

Summertime response of ozone and fine particulate matter to mixing layer meteorology over the North China Plain

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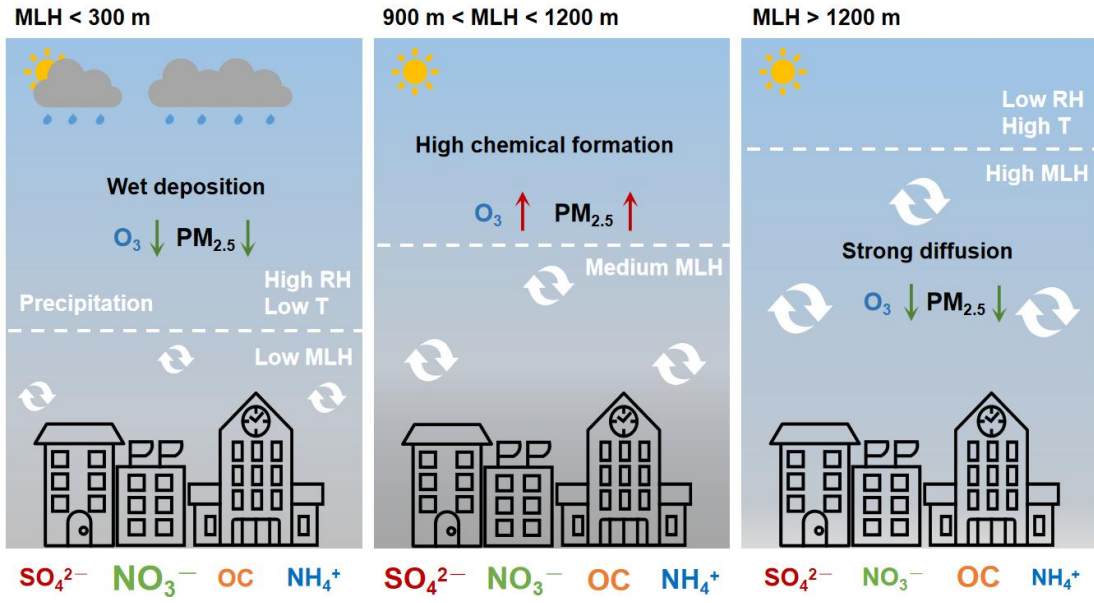
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Abstract. Measurements of surface ozone (O_3), $PM_{2.5}$ and its major secondary components (SO_4^{2-} , NO_3^- , NH_4^+ , and OC), mixing layer height (MLH) and other meteorological parameters were made in the North China Plain (NCP) during warm seasons (June–July) in 2021. The observation results showed that the summertime regional MDA8 O_3 initially increased and reached the maximum ($195.88 \mu\text{g m}^{-3}$) when MLH at around 900–1800 m, then turned to decrease with further evolution of MLH. Interestingly, synchronized increases in $PM_{2.5}$ concentration along with the development of the mixing layer (MLH < 1200 m) have been witnessed, and the positive response of $PM_{2.5}$ to MLH was significantly associated with the increase in SO_4^{2-} and OC. It was found that this increasing trend of $PM_{2.5}$ with elevated MLH was not only determined by the effect of wet deposition process but also by the enhanced secondary chemical formation, which was related to appropriate meteorological conditions ($50 \% < RH < 70 \%$) and increased availability of atmospheric oxidants. Air temperature played a minor role in the change characteristics of $PM_{2.5}$ concentration, but greatly controlled the opposite change characteristics of SO_4^{2-} and NO_3^- . The concentrations of $PM_{2.5}$ and its major secondary components, as well as SOR and NOR, increased synchronously with elevated MDA8 O_3 concentration, and the initial increase of $PM_{2.5}$ along with the increased MLH corresponded well with that of MDA8 O_3 . We highlight that the correlation between MLH and secondary air pollutants should be treated with care in hot seasons, and the superposition-composite effects of $PM_{2.5}$ and O_3 along with the evolution of mixing layer should be considered when developing $PM_{2.5}$ - O_3 coordinated control strategies.



31
32

33 **1 Introduction**

34 Surface ozone (O_3) and $PM_{2.5}$ (atmospheric fine particles with an aerodynamic diameter of less
35 than $2.5\ \mu\text{m}$) are important air pollutants in the atmosphere and have aroused a lot of attention from the
36 public due to their adverse health impact (Jiang et al., 2018; Cohen et al., 2017; Gao and Ji, 2018).
37 Even though stringent clean air actions have been implemented in China during the past decade, high
38 concentrations of O_3 and/or $PM_{2.5}$ exceeding the national air quality standards still occurred during
39 warm seasons (June–August) in China, especially in the North China Plain (NCP), the economic center
40 of China (Dai et al., 2023). O_3 is a secondary pollutant originated from photochemical oxidation of
41 volatile organic compounds (VOC) and carbon monoxide (CO) in the presence of nitrogen oxides
42 (NO_x), and $PM_{2.5}$ is mainly determined by the atmospheric processes of emissions and secondary
43 formation from gaseous precursors. In addition to air pollutant emissions, meteorological conditions
44 play critical roles in the formation of $PM_{2.5}$ and O_3 (Miao et al., 2021). Mixing layer height (MLH),
45 which influences the vertical mixing within the pollution mixing layer and determines the dilution of
46 pollutants emitted near the ground (Haman et al., 2014; Zhu et al., 2018; Lou et al., 2019), often serves
47 as a critical physical parameter in atmospheric environmental evaluation. Elucidating the association of
48 MLH with surface O_3 and $PM_{2.5}$ is fundamental for the development of $PM_{2.5}$ - O_3 coordinated control
49 strategies.

50 The response of air pollution to MLH was changeable and complicated (Miao et al., 2021).
51 Previous works frequently assumed that the narrowing of mixing layer resulted in accumulation of
52 pollutants near the ground and the increase in MLH was expected to reduce $PM_{2.5}$ concentration due to
53 dilution (Murthy et al., 2020; Du et al., 2013). However, the relationship between mixing layer
54 structure and $PM_{2.5}$ concentration depends on the site, observation period, and the properties of MLH
55 retrievals (Geiß et al., 2017; Lu et al., 2019). Even though the link between $PM_{2.5}$ concentrations and
56 MLH has been investigated in many studies, most observations were conducted in winter conditions
57 and few studies in hot seasons. Interestingly, in some cities, such as Delhi (Murthy et al., 2020) and
58 Shanghai (Pan et al., 2019; Miao et al., 2021), the increase in $PM_{2.5}$ has been observed when MLH
59 increased during summertime. As for O_3 , the relationship between the changes in the MLH and O_3
60 concentrations is very complex. Both increase or decrease of O_3 has been observed corresponded to the
61 growth of MLH. First, O_3 concentration decreases along with the increase of MLH owing to dilution.

