

Observations of the vertical distributions of summertime atmospheric pollutants in Nam Co: OH production and source analysis

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1 Abstract

2 The Tibetan Plateau (TP) plays a key role in regional environment and global climate change, however,
3 the lack of vertical observation of atmospheric species, such as HONO and O₃, hinders a deeper
4 understanding of the atmospheric chemistry and atmospheric oxidation capacity (AOC) on the TP. In
5 this study, we conducted multi-axis differential optical absorption spectroscopy (MAX-DOAS)
6 measurements at Nam Co, the central TP, to observe the vertical profiles of aerosol, water vapor (H₂O),
7 NO₂, HONO and O₃ from May to July 2019. In addition to NO₂ mainly exhibiting a Gaussian shape
8 with the maximum value appearing at 300-400 m, other four species all showed an exponential shape
9 and decreased with the increase of height. The maximum values of monthly averaged aerosol (0.17
10 km⁻¹) and O₃ (66.71 ppb) occurred on May, H₂O (3.68×10^{17} molec cm⁻³) and HONO (0.13 ppb)
11 appeared on July, while NO₂ (0.39 ppb) occurred on June at 200-400 m layer. H₂O, HONO and O₃ all
12 exhibited a multi-peak pattern, and aerosol appeared a bi-peak pattern for their averaged diurnal
13 variations. The averaged vertical profiles of OH production rates from O₃ and HONO all exhibited an
14 exponential shape decreasing with the increase of height with maximum values of 2.61 ppb/h and 0.49
15 ppb/h at the bottom layer, respectively. The total OH production rate contributed by HONO and O₃ on
16 the TP was obviously larger than that in low-altitude areas. In addition, source analysis for HONO and
17 O₃ at different height layers were conducted. The heterogeneous reaction of NO₂ on wet surfaces was a
18 significant source of HONO. The maximum values of HONO/NO₂ appeared around H₂O being $1.0 \times$
19 10^{17} molec cm⁻³ and aerosol being lager 0.15 km⁻¹ under 1.0 km, and the maximum values usually
20 accompanied with H₂O being $1.0\text{-}2.0 \times 10^{17}$ molec cm⁻³ and aerosol being lager 0.02 km⁻¹ at 1.0-2.0 km.
21 O₃ was potentially sourced from south Asian subcontinent and Himalayas through long-range transport.
22 Our results enrich the new understanding of vertical distribution of atmospheric components and
23 explained the strong AOC on the TP.

24

25 1 Introduction

26 The TP spans 2.5 million square kilometers with an average altitude of over 4000 m. Therefore, the TP
27 is called the “Third Pole” of the earth (Ma et al., 2020; Kang et al., 2022). It is the home to tens of
28 thousands of glaciers and nourishes more than 10 of Asia’s rivers, thus it also acts the role of “Water
29 Tower of Asia” (Qu et al., 2019; Ma et al., 2022). Due to its special topography, the TP is the heat
30 source of atmosphere due the strong solar radiation, which as the driven force to profoundly affect the
31 regional atmospheric circulation, global weather conditions and climate change (Yanai et al., 1992;
32 Boos et al., 2010; Chen et al., 2015; Liu et al., 2022; Zhou et al., 2022). Monsoon rainfall in Asia, flood
33 over the Yangtze River valley, and El Niño in the Pacific Ocean are strongly associated with the TP
34 (Hsu et al., 2003; Li et al., 2016; Lei et al., 2019). In addition, the cyclone circulations caused by the
35 TP heat source also can inhibit the diffusion of atmospheric pollutants in the areas around the TP, such
36 as the Sichuan Basin, causing regional pollution (Zhang et al., 2019). Therefore, observations of the
37 atmospheric species on the TP are essential to enhance the in-depth understanding of its atmospheric
38 physicochemical processes.

39 However, deciphering the atmospheric environment of the TP is highly challenging and dangerous, due
40 to its complex topography and harsh environment (Barnett et al., 2005; Bolch et al., 2012; Cong et al.,
41 2015; Kang et al., 2016). In order to unveil the feature of atmospheric composition over the TP and
42 their corresponding climate feedback, a large number of field observation stations have been
43 established, and a series of field campaigns have continued to be carried out recently, especially after
44 the performance of “the Second Tibetan Plateau Scientific Expedition and Research Program” (Che and
45 Zhao 2021; Wang et al., 2021; Ran et al., 2022). The China National Environmental Monitoring Center
46 (CNEMC) has established an in-situ monitoring network with more than 12 stations over the TP, such
47 as Lhasa, Shigatse, Shannan, Nyingchi, Nagqu, Ngari, Qamdo, Diqing, Aba, Guoluo, Xining, and
48 Haixi, to continuously monitor the surface concentrations of six atmospheric components (i.e. PM₁₀,
49 PM_{2.5}, NO₂, SO₂, O₃ and CO) since 2013 (Gao et al., 2020; Li et al., 2020; Sun et al., 2021). The
50 Institute of Tibetan Plateau Research, Chinese Academy of Sciences, has also established six long-term
51 field observation stations to measure meteorological parameters and small amounts of atmospheric
52 composition (i.e. black carbon, aerosol optical density (AOD)) (Ma et al., 2020). In addition, scientists
53 are relying on advancements in satellite remote sensing technology, such as the tropospheric
54 monitoring instrument (TROPOMI), the ozone monitoring instrument (OMI), the moderate-resolution
55 imaging spectroradiometer (MODIS) and the cloud-aerosol lidar and infrared pathfinder satellite
56 observation (CALIPSO), to monitor the spatial and temporal evolutions of atmospheric composition on
57 the TP (Zhu et al., 2019; Li et al., 2020; Rawat and Naja 2022). Their advantage is to obtain the column
58 densities of pollutants in a large-scale space of the TP. Although CALIPSO could detect aerosol
59 vertical profiles, the spatiotemporal resolution (i.e. ~5.0 km horizontal resolution, 0.06 km vertical
60 resolution and ~16 d temporal resolution) is limited and the data uncertainty in the planetary boundary
61 layer (PBL) is large due to the low signal-to-noise ratio (Huang et al., 2007). However, several studies
62 also revealed that the formation, aging and transport processes of atmospheric composition on the TP
63 occurs not only near the ground surface but also at high altitudes (Xu et al., 2020; Xu et al., 2022). The
64 high PBL on the TP caused by its strong solar radiation and undulating terrain promotes the
65 atmospheric exchange between the bottom troposphere and stratosphere (Yang et al., 2003; Seidel et al.,
66 2010). Therefore, the lack of vertical profiles of hinders the understanding of the evolution of trace
67 gases and their environmental and climate effects over the TP. In recent years, balloon and lidar
68 vertical measurements on the TP are occasionally carried out (Fang et al., 2019; Zhang et al., 2020;
69 Dong et al., 2022), but their limited detection species (i.e. aerosol and O₃) and high cost are obstacles
70 that limit long-term continuous observation and the conduction of more in-depth scientific research.
71 MAX-DOAS has the technical advantage of low-cost continuous observation of multiple atmospheric
72 components (i.e. aerosol, O₃ and their precursors) (Wang et al., 2018; Ma et al., 2020; Cheng et al.,
73 2021; Xing et al., 2021; Li et al., 2022; Cheng et al., 2023a, 2023b). Combining these data with better
74 scientific models can reduce the modeling bias and promote to better understand the physical, chemical
75 and dynamical processes.

76 The strong convergent airflow formed under the combined action of monsoon, subtropical anticyclone
77 and the airflow of subtropical westerlies could promote the accumulation of O₃ on the TP in summer

78 (Ye and Gao 1997). Therefore, several studies have revealed the high O₃ concentration on the TP (Li et
79 al., 2022; Yang et al., 2022; Yu et al., 2022). The strong solar radiation, high O₃ concentration and
80 relatively high humidity on the TP provide great potential for high OH production. Lin et al. (2008) and
81 Ye (2019) also confirmed that the high OH over the TP is mainly related to the reaction between O(¹D)
82 and H₂O. The O(¹D) is produced from the photolysis of O₃ by UV radiation. Therefore, a hypothesis of
83 “strong AOC over the TP” was put forward. Previous studies pointed out that HONO also play an
84 important role in AOC at low-altitude areas, and its contribution to OH can reach 40-60%, and even
85 more than 80% in the early morning (Michoud et al., 2012; Ryan et al., 2018; Xue et al., 2020).
86 However, few HONO studies on the TP have been reported. Our previous study operated at the
87 Qomolangma Atmospheric and Environmental Observation and Research Station, Chinese Academy of
88 Sciences (QOMS-CAS) revealed that the HONO mainly distributed in the lower PBL and peaked in
89 summer with 1.11 ppb, which is comparable to the average level of HONO in other low-altitude areas
90 (Luo et al., 2010; Xing et al., 2021a, 2021b; Yang et al., 2021). It indicates that it is also necessary to
91 study the contribution of HONO to AOC on the TP. Furthermore, understanding the vertical
92 distribution of OH is of great significance for learning about the atmospheric chemical processes and
93 the evolution of atmospheric components on the TP (Zhou et al., 2015). Identifying the sources of O₃
94 and HONO is the basis for studying the AOC on the TP. The limited researches concluded that the
95 atmospheric HONO on the TP is mainly sourced from the emissions of vehicles, biomass burning and
96 soil, except for the NO₂ heterogeneous reaction on aerosol surfaces (Xing et al., 2021). The lower
97 tropospheric O₃ on the TP is mainly dominated by local photochemical reactions, regional horizontal
98 transport, vertical mixing and the intrusion from stratosphere (Yin et al., 2017; Xu et al. 2018).

