topography, exhibits slightly lower peaks (1.0×10^{-4} –
636 1.5×10^{-4} $ppb \cdot s^{-1}$). SXU reaches $\sim 1.7 \times 10^{-4}$ $ppb \cdot s^{-1}$. At all these sites, $P(OH)O_3$
637 decreases with altitude and generally falls below 1.0×10^{-4} $ppb \cdot s^{-1}$ above 1 km. East
638 China stations (TS, NUIST, LA, HNU, AHU, and CF) display similar peak timing
639 (12:00–14:00), with near-surface maxima of 1.0×10^{-4} – 1.8×10^{-4} $ppb \cdot s^{-1}$. TS, NUIST,
640 HNU, AHU, and CF reach $\sim 1.5 \times 10^{-4}$ – 1.8×10^{-4} $ppb \cdot s^{-1}$, while LA peaks at $\sim 1.4 \times 10^{-4}$
641 $ppb \cdot s^{-1}$. In South China (GIG, SUST, GZ_ZL, GZ_NS, GZ_TM, GZ_GY, and
642 GZ_DXC), the maxima occur slightly later (13:00–15:00) and are generally higher
643 (1.3×10^{-4} – 1.7×10^{-4} $ppb \cdot s^{-1}$), with GZ_GY reaching $\sim 1.9 \times 10^{-4}$ $ppb \cdot s^{-1}$ and GZ_NS,
644 GZ_TM, and GZ_DXC $\sim 1.8 \times 10^{-4}$ $ppb \cdot s^{-1}$. Southwest China (CDAES and CQ) shows
645 peaks of $\sim 1.5 \times 10^{-4}$ – 2.5×10^{-4} $ppb \cdot s^{-1}$, including $\sim 1.9 \times 10^{-4}$ $ppb \cdot s^{-1}$ at CQ and
646 $\sim 1.5 \times 10^{-4}$ $ppb \cdot s^{-1}$ at CDAES. The Central China site LY exhibits a peak of $\sim 1.5 \times 10^{-4}$
647 $ppb \cdot s^{-1}$, comparable to those in North and East China, indicating broadly similar
648 ozone photochemical efficiencies across these regions.



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Figure 10. Mean vertical profiles of OH radicals produced by O₃ photolysis.

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652 3.4 Validations with independent data

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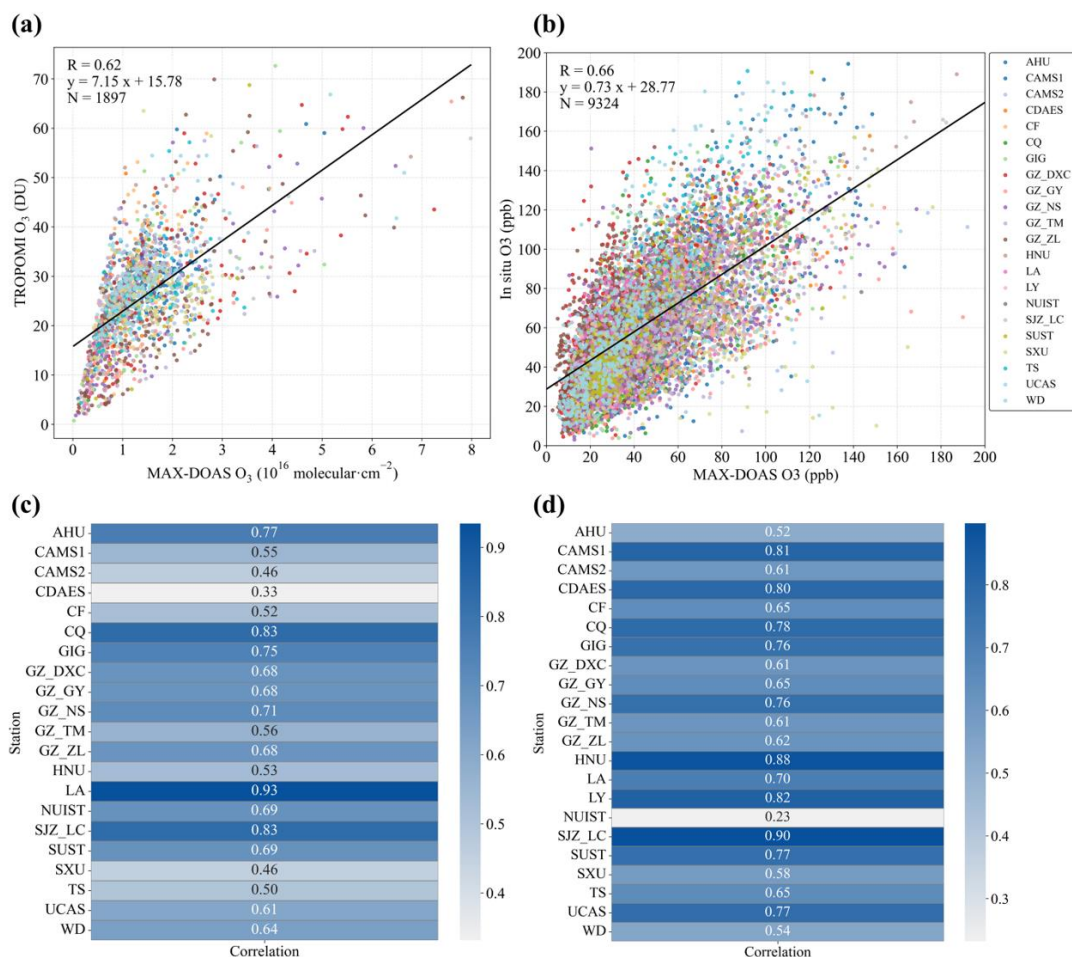
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The dataset was validated using two independent approaches. First, O₃ VCD retrieved from the MAX-DOAS network for 2021–2024 were evaluated against coincident TROPOMI satellite observations. MAX-DOAS measurements were averaged within ± 30 min of the TROPOMI overpass (13:30–14:00 Beijing Time), and TROPOMI pixels were spatially averaged over a $7 \text{ km} \times 5.5 \text{ km}$ area center on each site, consistent with the native spatial resolution of TROPOMI. As shown in Figure 11a, the two datasets exhibit a strong linear relationship, with a Pearson correlation coefficient of $R = 0.62$ ($N = 1897$); site-resolved correlations are given in Figure 11c. Second, near-surface O₃ concentrations retrieved at the 22 hyperspectral sites were compared with in situ measurements from the nearest CNEMC over the same period. Site pairs were selected following the spatial representativeness criteria of Song et al. (2023b). 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