62 Second, an increase in MLH generally promotes the downward mixing of upper air containing higher
63 O₃ (Ma et al., 2021; Haman et al., 2014; Xu et al., 2018). In addition, the meteorological conditions
64 along with the changes of MLH can influence O₃ concentrations through effecting O₃ gaseous
65 precursors or production rates (Porter and Heald, 2019; Zhang et al., 2022). The combined effects of
66 these processes ultimately determine whether O₃ decreases or increases.

67 Other meteorological variables in the mixing layer were also found to significantly affect PM_{2.5}
68 and O₃ concentrations. The poor air quality in the NCP was tightly associated with near-surface
69 southerly winds and warm stagnant conditions during summertime (Zhang et al., 2015a). The increase
70 in PM_{2.5} concentration often coincided with high relative humidity (RH) conditions (Liu et al., 2017b),
71 which was beneficial to liquid-phase heterogeneous reactions and fine particle hygroscopic growth
72 (Seinfeld and Pandis, 2006; Wang et al., 2016; Zhang et al., 2015b). Temperature was essential to
73 secondary chemical reaction (Dawson et al., 2007). The increase in temperature can promote chemical
74 reaction rates, but also stimulate the evaporation of semi-volatile aerosol components, such as nitrate
75 (Wen et al., 2018). For O₃, elevated O₃ concentrations generally happened on days with strong sunlight
76 and low wind speeds, which favored the photochemical production and the accumulation of O₃ and its
77 precursors. Several studies have shown that O₃ was significantly positive correlated with temperature,
78 but negatively correlated with RH (Li et al., 2021; Hou and Wu, 2016; Steiner et al., 2010).

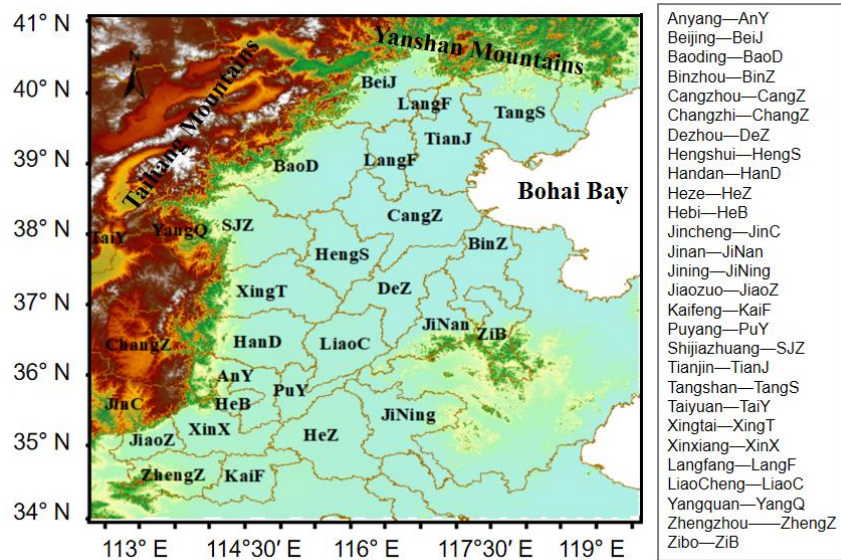
79 Long-term PM_{2.5} composition measurements in the NCP showed an increase in the contributions
80 of secondary species, e.g., sulfate (SO₄²⁻), nitrate (NO₃⁻), ammonium (NH₄⁺), and organic matter (OM),
81 to total PM_{2.5} in recent years (Cheng et al., 2019; Wang et al., 2022b). As air quality improved (PM_{2.5}<
82 50 μg m⁻³), the correlation between O₃ and PM_{2.5} tended to change from negative to positive in China
83 (Chu et al., 2020). One speculative reason for this phenomenon is that PM_{2.5} does not reduce actinic
84 flux and HO₂ radical significantly when the PM_{2.5} concentration was low. On the other hand, PM_{2.5} and
85 O₃ tend to be positively correlated possibly due to their common precursors, such as VOCs and NO_x,
86 and their simultaneous generation in photochemical reactions. In addition, the generation of O₃ would
87 cause the enhancement of atmospheric oxidation capacity, and catalyze the generation of the secondary
88 PM_{2.5} (Cheng et al., 2019; Kang et al., 2021; Wu et al., 2022). Even though some studies have
89 discussed the correlations between MLH and some secondary pollutants, the understanding of the
90 interaction between O₃ and PM_{2.5} (including its major components) along with the evolution of mixing
91 layer during warm seasons remained poor owing to the limited observations of PM_{2.5} chemical species

92 involved. The regional-scale observation can represent the variation characteristics for this area and
 93 avoid the spatial heterogeneity between the sites. However, to the best of our knowledge, previous
 94 observational studies were mostly limited to specific cities. Therefore, it's encouraged to analyze
 95 multiple data sources to determine overall trends rather than making conclusions based on a single
 96 dataset.

97 According to the hourly concentrations of PM_{2.5} and MDA8 O₃ in China over the years of
 98 2013–2020, the months of June and July can well represent the typical characteristics of O₃–PM_{2.5}
 99 coordinated pollution during warm seasons in the North China Plain (NCP) (Dai et al., 2023). To
 100 enhance the understanding of the linkages between mixing layer structure and air pollution, in this
 101 study, a regional-scale field observation of meteorological factors, O₃, PM_{2.5} concentration and its
 102 secondary composition were conducted in the NCP from June 1 to July 31, 2021. For the first time, the
 103 potential association among ground-level observed O₃, PM_{2.5} and its dominant components, and mixing
 104 layer meteorological conditions will be explored in the NCP during summertime.

105 2 Data and methods

106 2.1 Measurements



107
 108 **Figure 1.** Location of monitoring stations in the NCP.

109 In this study, observation was made in the North China Plain (NCP) from June 1 to July 31, 2021.
 110 The air pollution observation stations in the NCP covered two megacities (BeiJ and TianJ) and 26