99 In this study, we firstly analyzed the temporal and vertical characteristics of several atmospheric
100 components (i.e. aerosol, H₂O, NO₂, HONO and O₃) based on MAX-DOAS observations in Nam Co.
101 Afterwards, the contributions of O₃ and HONO to OH in the vertical space were discussed through the
102 tropospheric ultraviolet and visible (TUV) radiative transfer model and MAX-DOAS measurements.
103 Finally, the potential sources of O₃ and HONO at different altitudes were analyzed based on the
104 MAX-DOAS retrievals.

105 **2 Method and methodology**

106 **2.1 Site**

107 The Nam Co Monitoring and Research Station for Multisphere Interactions, CAS (NAMORS)
108 (30.774°N, 90.988°E; 4730 m a.s.l.) is located at the southeast banks of Nam Co lake and the foothills
109 of the northern Mt. Nyainqêntanglha (Fig. 1). The station land is covered by alpine meadows with soil
110 type of sandy silt loam. The southwest monsoon can carry abundant moisture from Indian Ocean to this
111 station in summer to increase humidity and precipitation there. Moreover, due to the summertime huge
112 evaporation from Nam Co lake, the atmospheric H₂O around CAS (NAMORS) is more abundant than
113 in other areas of the TP, resulting in lush grass vegetation and making the area around this station an
114 important summertime pasture. In addition, there are not large industries and cities within 100 km of
115 the CAS (NAMORS). The closest town to CAS (NAMORS) is Dangxiong county which is about 60
116 km away from this station and lower about 500 m than this station. Only a small number of vehicles
117 pass through this area during summer tourism season. Therefore, no obvious anthropogenic sources of
118 air pollutants exist near this station. Averaged spatial distributions of AOD, O₃, NO₂ and HCHO
119 monitored by satellite from May to July 2019 are shown in Figure S1. Elevated AOD, NO₂, and O₃ are
120 mainly distributed in South Asian subcontinent (e.g. India and Nepal), the southern foothills of the
121 Himalayas, which is located in the upwind direction of the southwest monsoon potentially affecting the
122 atmospheric composition over CAS (NAMORS).

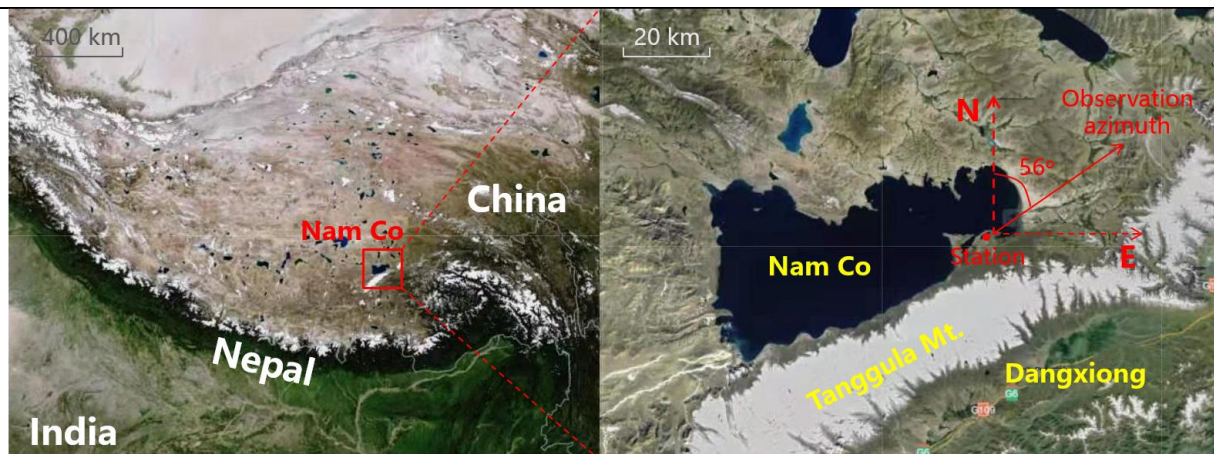


Figure 1. Geographical location of CAS (NAMORS) on the Tibet plateau.

2.2 Measurements

2.2.1 Instrument setup and spectral analysis

The MAX-DOAS instrument installed at CAS (NAMORS) was operated from 01 May to 09 July 2019. It consists of three major parts: telescope unit, spectrometer unit and control unit. The detailed description of this instrument can be found in Xing et al. (2021). In this study, the elevation angle sequence was set to 1, 2, 3, 4, 5, 6, 8, 10, 15, 30, and 90° with an exposure time of 60 s to each individual spectrum. The azimuth angle was set to 56° pointing to Nagqu direction. Moreover, only spectra collected under solar zenith angle (SZA) less than 75° was used for spectral analysis to avoid the strong stratospheric absorption.

The differential slant column densities (DSCDs) of O₄, H₂O, NO₂, HONO and O₃ were retrieved using QDOAS software (<http://uvvis.aeronomie.be/software/QDOAS/>) developed by Royal Belgian Institute for Space Aeronomy (BIRA-IASB). The zenith spectrum measured at every sequence were selected as scan Fraunhofer reference spectrum. The retrieval configurations of O₄, H₂O, NO₂, HONO and O₃ followed Xing et al. (2017), Lin et al. (2020), Xing et al. (2021), Wang et al. (2020) and Wang et al. (2018), respectively. The detailed DOAS fit settings of above five species were listed in Table 1. Corrected I₀ (Aliwell et al., 2002) was used in this study. Fig. 2 shows a typical DOAS retrieval example for above five species. DOAS fit results with root mean square (RMS) values larger than 5 × 10⁻⁴, 5 × 10⁻⁴, 5 × 10⁻⁴, 1 × 10⁻³, and 6 × 10⁻⁴ for O₄, H₂O, NO₂, HONO, and O₃, respectively, were filtered out. In addition, we calculated color index (CI) to remove cloud effect (Wagner et al., 2016). The data filter criteria according to CI followed by Ryan et al. (2018) and Xing et al. (2020). Afterwards, the quantified DSCDs of O₄, H₂O, NO₂, HONO, and O₃ remained 91.33%, 91.97%, 92.16%, 86.42% and 81.09%, respectively.

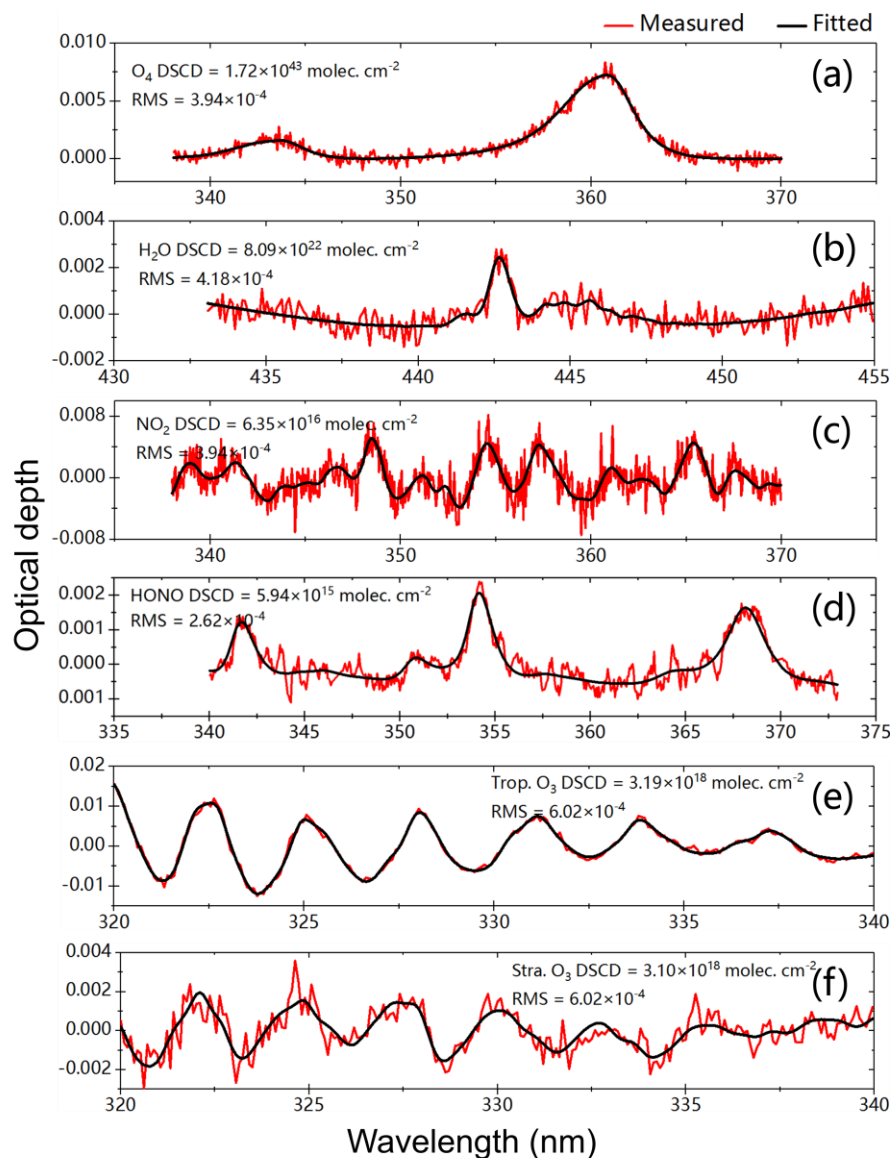
2.2.2 Vertical profile retrieval

The vertical profiles of aerosol and trace gases (i.e. H₂O, NO₂, HONO and O₃) were retrieved using algorithm based on optimal estimation method (OEM). A linearized pseudo-spherical vector discrete ordinate radiative transfer model VLIDORT was used as forward model and a Gauss-Newton (GN) scheme was used as the inversion strategy (Wedderburn et al., 1974). The detailed description of this algorithm can be found in Liu et al. (2021), Xing et al. (2021) and Wang et al. (2018). The detailed retrieval processes were depicted in Sect. S1 of the supplement. In this study, the initial a priori profile shape of above five species was set to exponential decreasing shape, and the AOD and vertical column densities (VCDs) simulated by weather research and forecasting model coupled chemistry (WRF-Chem) were also used as initial input a priori information to constrain the retrieval process. For the O₃ profile retrieval, the stratospheric O₃ profile was deducted using TROPOMI O₃ profile (Zhao et al., 2021). We set 20 vertical layers from 0.0 to 4.0 km with a vertical resolution of 0.2 km. The correlation height was set to 1.0 km. Moreover, the surface albedo, single scattering albedo and asymmetry parameter were set to fixed constant of 0.08, 0.85 and 0.65, respectively (Irie et al., 2008). The retrieved vertical profiles

161 were removed under the condition of degree of freedom (DOF) and relative error less than 1.0 and
 162 100%, respectively.