667 both sites sampled comparable urban or suburban atmospheric conditions. The
 668 comparison (Figure 11b) shows a significant positive correlation ($R = 0.66$, $N = 9324$),
 669 demonstrating good consistency between MAX-DOAS–derived surface O_3 and
 670 ground-based observations. The correlations for each hyperspectral site and its nearest
 671 CNEMC are summarized in Figure 11d. Together, these two independent validations
 672 demonstrate reasonable consistency and provide confidence in the dataset used in this
 673 study.



674 Figure 11. (a) Correlation between O_3 column densities retrieved from hyperspectral
 675 ground-based stations and TROPOMI satellite observations; (b) correlation between
 676 hyperspectral O_3 column densities and in situ O_3 measurements from the nearest
 677 CNEMC; (c) site-specific correlations between hyperspectral and TROPOMI O_3
 678 column densities; (d) site-specific correlations between hyperspectral O_3 column
 679 densities and in situ O_3 at the nearest CNEMC.

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682 4. Data availability

683 The vertical profiles of HONO and O_3 , and the vertical profiles of OH radicals
 684 over the major regions of China presented in this study, are freely available in .xlsx
 685 format at Zenodo (<https://doi.org/10.5281/zenodo.18489836>; Zou et al., 2026).

686

687 5. Summary

688 We developed and released a comprehensive dataset of vertical profiles of
689 HONO and O₃, and the associated OH radical production rates, P(OH)_{HONO} and
690 P(OH)_{O₃}, derived from the Chinese hyperspectral vertical remote-sensing network for
691 2021–2024. The dataset spans 22 representative sites across North, East, Central,
692 South, and Southwest China, covering a wide range of climatic regimes and surface
693 types, and represents one of the most extensive publicly available collections in China
694 in terms of spatial coverage and vertical resolution of photochemical parameters
695 relevant to OH precursors. Independent validation against TROPOMI satellite
696 retrievals and in situ measurements from the CNEMC demonstrates robust
697 consistency. Mean diurnal profiles within 0–4 km reveal pronounced regional and
698 vertical contrasts in HONO and O₃ driven photochemistry. Both J(HONO) and
699 J(O(¹D)) exhibit radiation controlled, single-peaked diurnal cycles, with maxima
700 around local noon (11:00–14:00), and remain elevated in the upper mixed layer and
701 the lower free troposphere, reflecting the combined effects of radiative transfer and
702 aerosol extinction on the vertical distribution of photolysis rates. Accordingly,
703 P(OH)_{HONO} and P(OH)_{O₃} peak near the surface and decrease with height, indicating
704 that the boundary layer is the primary daytime source region of OH. At several
705 plateau and mountainous sites, however, the lower free troposphere also shows a
706 substantial radical production potential. Urban and highly industrialized sites exhibit
707 higher photolysis rates and OH production, reflecting the combined effects of high
708 precursor concentrations and strong radiation, while high-altitude clean-background
709 sites, despite lower near-surface concentrations, maintain relatively large photolysis
710 rates and significant OH production at middle and upper levels due to weaker aerosol
711 extinction and stronger shortwave radiation, showing a vertical photochemical
712 structure distinct from that over plains.

713 With continuous temporal coverage ranging from five months to 3.5 years and
714 multi-site vertical profiling, this dataset provides a valuable foundation for: (1)
715 quantifying the relative contributions of HONO and O₃ photolysis to the OH budget
716 in the boundary layer and the lower free troposphere; (2) constraining radical initial
717 conditions and radiative parameterizations in regional and global chemical transport
718 models; (3) enabling cross-validation and synergistic inversion among ground-based,
719 UAV, and satellite observations; (4) advancing studies of photochemical pollution
720 formation, secondary aerosol production, and atmospheric oxidation capacity; and (5)
721 supporting air-quality management and policy development as a complementary
722 national monitoring resource. (6) serving as a critical benchmark for assessing and
723 reducing uncertainties in vertical parameterization schemes (e.g., turbulent mixing,
724 photolysis rates, heterogeneous reactions) within atmospheric chemical transport
725 models. However, it is important to note the limitations of the current dataset. The
726 time span (2021–2024) limits the capacity for robust analysis of long-term interannual
727 trends driven by climate change or policy shifts. The observed photochemical regimes
728 may not fully represent conditions during extreme climatic years outside this period,

729 and the climatological representativeness of sites with shorter operational histories
730 requires continued data accumulation.

731

732 **Declaration of competing interest**

733 The authors declare that they have no known competing financial interests or personal
734 relationships that could have appeared to influence the work reported in this paper.

735

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740

741 **Author contributions**

742 All authors contributed to the generation of the dataset described in this paper. Tiliang
743 Zou, Chengzhi Xing, and Cheng Liu wrote the manuscript, while all other authors
744 participated in its revision.

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