111 surrounding cities. The geographic locations of these stations are marked in Figure 1. The North China
112 Plain (NCP) is surrounded to the west by the Taihang Mountains, to the north by the Yan Mountains,
113 and to the east by the Bohai Sea. The hourly concentrations of ground-level O₃, PM_{2.5} and its major
114 components (SO₄²⁻, NO₃⁻, NH₄⁺, and OC), and meteorological variables, including air temperature,
115 relative humidity (RH), wind speed (WS) and direction (WD), and 24-h accumulated precipitation, at
116 the sites were obtained from the platform of National Atmospheric Particulate
117 Chemical-Speciation-Network, which is established for improving the understanding of the heavy
118 pollution formation mechanism in the NCP and supporting the decision-making of local governments
119 and state administration. Hourly SO₂, NO₂, O₃, PM_{2.5} and its chemical compositions were recorded at
120 the PM_{2.5} component network, which was selected followed the Technical Regulation for Selection of
121 Ambient Air Quality Monitoring Station published by the Ministry of Ecology and Environment of the
122 People's Republic of China (HJ664-2013). The monitoring sites of PM_{2.5} component network were
123 mostly set up within the cities, and can reflect the average pollution level of each city. Details for the
124 near-ground observation stations of PM_{2.5} component network were shown in Table S1. Mass
125 concentrations of SO₄²⁻, NO₃⁻, and NH₄⁺ in PM_{2.5} were continuously measured at a 1-h resolution by
126 MARGA (model ADI 2080) or AIM-IC (URG 9000D) equipped with a PM_{2.5} sampling inlet. These
127 two IC-based online instruments have shown good performance through instrument intercomparison
128 studies or comparison to offline filters under clean to moderately polluted conditions (Markovic et al.,
129 2012; Wu and Wang, 2007; Park et al., 2013; Rumsey et al., 2014). Organic carbon (OC) was measured
130 online by Sunset Semi-Continuous Carbon Analyzer (Sunset Laboratory Inc, USA). The concentration
131 of OM can be obtained by multiplying the OC concentration by a factor of 1.6 (Li et al., 2021). PM_{2.5},
132 O₃, NO₂ and SO₂, were recorded hourly based on Thermo Scientific samplers and analyzers. Detailed
133 descriptions of these online sampling instruments can be found in our previous works (Kong et al.,
134 2018; Liu et al., 2017a; Pang et al., 2020; Wang et al., 2022b). The meteorological variables were
135 recorded in the national meteorological observation stations, and the information of each station can be
136 obtained from the public website of China Meteorological Administration
137 (<http://data.cma.cn/data/cdcindex/cid/0b9164954813c573.html>). The temporal resolution of air
138 temperature, RH, WS and WD was 1-h. To avoid the influence of diurnal boundary layer cycles, in this
139 article we focused on the relationships between daily mean air pollutants and meteorological factors.
140 The daily mean meteorological factors, PM_{2.5} and its major secondary components were calculated

141 from the hourly data; daily O₃ concentration was characterized by the maximum daily 8 h average
142 ozone (MDA8 O₃). Details for the near-ground observation species and the metrics were shown in
143 Table S2.

144 To better demonstrate the overall change characteristics of regional air pollution and
145 meteorological conditions during the observation period, the occurrence frequency (%), which means
146 the proportion of the number of cities at each air pollutant or methodology level, was calculated based
147 on the following equation:

$$148 \text{ Occurrence frequency}_X^{\text{level}} = \frac{N_X^{\text{level}}}{\text{Total } N_X} \times 100 \% \quad (1)$$

149 where X means the air pollutants or methodology factors, N_X^{level} represents the number of cities at each
150 X level, Total N_X represents the total number of cities.

151 2.2 The calculation of mixing layer heights

152 In recent years, many works have progressed in the atmospheric boundary layer characteristics,
153 and analyzed the impacts of these parameter on air pollution. Planetary boundary layer (PBL), as one
154 of the critical parameters to air quality modeling, has been well explored. However, PBL usually refers
155 to the large-scale Ekman dynamic boundary layer (Haugen et al., 1971; Wang et al., 2014; Zhang et al.,
156 2005). The way with which boundary layer describes the influences of air pollution is easily duplicated
157 and confused (Niu et al., 2017). It is unreasonable to some extent, if the characteristic of the air
158 pollution related to near-surface boundary layer is evaluated by using the concept of PBL. For air
159 pollution measurement, one of selected functionalities of parameterization scheme for pollution mixing
160 layer is to judge whether an air mass over a specific locality satisfies the “static and stable” attribute or
161 not. Therefore, in this work, to express the basic physics for diagnosing meteorological conditions, we
162 used the concept of pollution mixing layer height (MLH) proposed by Wang et al. (2017), which was
163 based on the classical synoptic theory according to the level of convective condensation layer, and the
164 details of this method can be seen in previous work (Wang and Yang, 2000; Wang et al., 2017).

165 To be specific, we define the height close to the cloud base as the height of super-saturation layer
166 (H_{SSL}), and the isentropic atmospheric process meets the level of convective condensation layer
167 (LCL) in the super-saturation state, i.e., it is very close to the H_{SSL}. Iterative algorithm is used to
168 work out the H_{SSL} (Wang and Yang, 2000):

169 $H_{SSL} \approx LCL = 6.11 \times 10^2 \times \left(\frac{0.622 + 0.622 \frac{e_s}{p - e_s}}{0.622 \frac{e_s}{p - e_s}} \right),$ (2)

170 $e_s = 6.22 \times \exp \frac{17.13(T - 273.16)}{T - 38},$ (3)

171 where e_s represents saturated water vapor pressure, T is temperature (K). Eq. (2) can be used to
 172 calculate the H_{SSL} which is favorable for pollutant mixing and represented by (P). Below this height,
 173 the atmosphere gets supersaturated, causing the pollution mixing and wetting process in the low
 174 altitude to continue, so this height is called the height of pollution mixing layer (MLH). Thus, MLH
 175 can be derived in the following expression:

176 $MLH \approx H_{SSL} \approx LCL = 6.11 \times 10^2 \times \left(\frac{0.622 + 0.622 \frac{e_s}{p - e_s}}{0.622 \frac{e_s}{p - e_s}} \right),$ (4)

177 According to the relationship between air pressure and height, the units of MLH can be converted to
 178 the height expression in meters:

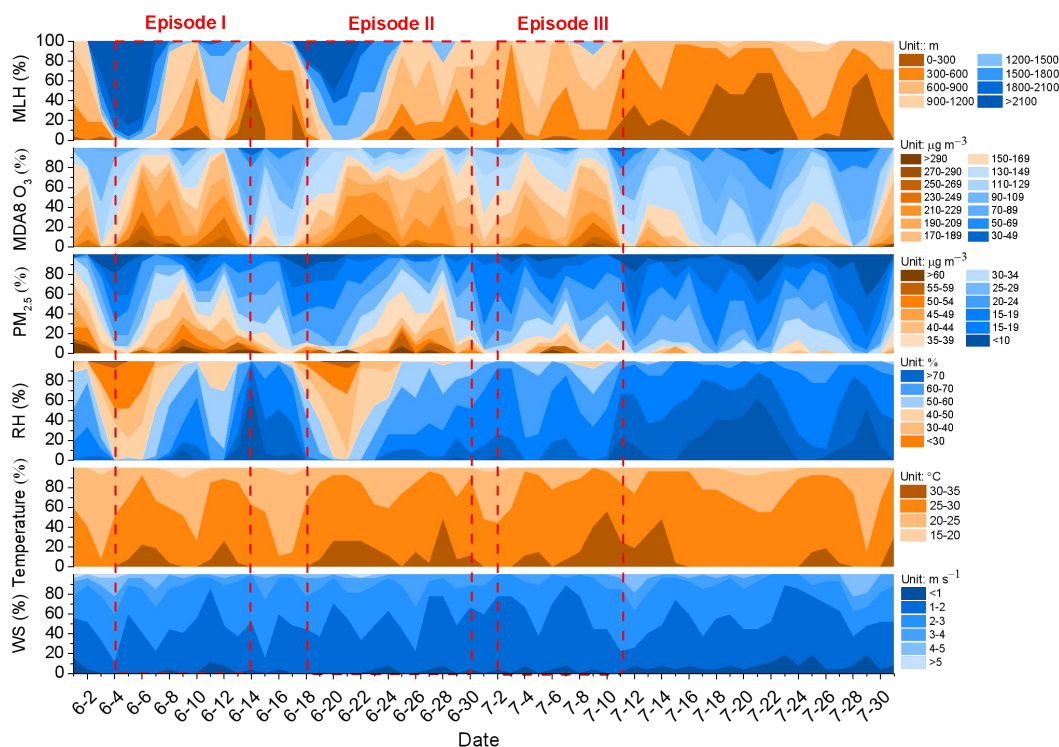
179 $\int_{p_0}^{p_z} dp = - \int_0^z \rho_0 g dz,$ (5)