163 Table 1. Detailed DOAS retrieval settings for O₄, H₂O, NO₂, HONO and O₃.

Parameter	Data source	Fitting intervals (nm)				
		O ₄	H ₂ O	NO ₂	HONO	O ₃
Wavelength range		338-370	433-455	338-370	340-373	320-340
NO ₂	298K, I ₀ -corrected, Vandaele et al. (1998)	✓	✓	✓	✓	✓
NO ₂	220K, I ₀ -corrected, Vandaele et al. (1998)	✓	✓	✓	✓	×
O ₃	223K, I ₀ -corrected, Serdyuchenko et al. (2014)	✓	✓	✓	✓	✓
O ₃	243K, I ₀ -corrected, Serdyuchenko et al. (2014)	✓	×	✓	✓	×
O ₃	293K, I ₀ -corrected, Serdyuchenko et al. (2014)	×	×	×	×	✓
O ₄	293K, Thalman and Volkamer (2013)	✓	✓	✓	✓	✓
HCHO	298K, Meller and Moortgat (2000)	✓	×	✓	✓	✓
Glyoxal	298K, Volkamer (2005)	×	✓	×	×	×
H ₂ O	HITEMP (Rothman et al. 2010)	✓	✓	✓	✓	×
BrO	223K, Fleischmann et al. (2004)	✓	×	✓	✓	×
HONO	296K, Stutz et al. (2000)	×	×	×	✓	×
Ring	Calculated with QDOAS	✓	✓	✓	✓	✓
Polynomial degree		Order 3	Order 3	Order 3	Order 5	Order 3
Intensity offset		Constant	Constant	Constant	Constant	No



164

165 Figure 2. DOAS fit examples of O₄, H₂O, NO₂, HCHO, tropospheric O₃ and stratospheric O₃. The red
 166 line and black line represent the measured and fitted results, respectively.

167 2.2.3 Error analysis

168 The error sources can be divided into four different types: smoothing error, noise error, forward model
 169 error, and model parameter error (Rodgers, 2004). However, in terms of this classification, some errors
 170 are difficult to be calculated or estimated. For example, the forward model error, which is caused by an
 171 imperfect representation of the physics of the system, is hard to be quantified due to the difficulty of
 172 acquiring an improved forward model. Given calculation convenience and contributing ratios of
 173 different errors in total error budget, we mainly took into account following error sources, which were
 174 smoothing and noise errors, algorithm error, cross section error, and uncertainty related to the aerosol
 175 retrieval (only for trace gas). In this study, we estimated the contribution of different error sources to
 176 the AOD and VCDs of trace gases, and near-surface (0–200 m) trace gases' concentrations and aerosol
 177 extinction coefficients (AECs), respectively. The detailed demonstrations and estimation methods are
 178 displayed below.

- 179 a. Smoothing errors arise from the limited vertical resolution of profile retrieval. Noise errors denote
 180 the noise in the spectra (i.e., the error of DOAS fits). Considering the error of the retrieved state
 181 vector equaling the sum of these two independent errors, we calculated the sum of smoothing and
 182 noise errors on near-surface concentrations and column densities, which were 13 and 5 % for
 183 aerosols, 13 and 36 % for H₂O, 12 and 14 % for NO₂, 18 and 21 % for HONO, and 12 and 32 % for
 184 O₃, respectively.
- 185 b. Algorithm error is denoted by the differences between the measured and simulated DSCDs. This
 186 error contains forward model error from an imperfect approximation of forward function, parameter
 187 error of forward model, and other errors, such as detector noise (Rodgers, 2004). Algorithm error is
 188 a function of the viewing angle, and it is difficult to assign this error to each altitude. Thus, this
 189 error on the near-surface values and column densities is estimated through calculating the average
 190 relative differences between the measured and simulated DSCDs at the minimum and maximum
 191 elevation angle (except 90°), respectively (Wagner et al., 2004). In this study, we estimated these
 192 errors on the near-surface values and the column densities at 4 and 8 % for aerosols, 3 and 11 % for
 193 NO₂, and 20 and 20 % for HONO referring to Wang et al. (2017, 2020), 1 and 8 % for H₂O
 194 referring to Lin et al. (2020), and 6 and 10 % for O₃ referring to Ji et al. (2023), respectively.
- 195 c. Cross section error arises from the uncertainty in the cross section. According to Thalman and
 196 Volkamer, (2013), Lin et al. (2020), Vandaele et al. (1998), Stutz et al. (2000), and Serdyuchenko
 197 et al. (2014), we adopted 4, 3, 3, 5, and 2 % for O₄ (aerosols), H₂O, NO₂, HONO and O₃,
 198 respectively.
- 199 d. The profile retrieval error for trace gases is sourced from the uncertainty of aerosol extinction
 200 profile retrieval and propagated to trace gas profile. This error could be roughly estimated based on
 201 a linear propagation of the total error budgets of the aerosol retrievals. The errors of the learned
 202 four trace gases were roughly estimated at 14 % for VCDs and 10 % for near-surface
 203 concentrations, respectively.

204 The total uncertainty was the sum of all above errors in the Gaussian error propagation, and the error
 205 results were listed in Table 2. We found that the smoothing and noise errors played a dominant role in
 206 the total uncertainties of aerosol and trace gases. Moreover, improving the accuracy and temperature
 207 gradient of the absorption cross section is another important means to reduce the uncertainty of the
 208 vertical profiles in the future, especially for O₃.

209 Table 2. Error budget estimation (in %) of the retrieved near-surface (0–200 m) concentrations of trace
 210 gases and AECs, and AOD and VCDs.

		Error sources				Total
		Smoothing and noise errors	Algorithm error	Cross section error	Related to the aerosol retrieval	
Near-surface	aerosol	13	4	4	/	14
	H ₂ O	13	1	3	14	19
	NO ₂	12	3	3	14	18
	HONO	18	20	5	14	29

	O ₃	12	6	2	14	19
VCD or AOD	AOD	5	8	4	/	10
	H ₂ O	36	8	3	10	38
	NO ₂	14	11	3	10	20
	HONO	21	20	5	10	31
	O ₃	32	10	2	10	35

211

212 **2.3 TUV model**

213 The calculation of photolysis rates of HONO and O₃ used TUV radiation model
 214 (<https://www2.acom.ucar.edu/modeling/tropospheric-ultraviolet-and-visible-tuv-radiation-model>)
 215 based on a full FORTRAN code. In order to ensure the accuracy of model running, we only selected
 216 data in sunny and cloudless days. Moreover, we developed a cloud classification method based on the
 217 diurnal variations of Color Index ($CI=I_{330}/I_{360}$) in Figure S2. The initial input parameters were as
 218 follows: the AOD at 361 nm was derived from aerosol extinction profiles measured by MAX-DOAS;
 219 the daily total ozone column density was measured by TROPOMI with a value range of 260-280 DU;
 220 the single scattering albedo (SSA) was calculated based on the regression analysis of multi-wavelength
 221 (361 and 477 nm) O₄ absorptions measured by MAX-DOAS (Xing et al., 2019); fixed Ångström
 222 exponents of 0.508, 0.581 and 0.713 were used in May, June and July, respectively, referring to Xia et
 223 al. (2011).

224 **2.4 Backward trajectory, PSCF and CWT analysis**

225 The 48-h backward trajectories at five heights of 200, 600, 1000, 1400 and 1800 m were calculated
 226 using the Hybrid Single-particle Lagrangian Integrated Trajectory (HYSPLIT) model based on the
 227 Global Data Assimilation System (GDAS) to identify the major transport pathways of O₃ (Draxler and
 228 Hess, 1998). Moreover, the calculated backward trajectories were clustered into three groups using
 229 Ward's variance method and Angle Distance algorithm (Ward 1963; Wang et al., 2006).

230 In order to determine the potential source locations of O₃ over CAS (NAMORS), the Potential Source
 231 Contribution Function (PSCF) model and Concentration Weighted Trajectory (CWT) model were used
 232 (Hong et al., 2019; Ou et al., 2021). The PSCF was calculated through the number of air trajectory
 233 endpoints being divided by the number of air trajectory endpoints. Moreover, a weighting function was
 234 introduced to reduce the increased uncertainties of PSCF with the increase of the distance between the
 235 grid and sampling point. In this study, the set of this weighting function referred to Yin et al. (2017).
 236 CWT can be used to calculate the weight concentration through averaging the concentrations
 237 associated with trajectories crossing the grid cell. Above weighting function was also introduced to
 238 calculate the WCWT (Hsu, et al., 2003). The detailed description of these two models can be found in
 239 Wang et al., 2006.

240 **2.5 Ancillary data**

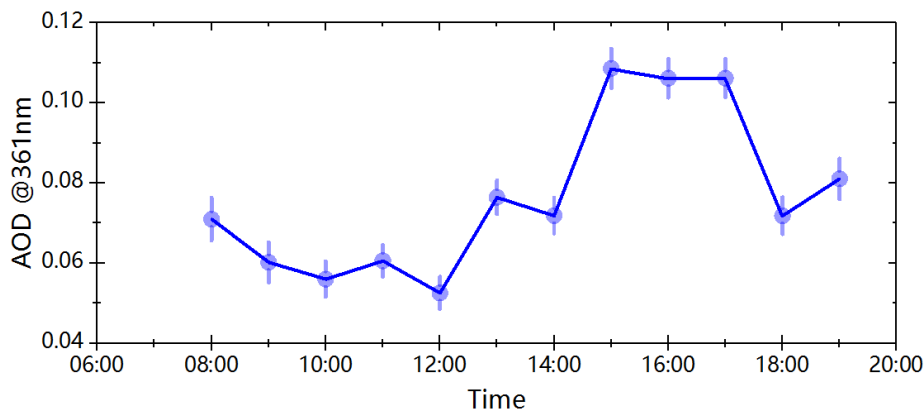
241 The surface NO₂, HONO and O₃ concentrations used to validate the corresponding MAX-DOAS
 242 measurements were monitored by broadband cavity enhanced spectrometer (BBCES) (Fang et al.,
 243 2017), long path absorption photometer (LOPAP) (Kleffmann et al., 2008) and Thermo Electron 49i
 244 (Shi et al., 2009), respectively. The PBL height was simulated using WRF with spatiotemporal
 245 resolutions of 20×20 km² and 1.0 hour (detailed configurations in Sect. S3 of the supplement).
 246 Moreover, the large-scaled spatial distributions of AOD, O₃ and NO₂ over CAS (NAMORS) were
 247 monitored by Himawari-8 (Bessho et al., 2016), OMI (Veefkind et al., 2004) and TROPOMI (Griffin et
 248 al., 2018; Su et al., 2020), respectively.

249 **3 Results**

250 **3.1 Overview of the measurements**

251 Figure 3 showed the averaged diurnal variation of AOD from 1st May to 9th July 2019, with an average
 252 value of 0.076 km⁻¹ during 08:00-19:00. The AOD was 0.071 km⁻¹ at 08:00, and then gradually
 253 decreased to a minimum value of 0.052 km⁻¹ at 12:00. Subsequently, the AOD increased significantly,

254 reaching maximum values during 15:00-17:00 (average of 0.107km^{-1}), which was about 1.408 times
 255 the diurnal average value. Considering the diurnal variation of wind speed (Figure S3), such an
 256 enhancement of AOD may be related to the long-range transport of aerosol from southern Asia (Yang
 257 et al., 2020; Bi et al., 2023). Moreover, 15:00-17:00 was the active time of tourists and local residents
 258 (i.e. cooking), and these kinds of anthropogenic sources contributed to the atmospheric AOD of
 259 NAMORS through short-distance transport (Yin et al., 2017; Zhang et al., 2017). After 17:00, the
 260 AODs decreased rapidly to 0.071 km^{-1} at 18:00 and 0.081 km^{-1} at 19:00, respectively.



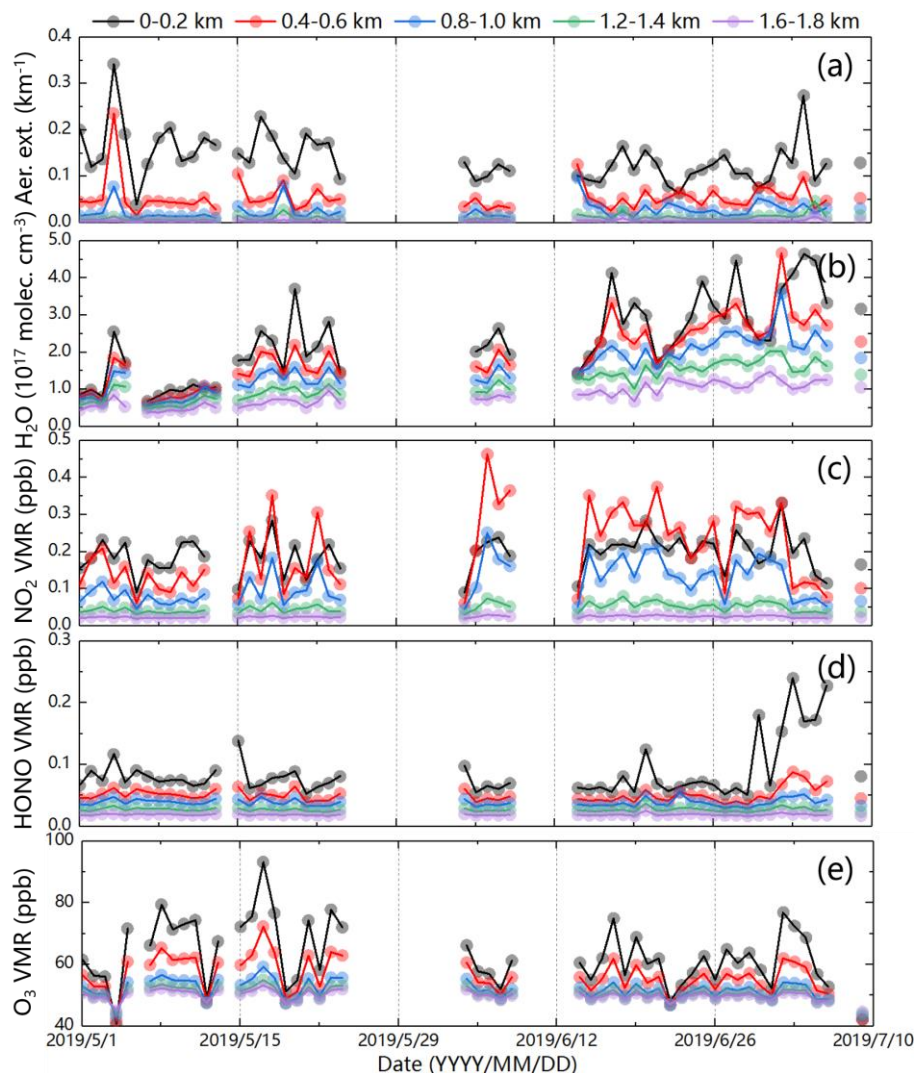
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262 Figure 3. Averaged diurnal variation of AOD at CAS (NAMORS). The error bars represent the mean retrieved errors
 263 of AOD.

264 As shown in Figure S4, the diurnal variation of PBL in Nam Co from May to July 2019 was lower in
 265 the early morning and late afternoon, but higher between 11:00 and 17:00, a relatively long period,
 266 with the maximum PBL larger than 2.0 km. Zhang et al. (2017) and Yang et al., (2017) also reported
 267 that the PBL in Nam Co was usually larger than 1.0 km during daytime in spring and summer. In order
 268 to investigate the height-dependent variations of aerosol, H_2O , NO_2 , HONO and O_3 within the PBL
 269 during the measurements, five height layers under the PBL (0.0-0.2 km, 0.4-0.6 km, 0.8-1.0 km,
 270 1.2-1.4 km and 1.6-1.8 km) were thus selected.

271 Figure 4 showed the time series of the daily averaged aerosol, H_2O , NO_2 , HONO and O_3 at above five
 272 layers from 1st May to 9th July 2019. Aerosol mainly distributed at 0.0-0.2 km with an average
 273 extinction coefficient of 0.138 km^{-1} , and the ratios of aerosol extinction at 0.4-0.6 km, 0.8-1.0 km,
 274 1.2-1.4 km and 1.6-1.8 km to those at 0.0-0.2 km were 39.34%, 18.77%, 7.29% and 2.62%,
 275 respectively. That indicated that the aerosol was usually local-emitted at the surface, and the
 276 occasionally appearance of strong aerosol extinction at 0.4-0.6 km, such as 13th and 30th June, was
 277 associated with long-range transport from south Asia (Figure S5, Wan et al., 2015; Li et al., 2016). The
 278 average concentration of H_2O at 0.0-0.2 km was $2.35 \times 10^{17}\text{ molec cm}^{-3}$, and the ratios of H_2O at
 279 0.4-0.6 km, 0.8-1.0 km, 1.2-1.4 km and 1.6-1.8 km to those at 0.0-0.2 km were 83.40%, 68.08%,
 280 50.64% and 35.74%, respectively, which should attribute to the transport of H_2O from southern Asia
 281 driven by the Indian ocean monsoon and the elevated evaporation from Nam Co lake to lead to its not
 282 obvious vertical gradient (Figure S6, Lei et al., 2014; Zhu et al., 2019). The average concentration of
 283 NO_2 at 0.0-0.2 km was 0.193 ppb, and its high concentration mainly distributed at 0.4-0.6 km after 15th
 284 May. The ratios of NO_2 at 0.4-0.6 km, 0.8-1.0 km, 1.2-1.4 km and 1.6-1.8 km to those at the bottom
 285 layer were 104.03%, 59.05%, 24.62% and 12.84%, respectively. The elevation of the distribution
 286 height of high concentration NO_2 should be attributed to the transport process from the NO_x produced
 287 by ice and snow on the top of Mt. Tanggula under strong ultraviolet radiation (Boxe et al., 2005; Fisher
 288 2005; Lin et al., 2021). As depicted in Figure S7, the WPCF passing through Mt. Tanggula showed
 289 high values at 300-400 m layer, especially at 400 m (> 0.3). It also indirectly indicated that the
 290 important contribution to NO_x from ice and snow on the top of mountains under strong ultraviolet
 291 radiation on the TP. HONO mainly distributed at 0.0-0.2 km with an average value of 0.087 ppb, and
 292 the ratios of HONO at 0.4-0.6 km, 0.8-1.0 km, 1.2-1.4 km and 1.6-1.8 km to those at 0.0-0.2 km were
 293 58.49%, 44.64%, 31.30% and 21.67%, respectively. That indicated that the primary and secondary
 294 sources of HONO were mainly at the surface (Section 4.2). The vertical gradient of daily averaged O_3
 295 concentration was also not obvious, which was associated with its vertical mixing and photochemical

296 production (Yin et al., 2017). As shown in Figure S8, the corresponding TROPOMI O₃ profiles in Nam
 297 Co and O₃ profiles measured by lidar and ozonesonde around Nam Co reported in several previous
 298 studies also exhibited an exponential shape (Fang et al., 2019; Zhang et al., 2020; Yu et al., 2022). The
 299 O₃ average concentration at 0.0-0.2 km was 63.030 ppb, and the ratios of O₃ at 0.4-0.6 km, 0.8-1.0 km,
 300 1.2-1.4 km and 1.6-1.8 km to those at surface were 89.25%, 82.44%, 80.16% and 79.13%, respectively.