180 where z is the height, ρ_0 is the density of gas, p_z and p_0 represent the air pressure in the height of z and
 181 0, respectively.

182 Several works have verified the reliability of the results based on this method. With this method,
 183 Wang et al. (2017) well characterized the features of mixing layer height in highly-sensitive areas of
 184 pollution in China. Wang et al. (2022c) also used this method to explore the $PM_{2.5}$ and O_3
 185 superposition-composite pollution event during spring 2020 in Beijing, China, and the hourly evolution
 186 of MLH, O_3 , and $PM_{2.5}$ during the observation period were analyzed. In addition, Niu et al. (2017) has
 187 applied this method in Beijing, and the results showed that the pollution mixing layer can well present
 188 the change characteristics of haze pollution process. In this work, we further clarified the concept of
 189 MLH, and applied this method to investigate the impacts of MLH upon the change characteristics of
 190 ozone and fine particulate matter.

191 **3 Results and discussions**

192 **3.1 General characteristics**



193

194 **Figure 2.** The occurrence frequency (%) of PM_{2.5}, MAD8 O₃, and meteorological factors under
 195 different levels in the NCP from June 1 to July 31, 2021. The color shading represents different
 196 categories classified by PM_{2.5}, MDA8 O₃, and meteorological factors. The red dash boxes represent
 197 three typical PM_{2.5} and O₃ co-polluted episodes: June 4–14 (Episode I), June 18–29 (Episode II), and
 198 July 2–11 (Episode III), 2021.

199 The summertime change characteristics of ground-level meteorological factors (MLH, RH,
 200 temperature, and WS), MDA8 O₃, PM_{2.5} and its major components in the NCP were demonstrated in
 201 Figure 2 and Figure S1. The primary atmospheric pollutant in the NCP during the summertime was O₃,
 202 and the concentrations of MDA8 O₃ averaged over all sites in the NCP varied from 74.94 µg m⁻³ to
 203 219.28 µg m⁻³, with the mean value of 151.72 µg m⁻³ (Table 1). The O₃ pollution lasted nearly the
 204 whole observation period, characterized by frequent and long-lasting pollution episodes. The PM_{2.5}
 205 concentration was much lower comparing with ozone, with the mean, maximum, and minimum of the
 206 regional daily mean PM_{2.5} concentration as 25.62 µg m⁻³, 45.62 µg m⁻³, and 11.32 µg m⁻³, respectively,
 207 during the observation period. NO₃⁻ was the prominent PM_{2.5} component, with the mean concentration
 208 of 7.76 µg m⁻³. According to the National Ambient Air Quality Standard of China (GB3095-2012), the
 209 daily PM_{2.5} averages in “2+26” cities can meet the Level II standard of 75 µg m⁻³, while exceeding the
 210 level I standard (35 µg m⁻³). As showed in Figure 2, regional PM_{2.5} pollution processes corresponded
 211 well with the increasing processes of MDA8 O₃. Here, we define a O₃–PM_{2.5} co-polluted episode as a

212 set of continuous days (longer than 4 days) with MDA8 O₃ and daily mean PM_{2.5} (in more than 10 %
 213 NCP cities) exceeding 160 µg m⁻³ and 35 µg m⁻³, respectively. According to this criterion, three typical
 214 O₃-PM_{2.5} co-polluted episodes were selected: June 4–14 (Episode I), June 18–29 (Episode II), and July
 215 2–11 (Episode III), 2021.

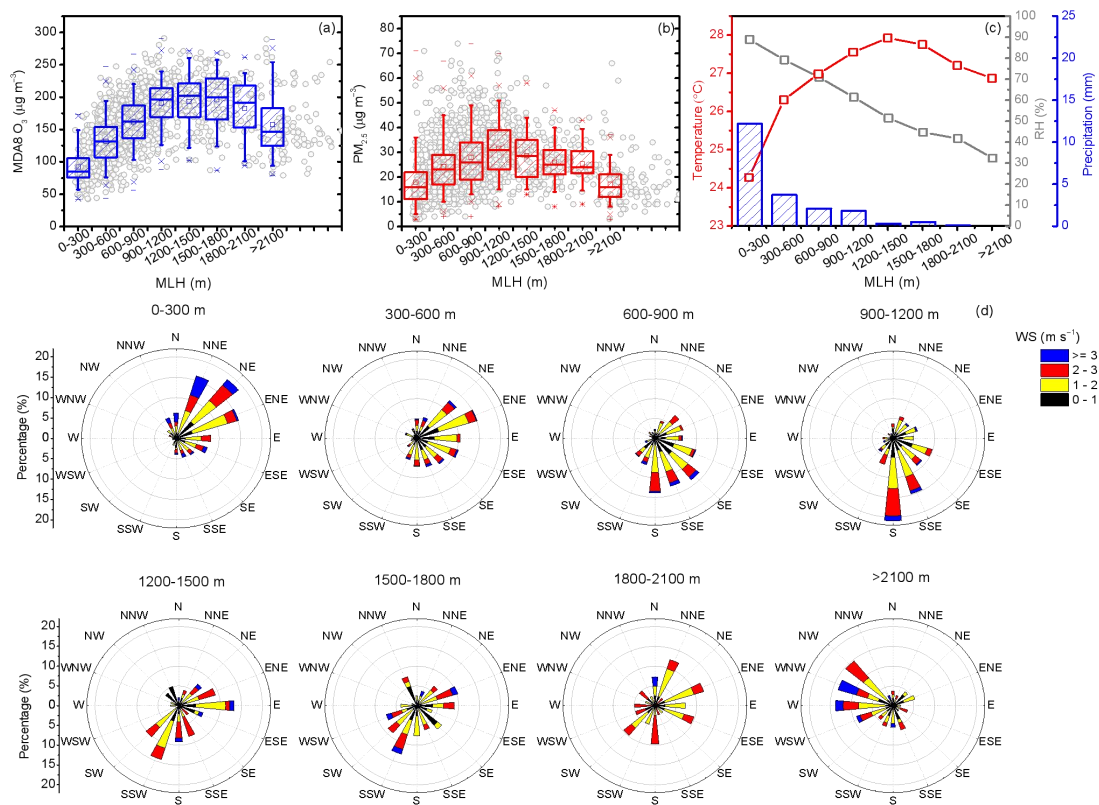
216 During these three typical episodes, the synchronous change characteristics of air pollutants and
 217 mixing layer meteorology were analyzed. In Episode I and II, when MLH higher than 2100 m, both
 218 MDA8 O₃ and PM_{2.5} concentrations were low. Along with the reduction of MLH (from 1800–2100 m
 219 to 1200–1800 m), regional MDA8 O₃ and PM_{2.5} concentration both gradually climbed up. When MLH
 220 fell in the range of 1200–1800 m, MDA8 O₃ concentration reached the maximum with about 80 %
 221 areas higher than 170 µg m⁻³. We found that there is a lag time between the concentration peak of
 222 MDA8 O₃ and that of PM_{2.5} along with the reduction of MLH. With the further decrease of MLH,
 223 MDA8 O₃ turned to decline, while PM_{2.5} kept stable or continued to increase when regional MLH in
 224 the range of 600–1200 m. In Episode III, the MLH in most cities was lower than 1200 m, and the
 225 regional MDA8 O₃ and PM_{2.5} pollution conditions were lighter than other episodes, with 80 % PM_{2.5}
 226 values lower than 35 µg m⁻³. It's interesting to note that the change characteristics of SO₄²⁻ and NO₃⁻
 227 were different (Figure S1), and the regional peaks of these two components were inconsistent,
 228 especially in Episode II. With the evolution of MLH, NO₃⁻ climbed up and peaked on June 24 when
 229 regional MLH lower than 900 m, while SO₄²⁻ reached the maximum on June 28 when MLH was around
 230 900–1500 m. This may be related to other synchronized mixing layer meteorology factors, such as RH
 231 and temperature. For example, the evolution of mixing layer often accompanied with changes in
 232 temperature. The increase in temperature can promote the chemical formation rate of these secondary
 233 components, but also stimulate the volatilization of NO₃⁻ to gaseous state (HNO₃) and lead to the
 234 decrease in NO₃⁻ concentration. Further analysis about the response of O₃, PM_{2.5} and its secondary
 235 components to different mixing layer meteorology factors will be conducted in the following sections.