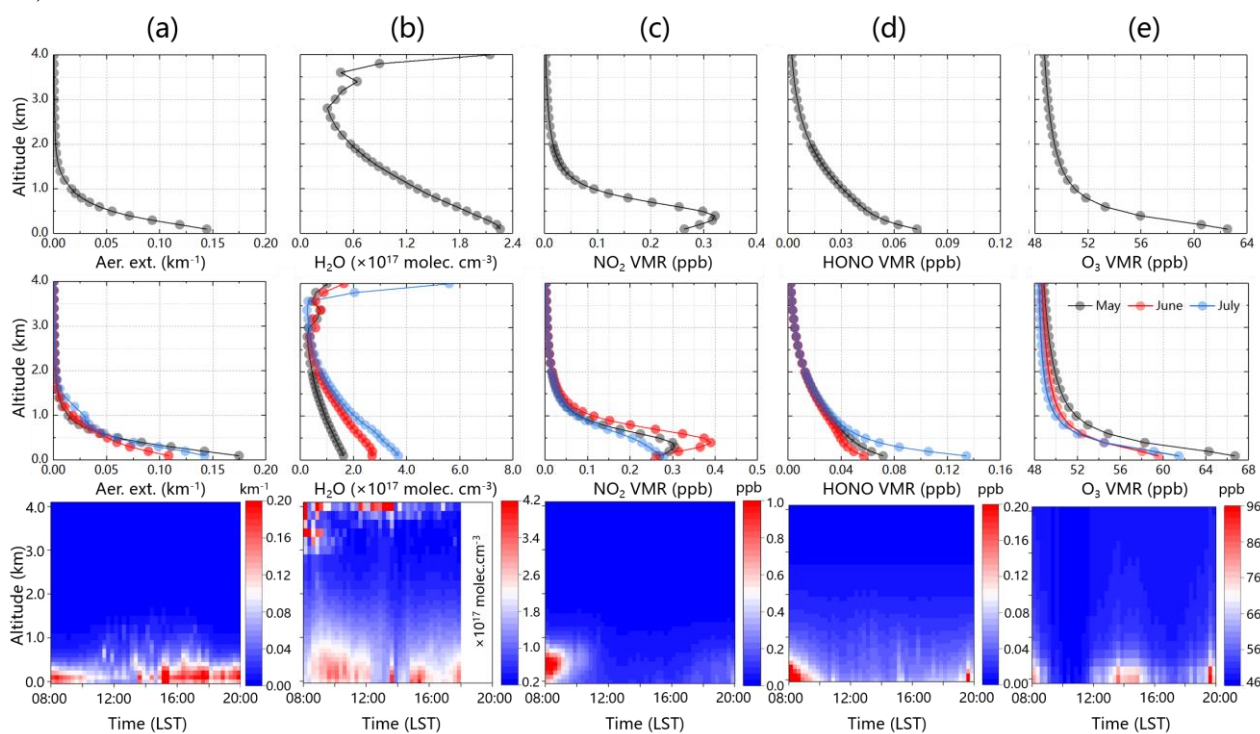


301
 302 Figure 4. Time series of daily averaged (a) aerosol extinction, (b) H₂O, (c) NO₂, (d) HONO, and (e) O₃
 303 monitored by MAX-DOAS at 0-0.2, 0.4-0.6, 0.8-1.0, 1.2-1.4 and 1.6-1.8 km five height layers from 01
 304 May to 09 July 2019.

305 3.2 Vertical distributions of aerosol, H₂O, NO₂, HONO and O₃

306 The first row in Figure 5 provided the averaged vertical profiles of aerosol, H₂O, NO₂, HONO and O₃
 307 from May to July 2019. We found that the vertical profiles of aerosol, H₂O, HONO and O₃ all
 308 exhibited an exponential shape with maximum values near the surface, while NO₂ exhibited a Gaussian
 309 shape with the maximum value of 0.321 ppb occurring at 0.3-0.4 km layer. In addition to the effect of
 310 NO_x transport, Xu et al. (2018) also revealed that the long-range high-altitude transport process from
 311 the northern south Asian subcontinent can significantly enhance the Nam Co's peroxyacetyl nitrate
 312 (PAN) level which is a reservoir of NO_x. As shown in the second row of Figure 5, the monthly
 313 averaged aerosol vertical profiles from May to July 2019 all exhibited an exponential shape, and varied
 314 in the order of May (0.17 km⁻¹) > July (0.14 km⁻¹) > June (0.11 km⁻¹). Xu et al. (2018) and Neupane et
 315 al. (2019) also reported a similar monthly variations of black carbon (BC) from May to July over the
 316 TP, and revealed that it was mainly associated with the anthropogenic emissions (i.e. biomass burning)
 317 and its transport from south Asia. The monthly averaged vertical profile of H₂O in May and July
 318 exhibited an exponential shape, while its maximum concentration layer slightly elevated to 0.1-0.2 km

319 in June which was related to the strongest monsoon transport (Figure S9). It varied in the order of July
 320 (3.68×10^{17} molec cm^{-3}) > June (2.71×10^{17} molec cm^{-3}) > May (2.26×10^{17} molec cm^{-3}), and its
 321 maximum concentration occurring in July was strongly associated with the enhanced evaporation from
 322 the Nam Co lake (Xu et al., 2011). The monthly averaged vertical profiles of NO_2 all exhibited a
 323 Gaussian shape from May to July, and its maximum values mainly distributed at 0.2-0.4 km layer
 324 varying in the order of June (0.39 ppb) > May (0.31 ppb) > July (0.28 ppb). It indicated that the
 325 regional transport from the NO_x produced from ice and snow under strong shortwave radiation (Figure
 326 S7), NO_2 emitted from vehicles due to the increased tourism, anthropogenic emissions from local
 327 residents (i.e. biomass burning and religious activities) played an important role in the vertical
 328 distribution characteristic of NO_2 (Boxe et al., 2005; Chen et al., 2019). The monthly averaged vertical
 329 profiles of HONO from May to July all exhibited an exponential shape, with maximum values near the
 330 surface varying in the order of July (0.13 ppb) > May (0.07 ppb) > June (0.06 ppb). The local direct
 331 emissions from biomass burning, vehicles and soil should be main sources of the surface HONO (Xing
 332 et al., 2021). Moreover, the heterogeneous reaction of NO_2 on wet surfaces should be another important
 333 source of HONO at different height layers (Section 4.2). For example, the aerosol extinction coefficient,
 334 and the concentrations of H_2O and NO_2 were all relatively large at the bottom layer in July,
 335 correspondingly, we observed the highest concentration of HONO near the surface in this month. The
 336 monthly averaged O_3 vertical profiles all showed an exponential shape from May to July, and its
 337 surface concentration varied in the order of May (66.71 ppb) > July (61.45 ppb) > June (59.55 ppb).
 338 This kind of monthly variation trend of O_3 was also reported by several previous studies (Yin et al.,
 339 2017; Xu et al., 2018). The O_3 in Nam Co was mainly sourced from stratospheric intrusion,
 340 photochemical reactions, long-range transport and local vertical mixing (Yin et al., 2017; Chen et al.,
 341 2019).



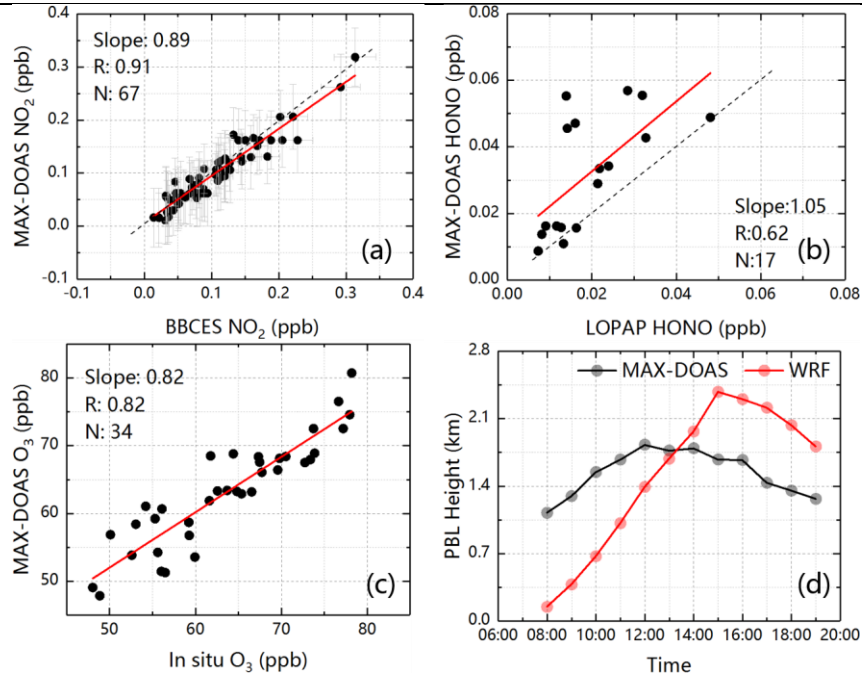
342
 343 Figure 5. Vertical profiles of (a) aerosol extinction, (b) H_2O , (c) NO_2 , (d) HONO, and (e) O_3 . The top
 344 row shows the averaged vertical profiles from 01 May to 09 July 2019. The middle row shows the
 345 monthly averaged vertical profiles. The bottom row shows the averaged diurnal vertical profiles from
 346 01 May to 09 July 2019.