236 Table 1. General information on O₃-PM_{2.5} co-polluted episodes from June 1 to July 31, 2021.

	Episode I			Episode II			Episode III			Summer		
	Ave.	Min	Max	Ave.	Min	Max	Ave.	Min	Max	Ave.	Min	Max
Gaseous pollutants (µg m ⁻³)												
MDA8												
O ₃	170.80	85.62	219.28	180.65	142.10	204.15	168.70	111.79	199.39	151.72	74.94	219.28
SO ₂	10.01	6.48	14.44	9.09	6.11	12.48	6.75	5.72	8.00	7.59	4.79	14.44

NO ₂	24.61	16.26	31.81	22.89	14.11	32.15	17.66	13.12	21.00	19.31	10.90	32.15
PM _{2.5} and its major components (µg m ⁻³)												
PM _{2.5}	30.55	15.74	42.67	28.33	17.22	42.52	25.05	20.84	31.75	25.62	11.32	45.62
NO ₃ ⁻	8.74	2.16	16.44	8.29	2.85	18.00	7.67	5.87	13.44	7.76	2.16	18.24
SO ₄ ²⁻	7.22	2.81	10.25	7.32	4.02	12.15	7.12	5.48	8.92	7.04	2.81	12.15
NH ₄ ⁺	5.51	1.42	9.34	5.52	2.27	9.29	5.38	4.46	8.21	5.30	1.42	9.88
OC	5.11	2.74	6.60	4.71	3.25	6.75	4.11	2.90	5.30	4.32	2.69	6.75
Meteorological variables												
MLH (m)	1342.73	305.93	2423.42	1190.36	626.51	2127.31	740.86	460.91	950.10	855.99	305.93	2423.42
T (°C)	26.24	23.86	28.91	27.41	25.53	28.76	27.58	24.85	30.14	26.69	22.48	30.14
RH (%)	57.01	32.78	90.54	56.90	37.04	70.60	71.45	64.64	80.38	68.70	32.78	90.54

237 3.2 Evolution of ozone with mixing layer meteorology



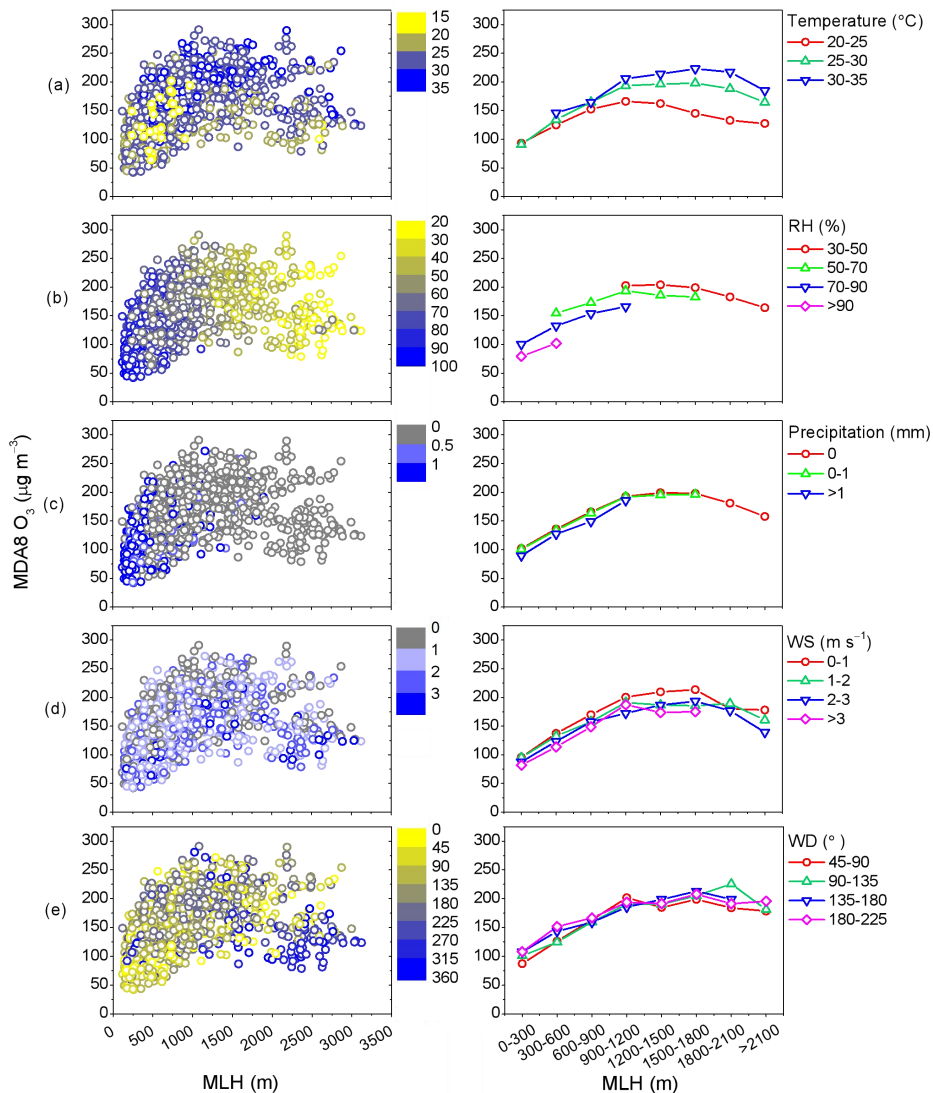
238 **Figure 3.** The variation characteristics of (a) MDA8 O₃, (b) PM_{2.5}, (c) temperature, RH, 24-h
 239 precipitation, and (d) WS and WD in different MLH conditions. Box plots in (a) and (b) show the inter
 240 quartile range (the distance between the bottom and the top of the box), median (the band inside the
 241 box), and 95 % confidence interval (whiskers above and below the box) of the data. S: south; N:
 242 E: east; W: west.