347 The third row in Figure 5 illustrated the averaged diurnal variations in vertical profiles of aerosol, H_2O ,
 348 NO_2 , HONO and O_3 from May to July 2019. Aerosol mainly distributed under 1.0 km, especially 0.6
 349 km, and its mixing height was gradually increased with the rise of the PBL height after 12:00.
 350 Moreover, the diurnal variation of aerosol showed a bi-peak pattern, which was in line with the
 351 investigation reported by Pokharel et al. (2019). The first peak occurred between 08:00-10:00, and

352 another appeared after 15:00. The first peak should be attributed to the local emission of aerosol and
353 the diurnal cycle of PBL (Zhang et al., 2017; Pokharel et al., 2019). The second peak was driven by
354 regional transport and the interaction between local sandy silt loam surface and local meteorology. The
355 high wind speed (> 4.5 m/s) at surface appeared after 15:00, which coincided with the appearance of
356 the second aerosol peak (Figure S3). Moreover, the high extinction during the second peak was
357 extended to 1.0 km associated with the wind speed larger than 8 m/s (Figure S10), which created a
358 favorable condition for high-altitude aerosol transport. H₂O mainly distributed under 1.0 km and above
359 3.0 km, and its diurnal variation exhibited a multi-peak pattern. The first peak appeared between
360 08:00-12:00, which was mainly affected by the monsoon driven long-range transport of H₂O (Cong et
361 al., 2009; Xu et al., 2020). The second and third peaks occurred at 15:00-16:00 and after 17:00,
362 respectively. In addition to long-range transport, the enhanced evaporation from the Nam Co lake also
363 significantly contributed to the appearance of these two peaks of H₂O (Xu et al., 2011). NO₂ mainly
364 distributed at 0.2-0.4 km, and peaked before 10:00 and after 18:00 which were dominated by the effects
365 of local emissions and regional transport from the NO_x formed through ice and snow on the top of Mt.
366 Tanggula under strong ultraviolet radiation (Figure S7) (Boxe et al., 2005; Fisher 2005; Chen et al.,
367 2019; Lin et al., 2021). Moreover, its diurnal mixing height was obviously correlated to the diurnal
368 evolution of PBL height. HONO mainly distributed under 1.0 km, especially 0.4 km. Its diurnal
369 variation showed a multi-peak pattern with three obvious peaks before 10:00, 15:00-16:00, and after
370 19:00. In addition to local emissions (i.e. vehicle emission, biomass burning and soil emission), the
371 heterogeneous reaction of NO₂ on wet surfaces should be also an important HONO source (Xing et al.,
372 2021). We found that there were larger aerosol extinction (> 0.12 km⁻¹) and higher concentrations of
373 NO₂ (> 0.20 ppb) and H₂O ($> 2.27 \times 10^{17}$ molec cm⁻³) around three HONO peaks. O₃ mainly
374 distributed under 0.4 km, and its diurnal variation exhibited a multi-peak pattern with three peaks
375 appearing before 09:00, 13:00-15:00 and after 19:00. The appearance of O₃ peaks was mainly
376 associated with the influence of the complex topography of the TP, long-range transport, local vertical
377 mixing and stratospheric intrusion (Yin et al., 2017; Chen et al., 2019; Qian et al., 2022). The active
378 photochemical reaction should be another important source of O₃, especially for its second peak at
379 13:00-15:00.

380 **3.3 Validation with independent data**

381 In order to validate the MAX-DOAS dataset, we extracted the concentrations of NO₂, HONO and O₃ at
382 the bottom layer (0.0-0.1 km) from their corresponding vertical profiles to compare with in situ
383 measurements. As shown in Figure 6(a-c), we found good agreements between MAX-DOAS and in
384 situ observations with Pearson correlation coefficients (R) of 0.91, 0.62 and 0.82 (regression slope of
385 0.89, 1.05 and 0.82) for NO₂, HONO and O₃, respectively. That indicated the good reliability of trace
386 gases from MAX-DOAS retrievals. Moreover, we also compared the MAX-DOAS PBL and WRF PBL,
387 and a similar variation trend was found. However, WRF PBL showed a significantly difference in
388 height values with MAX-DOAS PBL before 12:00. That should be due to the simulation uncertainties
389 for WRF model at Tibetan plateau with complex topography and meteorology (Yang et al., 2016; Xu et
390 al., 2019).



391

392 Figure 6. Validations of (a) MAX-DOAS NO₂ vs in situ NO₂ (error bars represent the retrieved errors
 393 of NO₂ from MAX-DOAS and BBCES), (b) MAX-DOAS HONO vs LOPAP HONO, (c)
 394 MAX-DOAS O₃ vs in situ O₃, and (d) MAX-DOAS PBL vs WRF PBL.

395 4 Discussion

396 4.1 OH production

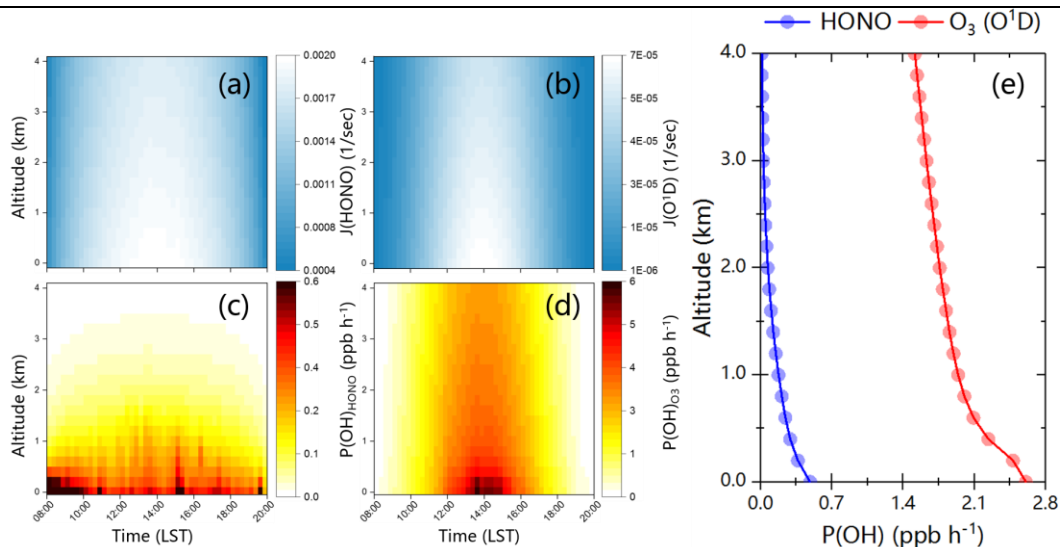
397 HONO and O₃ are two important precursors of OH radical to enhance the AOC (Kleffmann et al., 2005;
 398 Ryan et al., 2018; Xing et al., 2021). In order to evaluate the AOC on the TP, we tried to analyze the
 399 OH production from HONO and O₃ at different height layers through vertical observations and TUV
 400 calculations. The OH production rates from HONO and O₃ were calculated using the following two
 401 equations:

$$402 P(OH)_{HONO} = J(HONO) \times [HONO]$$

$$403 P(OH)_{O_3} = 2 \times f \times J(O(^1D)) \times [O_3]$$

404 Where $J(HONO)$ and $J(O(^1D))$ were the photolysis rates of HONO and O(¹D) calculated using TUV
 405 model. O(¹D) was the product from O₃ photolysis by UV radiation. f was the fraction of the process
 406 $O(^1D) + H_2O \rightarrow 2OH$.

407 Figure 7(a-b) showed the averaged diurnal vertical distributions of the photolysis rates $J(HONO)$ and
 408 $J(O(^1D))$ from May to July 2019. We found that the maximum $J(HONO)$ and $J(O(^1D))$ were all
 409 appeared at the bottom layer between 12:30 and 15:30 with values of 2.0×10^{-3} and $6.75 \times 10^{-5} \text{ s}^{-1}$,
 410 respectively. The maximum values were usually larger than that at low-altitude areas due to the
 411 stronger solar UV radiation on the TP (Su et al., 2008; Xing et al., 2021; Yang et al., 2021; Liu et al.,
 412 2022), but being consistent with the values on the TP reported by Lin et al. (2008). Moreover, it should
 413 be noted that the values of $J(HONO)$ and $J(O(^1D))$ all decreased with the increase of altitude, which
 414 was significantly different with previous studies in low altitudes (Ryan et al., 2018; Xing et al., 2021;
 415 Xu et al., 2021).



416
 417 Figure 7. Averaged diurnal vertical profiles of the (a) photolysis rate $J(HONO)$, (b) photolysis rate
 418 $J(O^1D)$, (c) OH radical production rates from HONO photolysis, (d) OH radical production rates from
 419 O_3 photolysis. (e) shows the averaged vertical profiles of OH radical production rates from HONO and
 420 O_3 photolysis from 01 May to 09 July 2019.