243 To quantify the effect of MLH on near-ground O₃ concentrations, relationships between MLH and

244 MDA8 O₃ were analyzed (Figure 3a). Here we used a data binning method to remove the expected
245 day-to-day atmospheric variability from sampling uncertainty (Dian et al., 2010), which has been
246 applied elsewhere (Lou et al., 2019). The MLH was grouped into 8 classes with 300 m width: 0–300,
247 300–600, 600–900, 900–1200, 1200–1500, 1500–1800, 1800–2100 and > 2100 m. It was found that
248 MDA8 O₃ concentration dramatically increased when MLH in the range of 0–900 m, and leveled off
249 when MLH at around 900–1800 m, with the maximum MDA8 O₃ of 195.88±42.76 μg m⁻³, then turned
250 to decrease with further development of MLH. This nonlinear relationship between MDA8 O₃ and
251 MLH was is consistent with the results conducted by Zhao et al. (2019) and Reddy et al. (2012). The
252 work by Zhao et al. (2019) found that O₃ concentration was the highest at medium boundary layer
253 heights (1200–1500 m) in Shijiazhuang, China. In India, days of higher O₃ concentrations were also
254 associated with higher boundary layer height (Reddy et al., 2012).

255 This relationship observed between MDA8 O₃ and MLH is very complex. Previous works have
256 shown that higher height of mixing layer can lead to the mixing of near surface air with the O₃ rich air
257 aloft, resulting in the observed enhancements in surface O₃ concentration (Reddy et al., 2012).
258 Concurrently, the evolution of mixing layer were strongly associated with the change of other
259 meteorological conditions, such as air temperature, RH and precipitation, which can also affect O₃
260 concentration (Haman et al., 2014). The combined effects of these processes ultimately determine
261 whether ground-level O₃ increases or not along the evolution of mixing layer. The increase of MLH
262 often coincided with higher air temperature, lower RH, and less precipitation (Figure 3c), which were
263 more conducive to O₃ production (Ma et al., 2021; Xu et al., 2018). As shown in Figure 4a–c, as MLH
264 kept constant, MDA8 O₃ concentration climbed up with increase in temperature but decrease in RH
265 and precipitation levels. Possible reasons for these results could be: (1) the increase in RH can
266 contribute to the depletion of O₃, and lead to weakened O₃ related photochemical reaction (Ma et al.,
267 2021; Yu, 2019); (2) due to higher RH or rain fall, gaseous precursors and O₃ can be washed out from
268 the atmosphere through wet deposition (Reddy et al., 2012); and (3) the rise of temperature can
269 accelerate the emission rate of gaseous precursors, such as biogenic VOCs and soil NO_x (Dang et al.,
270 2021; Porter and Heald, 2019), and also stimulate the photochemical reaction rate in the generation of
271 O₃ (Ma et al., 2021). Besides, wind fields could alter surface O₃ concentrations by transporting O₃ or its

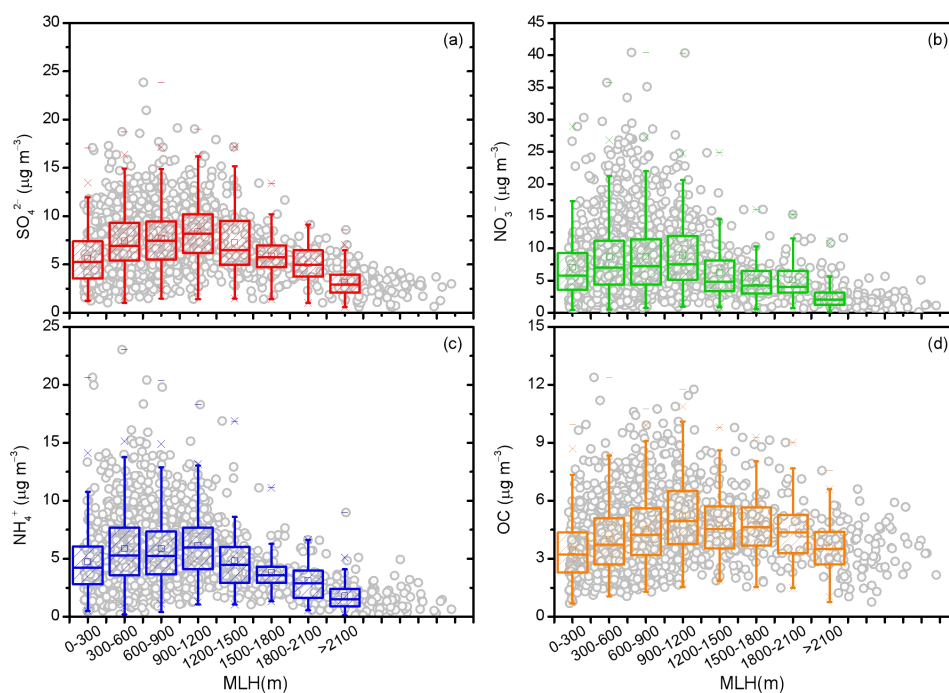
272 precursors in and out of this region (Ma et al., 2021). As shown in Figure S2, during the whole
 273 campaign, the NCP was dominated by winds from northeast and south (45–225°). Because more than
 274 75 % WD were in the rang of 45–225°, the WD was classified into 4 categories: 45–90, 90–135,
 275 135–180, and 180–225°. As shown in Figure 3d, along with the evolution of mixing layer, the WD
 276 gradually changed from northeast (MLH=0–600 m) to southeast (MLH=600–900 m) and south
 277 (MLH=900–1200 m). The southerly wind can transport the gaseous pollutants or O₃ from the southern
 278 part of the plain area to the northern part, and the Taihang mountains may block pollutant transport,
 279 leading to the accumulation of pollutants along the foot of the Taihang Mountains. It's noted that the
 280 concentration of MDA8 O₃ was higher when the plain dominated by southerlies (180–225°) when
 281 MLH lower than 1200 m (Figure 4e). In general, WS could affect the diffusion of air pollutants. Due to
 282 the limited dilution and dispersion effect of weak wind, the MDA8 O₃ concentrations at low wind speed
 283 (0–1 m s⁻¹) were relative higher than other WS conditions (Figure 4d).



284 **Figure 4.** The distribution characteristics of the MDA8 O₃ concentrations with the evolution of MLH
 285 under different (a) temperature, (b) RH, (c) precipitation, (d) WS, and (e) WD conditions.

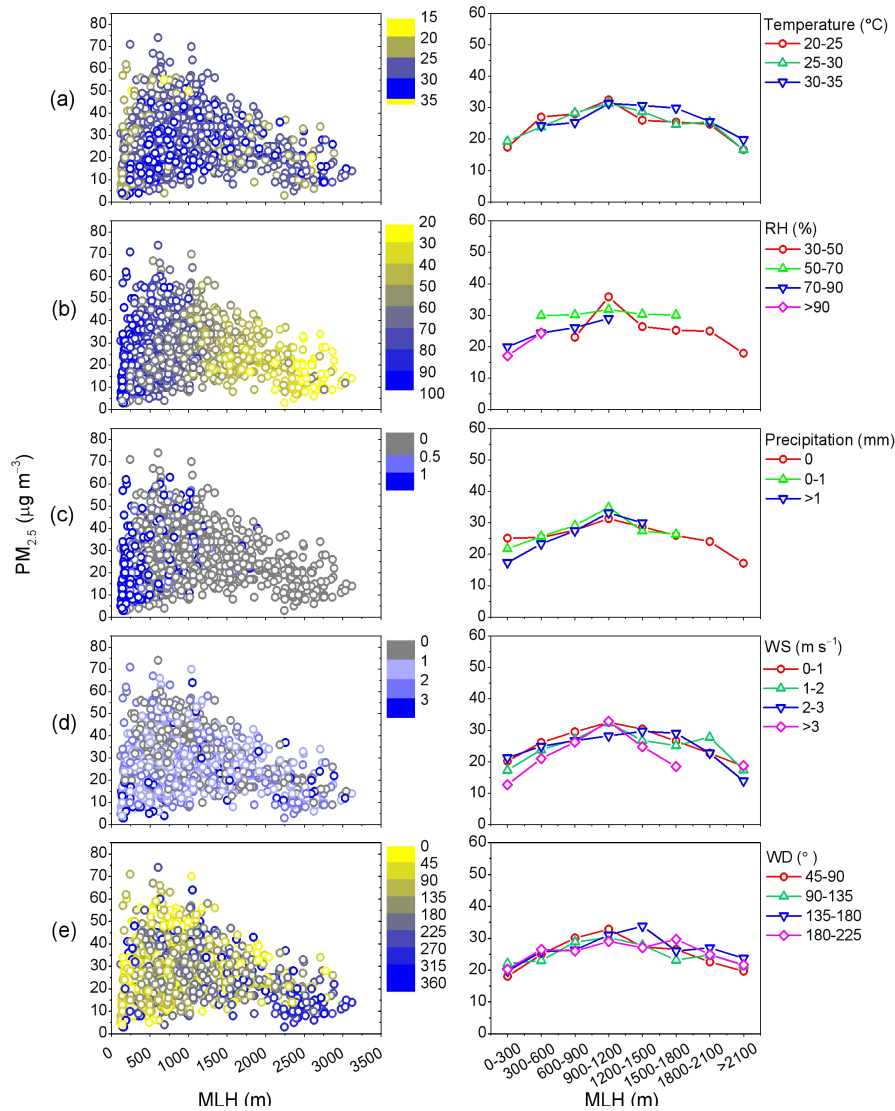
286 3.3 Evolution of PM_{2.5} and its secondary compositions with mixing layer meteorology

287 The concentration distribution of surface PM_{2.5} in different MLH bins has been shown in Figure
 288 3b. Interestingly, PM_{2.5} showed similar change profile as MDA8 O₃, which initially increased and then
 289 declined along with the growth of MLH. PM_{2.5} concentration reached the maximum of 31.65 μg m⁻³
 290 when MLH fell in the range of 900–1200 m, and the concentration has increased by 1.51 μg m⁻³
 291 through the rise phase for the variation of 100 m MLH. This phenomenon was quite different with the
 292 results in cold seasons (Pan et al., 2019; Du et al., 2013; Murthy et al., 2020). It has been suggested that
 293 the narrowing of mixing layer will compress air pollutants into a shallow layer, resulting in elevated
 294 pollution levels, thus MLH has been illustrated as the key factor to aggravate the haze events in large
 295 cities of China in winter. However, the response of PM_{2.5} concentration to MLH is not only determined
 296 by the vertical stratification of the mixing layer, but also by local sources, secondary chemical
 297 formation, wet deposition, and the wind field (Lu et al., 2019; Geiß et al., 2017; Pan et al., 2019; Miao
 298 et al., 2021; Lou et al., 2019). Noted that in this work there were still some extreme high PM_{2.5} values
 299 under low MLH condition as shown in Figure 3b, and this phenomenon will be discussed in the
 300 following part when exploring the effect of precipitation.



301
 302 Figure 5. The variation characteristics of (a) SO₄²⁻, (b) NO₃⁻, (c) NH₄⁺, and (d) OC in different MLH

303 conditions. Box plots show the inter quartile range (the distance between the bottom and the top of the
 304 box), median (the band inside the box), and 95 % confidence interval (whiskers above and below the
 305 box) of the data.



306 **Figure 6.** The distribution characteristics of the $PM_{2.5}$ concentrations with the evolution of MLH under
 307 different (a) temperature, (b) RH, (c) precipitation, (d) WS, and (e) WD conditions.