421 Figure 7(c-d) showed the averaged diurnal vertical profiles of OH production rates from HONO and O_3
 422 photolysis from May to July 2019. $P(OH)_{HONO}$ exhibited a multi-peak pattern which mainly appeared
 423 before 10:00, 15:00-16:00, and after 19:00 at 0-0.4 km with a maximum value of 0.81 ppb/h. While
 424 $P(OH)_{O_3}$ showed a unimodal pattern occurring at 13:00-15:00 under 0.4 km with a maximum value of
 425 6.20 ppb/h. The averaged vertical profiles of $P(OH)_{HONO}$ and $P(OH)_{O_3}$ during the observation were
 426 depicted in Figure 7(e). We found that the maximum values of $P(OH)_{HONO}$ (0.49 ppb/h) and $P(OH)_{O_3}$
 427 (2.61 ppb/h) all appeared at the bottom layer, and decreased with height. That indicated O_3 was an
 428 important contributor of OH production ($> 80\%$) on the TP, which was about 5-6 times to HONO.
 429 Moreover, the OH production rates from HONO and O_3 in other cities of China were depicted in Table
 430 3. The contribution percentage of O_3 to $P(OH)$ in Nam Co was significantly higher than that in other
 431 cities, which was due to the relatively high concentrations of O_3 and H_2O , and the strong radiation in
 432 Nam Co. In addition, $P(OH)_{HONO}$ in Nam Co was close to that in relatively dry areas (i.e. Beijing and
 433 Xianghe), but slightly lower than that in areas with relatively high humidity which can enhance the
 434 heterogeneous production of HONO (Ryan et al., 2018; Liu et al., 2019; Xing et al., 2021).

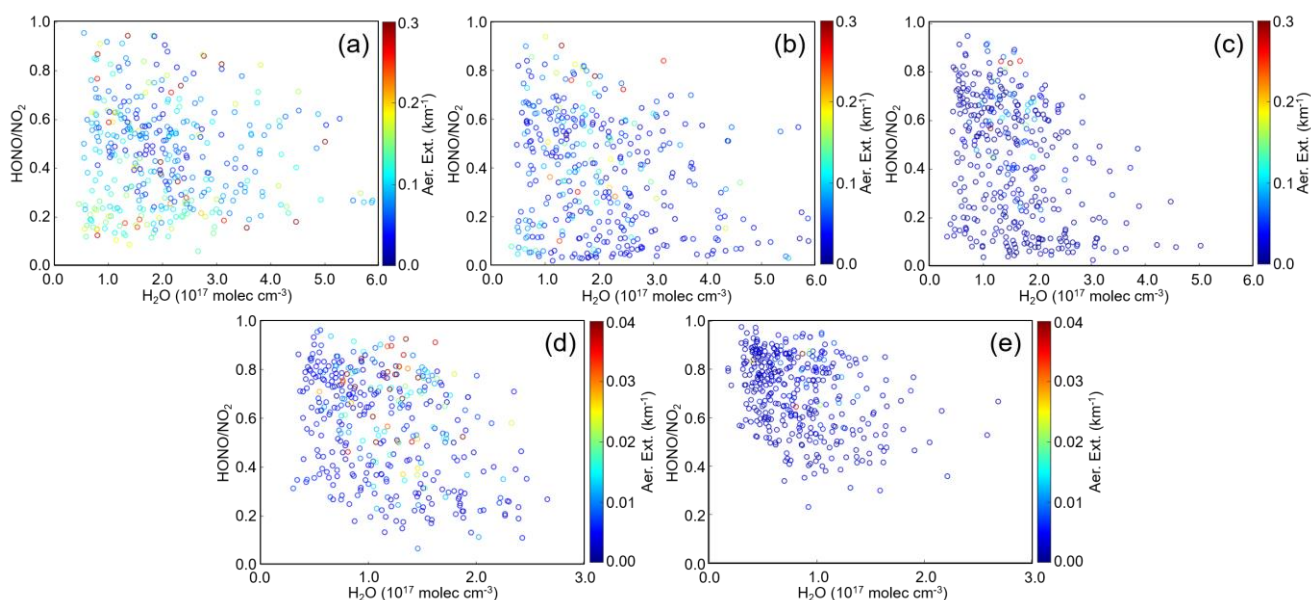
435 Table 3. The maximum OH production rates contributed from HONO and O_3 at different locations.

Location	Date	$P(OH)_{HONO}$ (ppb/h)	$P(OH)_{O_3}$ (ppb/h)	References
Xianghe (China)	Jul. 2008-Apr. 2009	~0.80 in Spring ~0.70 in Summer	~0.20 in Spring, ~0.45 in Summer	Hendrick et al. (2014)
Beijing (China)	Mar. 2010-Dec. 2012	~1.25 in Spring, ~0.70 in Summer	~0.10 in Spring, ~0.55 in Summer	Hendrick et al. (2014)
East China Sea (China)	Jun. 2017	~1.75	~1.20	Cui et al. (2019)
Chengdu (China)	Aug.-Sep. 2019	~3.25	-	Yang et al. (2021)
Qingdao (China)	Jul.-Aug. 2019	~1.30	~1.00	Yang et al. (2021)
Nam Co (China)	May-Jul. 2019	0.81	6.20	This study

436 4.2 Possible daytime HONO sources

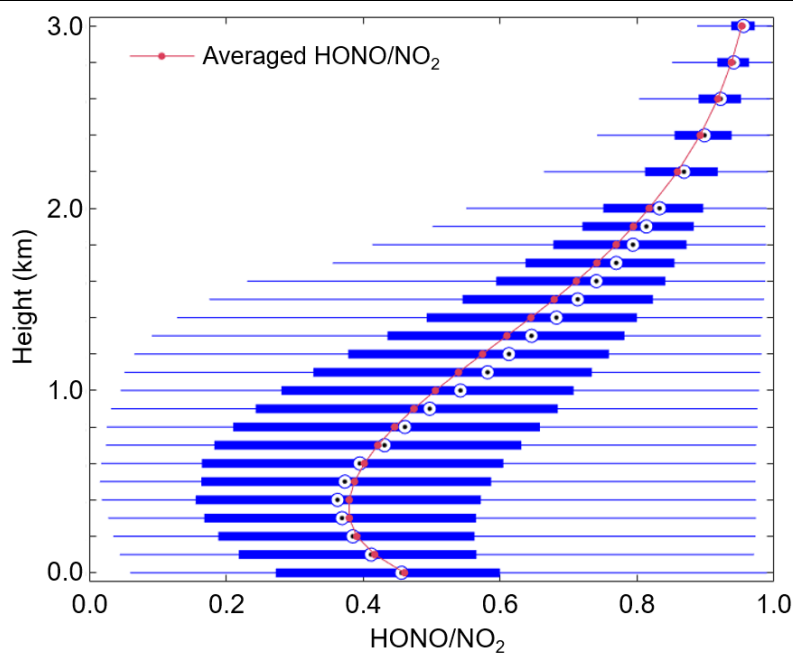
437 Atmospheric HONO mainly sourced from direct emission, homogeneous reaction and heterogeneous
 438 reaction (Fu et al., 2019; Ren et al., 2020; Chai et al., 2021; Crilley et al., 2021; Li et al., 2021). There
 439 were less anthropogenic emissions for HONO around NAMORS, however, the open burning of crop
 440 residues and soil emissions should be important HONO sources considering the pasture environment
 441 and large amounts of animal manure (Cui et al., 2021a; 2021b). Moreover, the background of low-level
 442 NO on the TP led to the homogeneous reaction not to be the main source of HONO at NAMORS
 443 (Lin et al., 2019; Xing et al., 2021; Li et al., 2022). Heterogeneous reaction of NO_2 on wet surfaces
 444 became an important potential source of HONO around NAMORS, which affected by the humidity,

445 temperature, solar radiation, aerosol concentration and corresponding specific surface area. In order to
 446 remove the effect of diurnal PBL evolution, we used HONO/NO₂ to indicate the extent of the
 447 heterogeneous reaction process. As shown in Figure 8, scatter plots between HONO/NO₂ and H₂O
 448 were illustrated. We found that the maximum value of HONO/NO₂ appeared around water vapor being
 449 around 1.0×10^{17} molec cm⁻³ under 1.0 km, and being around $0.5-1.0 \times 10^{17}$ molec cm⁻³ at 1.0-2.0 km
 450 height layer. This phenomenon of HONO/NO₂ firstly increasing and then decreasing with the
 451 increasing of H₂O (or relative humidity) was usually found in low-altitude areas in previous studies
 452 (Wang et al., 2013; Liu et al., 2019; Xing et al., 2021; Xu et al., 2021). When the H₂O was greater than
 453 above mentioned critical values at different heights, HONO/NO₂ gradually decreased, which was
 454 related to the efficient uptake of HONO and the decrease of NO₂ reactivity with the increase of H₂O
 455 (Liu et al., 2019; Xu et al., 2021). That indicated H₂O has significant enhancement for the conversion
 456 rate of NO₂ to HONO. Moreover, we found that the high value areas of HONO/NO₂ at above five
 457 height layers were all accompanied by high aerosol extinction (> 0.15 km⁻¹ under 1.0 km, and > 0.02
 458 km⁻¹ at 1.0-2.0 km). It indicated that aerosol surface has contribution to the heterogeneous reaction
 459 process of NO₂. The scatter plots between HONO and NO₂ at above five layers (Figure S11) also
 460 confirmed the possibility of the NO₂ heterogeneous reaction to generate HONO on the TP, and the
 461 contribution of atmospheric H₂O and aerosol extinction to this process.



462
 463 Figure 8. Scatter plots between HONO/NO₂ and H₂O colored by aerosol extinction at (a) 0.0-0.2 km, (b)
 464 0.4-0.6 km, (c) 0.8-1.0 km, (d) 1.2-1.4 km, and (e) 1.6-1.8 km from 1st May to 9th July 2019.

465 In Figure 9, the vertical profile of HONO/NO₂ from May to July 2019 was depicted. We found that
 466 HONO/NO₂ firstly decreased and then increased with the increasing of height, which was opposite to
 467 previous studies in low-altitude areas (Meng et al., 2020; Zhang et al., 2020; Xing et al., 2021; Xu et al.,
 468 2021). The minimum average HONO/NO₂ occurred at 0.3-0.4 km height layer with a value of 0.37.
 469 The relatively high values of HONO/NO₂ at the bottom layer should be related to the non-deducted
 470 HONO direct emissions.



471

472 Figure 9. Statistics for the vertical profile of HONO/NO₂ from 1st May to 9th July 2019. The left and
 473 right of the blue box represent the 25th and 75th percentiles, respectively; the dot within the box
 474 represents the mean.

475 4.3 Possible daytime O₃ sources

476 In addition to local photochemistry process, long-range transport was the main source of O₃ on the TP
 477 (Yin et al., 2017; Xu et al., 2018). To further understand the transport pathway and potential source of
 478 O₃, cluster analysis, WPSCF and WCWT models were used to assess the regional representivity of
 479 O₃ at five typical heights (200 m, 600 m, 1000 m, 1400 m and 1800 m). As shown in Figure S12 and
 480 Table 4, the backward trajectories arriving at NAMORS during the observation were classified into
 481 three clusters at 200 m, 600 m, 1400 m, 1800 m, and four clusters at 1000 m. We found that cluster 3
 482 was associated with the highest O₃ concentration at 200 m (65.48 ± 17.41 ppb) and 1800 m ($49.69 \pm$
 483 2.21 ppb), and cluster 1 were related to the highest O₃ concentration at 600 m (54.67 ± 6.94 ppb), 1000
 484 m (51.61 ± 3.84 ppb) and 1400 m (50.51 ± 2.89 ppb). These two clusters were all originating from
 485 northwestern of south Asian subcontinent passing through Himalayas, which was also reported by Yin
 486 et al. (2017) during springtime from 2011 to 2015. In Figure S13 and 10, WPSCF and WCWT analysis
 487 told us that the high O₃ concentration at above heights potentially sourced from northern India, central
 488 Pakistan, Nepal, western Bhutan and northern Bangladesh through long-range transport. It should be
 489 noted that the potential contribution to O₃ at NAMORS at 200 m from above potential source areas
 490 were all over 40 ppb. These contributions from the mentioned potential source areas at other four
 491 heights were also over 20-30 ppb. The massive fire emissions during springtime were an important
 492 source of O₃ in south Asia (Jena et al., 2015), and the obvious burning during the observation was
 493 observed in Figure S14. Moreover, the abundant precursors and high photochemical activity were
 494 another significant sources of O₃ in south Asia (Kumar et al., 2012; Sharma et al., 2017).

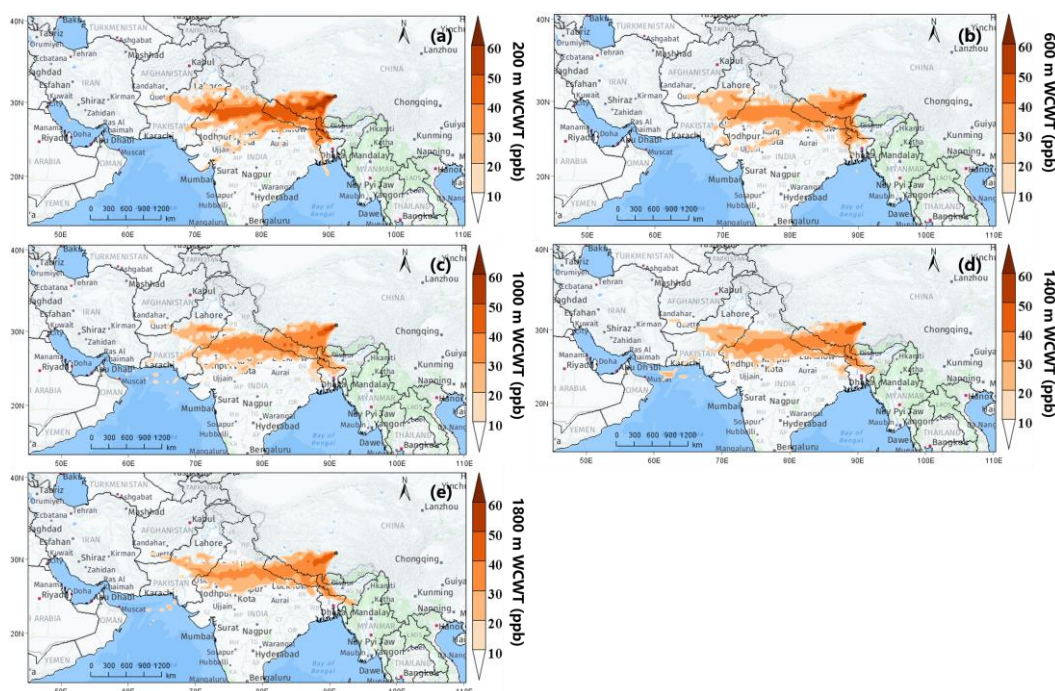
495 In addition, Figure 10 showed that the contribution of O₃ transported from Himalayas can even up to
 496 50 ppb, especially under 600 m. Several previous studies have revealed that the stratospheric O₃
 497 intrusion events were frequent in the Himalayas during spring and summer (Cristofanelli et al., 2010;
 498 Chen et al., 2011; Škerlak et al., 2014; Putero et al., 2016). Therefore, the O₃ from stratospheric
 499 intrusions in the Himalayas can affect the O₃ at NAMORS through long-range transport.

500 Table 4. Trajectory ratios and averaged O₃ concentration for all trajectory clusters arriving in Nam Co
 501 at 200 m, 600 m, 1000 m, 1400 m and 1800 m from May to July 2019.

	Cluster	Traj_ratio	O ₃ concentration (ppb)
			Mean \pm SD

200 m	1	55.86%	61.50 ± 18.15
	2	11.85%	54.57 ± 14.67
	3	32.28%	65.48 ± 17.41
	All	100.00%	61.14 ± 17.74
600 m	1	62.55%	54.67 ± 6.94
	2	14.32%	50.43 ± 6.64
	3	23.13%	53.27 ± 7.63
	All	100.00%	53.39 ± 7.26
1000 m	1	49.16%	51.61 ± 3.84
	2	8.81%	49.60 ± 3.99
	3	22.73%	50.72 ± 4.21
	4	19.30%	51.39 ± 4.49
	All	100.00%	50.98 ± 4.30
1400 m	1	80.14%	50.51 ± 2.89
	2	4.95%	49.12 ± 2.73
	3	14.92%	49.44 ± 3.85
	All	100.00%	50.07 ± 3.15
1800 m	1	83.75%	49.68 ± 2.55
	2	0.00%	49.07 ± 2.23
	3	16.25%	49.69 ± 2.21
	All	100.00%	49.59 ± 2.49

502



503

504 Figure 10. Spatial distributions of WCWT values for O₃ at (a) 200 m, (b) 600 m, (c) 1000 m, (d) 1400
 505 m, and (e) 1800 m height layers from 01st May to 09th July 2019 over CAS (NAMORS).

506 5 Summary and conclusions

507 MAX-DOAS measurements were performed to clarify the vertical distributions of several atmospheric
 508 components (aerosol, H₂O, NO₂, HONO and O₃), and to explore the AOC in vertical space in Nam Co
 509 from May to July 2019. The MAX-DOAS NO₂, HONO and O₃ agreed well with in situ measurements,
 510 with correlation coefficients of 0.91, 0.62 and 0.82, respectively. We found that the averaged vertical

511 profiles of aerosol, H₂O, HONO and O₃ all exhibited an exponential shape, while NO₂ showed a
512 Gaussian shape with a maximum value of 0.32 ppb appearing at 300-400 m. The maximum
513 concentrations of monthly averaged aerosol (0.17 km⁻¹) and O₃ (66.71 ppb) appeared on May, H₂O
514 (3.68×10^{17} molec cm⁻³) and HONO (0.13 ppb) appeared on July, and NO₂ (0.39 ppb) occurred on
515 June. For the diurnal variation, above five species all mainly distributed under 1.0 km, and mostly
516 exhibited a multi-peak pattern considering the effect of regional transport and local chemical reaction.

517 O₃ and HONO were important source of OH on the TP. The diurnal averaged OH production rate from
518 HONO during the observation exhibited a multi-peak pattern appearing before 10:00, 15:00-16:00 and
519 after 19:00 under 0.4 km with the maximum value of 0.81 ppb/h. The OH production rate from O₃
520 shown a unimodal pattern occurring at 13:00-15:00 under 0.4 km with the maximum value of 6.20
521 ppb/h which was obviously higher than that at low-altitude areas. In addition to direct emission, the
522 heterogeneous reaction of NO₂ on wet surfaces was also an important source of HONO in Nam Co. We
523 found that HONO/NO₂ first increasing and then decreasing with the increasing of H₂O. The maximum
524 value of HONO/NO₂ appeared around H₂O being around 1.0×10^{17} molec cm⁻³ under 1.0 km, and
525 being around $1.0\text{-}2.0 \times 10^{17}$ molec cm⁻³ at 1.0-2.0 km height layer. Moreover, high values of
526 HONO/NO₂ usually accompanied by high aerosol extinction. O₃ under 2.0 km were potentially sourced
527 from Himalayas, northern India, central Pakistan, Nepal, western Bhutan and northern Bangladesh
528 through long-range transport. Our results draw a picture of further understanding the spatial and
529 temporal variations in oxidation chemistry under PBL and provided a new perspective for source
530 analysis of major atmospheric components through vertical observation on the TP.

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540 Competing interests

541 All authors declare that they have no conflict of interest or financial conflicts to disclose.

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