308 The response of $PM_{2.5}$ concentrations to mixing layer structure was the net effect of the changes in
 309 $PM_{2.5}$ major chemical components, such as SO_4^{2-} , NO_3^- , NH_4^+ , and OC. Figure 5 showed the changes
 310 in the major $PM_{2.5}$ components due to the evolution of mixing layer. All the secondary components
 311 showed increasing trends when MLH lower than 1200 m, with SO_4^{2-} and OC showing the highest
 312 increment, followed by NO_3^- and NH_4^+ . When MLH changed from 300–600 m to 900–1200 m, the
 313 increment was not significant for NO_3^- and NH_4^+ . As NH_3 was generally abundantly supplied in the
 314 NCP, the formation of NH_4^+ was dominantly controlled by the reaction of ammonia with sulfate and

315 nitrate aerosols, and the changes in NH_4^+ were a consequence of the changes in SO_4^{2-} and NO_3^- (Chow
316 et al., 2022). When $\text{MLH} < 1200$ m, the mass fraction of NO_3^- was higher than SO_4^{2-} in $\text{PM}_{2.5}$ (Figure
317 S3), and the change characteristics of NH_4^+ along with the evolution of mixing layer were consistent
318 with that of NO_3^- . The mass ratio of SO_4^{2-} to NO_3^- gradually increased along with the development of
319 mixing layer. When MLH higher than 1200m, SO_4^{2-} surpassed NO_3^- and became the dominant $\text{PM}_{2.5}$
320 component. The difference in the relationships between these aerosol species and MLH reflected the
321 intrinsic complexity mechanisms of $\text{PM}_{2.5}$ formation, which were probably related to other
322 meteorological parameters, such as temperature, RH, precipitation, WS, and WD. In order to
323 understand how the other meteorological factors impacted the relationship between MLH and $\text{PM}_{2.5}$,
324 we demonstrated the statistics on the concentration distribution of $\text{PM}_{2.5}$ and its dominant components
325 with the increase of MLH under different RH, temperature, precipitation, WS, and WD conditions in
326 Figure 6 and Figure 7.

327 Temperature is not only essential to the secondary chemical reaction of trace gases, but also the
328 gas-particle partitioning of volatile $\text{PM}_{2.5}$ species. The response of $\text{PM}_{2.5}$ and its dominant components
329 to MLH followed similar change characteristics under different temperature conditions, all increasing
330 with the development of mixing layer when MLH lower than 1200m. The response of $\text{PM}_{2.5}$ to
331 temperature was largely the result of opposite changes in NO_3^- and SO_4^{2-} concentrations with a smaller
332 role played by organics (Figure 7). Specifically, as MLH kept constant, SO_4^{2-} concentration climbed up
333 with increasing temperature level, while the concentration of NO_3^- declined when temperature kept
334 going up. Higher temperature may promote faster oxidation of SO_2 to SO_4^{2-} , resulting in a significant
335 increase in SO_4^{2-} concentrations. Unlike SO_4^{2-} , which predominantly exists in the particle phase, NO_3^-
336 could be either presented as nitric acid (HNO_3) in the gas phase or as ammonium nitrate (NH_4NO_3) in
337 the particle phase (Chow et al., 2022). The temperature condition strongly influences the partitioning of
338 nitrate between gas and particle phase. Higher temperature prompts the partitioning of nitrate to HNO_3 ,
339 thus nitrate tends to exit in the gas phase, resulting in a significant decrease in NO_3^- and NH_4^+
340 concentrations.

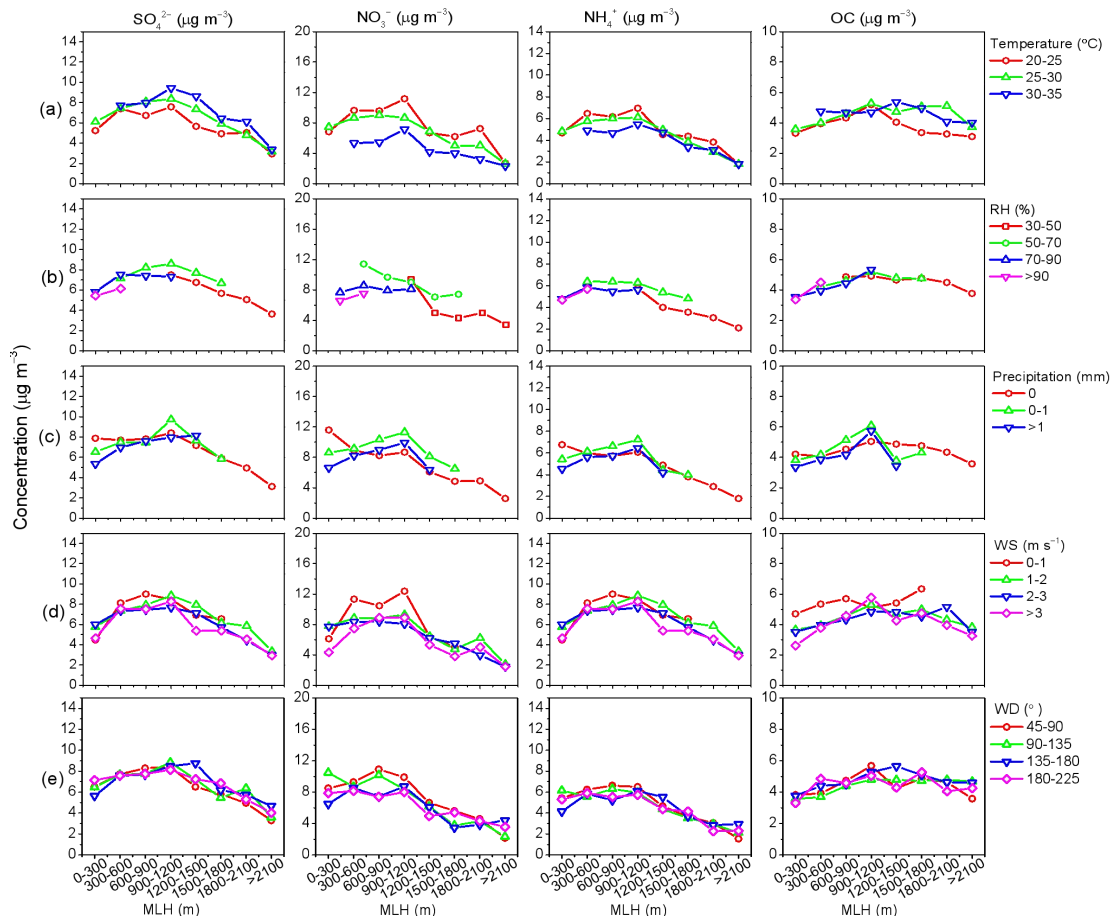
341 The response of $\text{PM}_{2.5}$ and its dominant components to the evolution of mixing layer was more
342 sensitive to RH, and distinct distribution characteristics under different RH ranges have been observed
343 in Figure 6 (b) and Figure 7 (b). When MLH fell in the range of 300–900 m, the concentration of $\text{PM}_{2.5}$
344 (Figure 6b) and its major components (Figure 7b) mostly decreased with RH elevating from 50–70 %

345 to 70–90 %. Previous works have shown that when RH higher than 60%, local humidity-related
346 physicochemical processes play important roles in transforming the gases to aerosols (Wang et al.,
347 2022d; Liu et al., 2020). We considered that the RH range from 50% to 70% was more beneficial to the
348 aqueous chemical production of major PM_{2.5} components, then led to the increase of PM_{2.5}
349 concentration. It is worth noting that when MLH in the range of 0–300 m, with RH increasing from
350 70–90 % to > 90 %, the concentration of PM_{2.5} (Figure 6b) and its major components (Figure 7b)
351 severely dropped, which was probably related to the fast hygroscopic growth and enhanced wet
352 deposition processes.

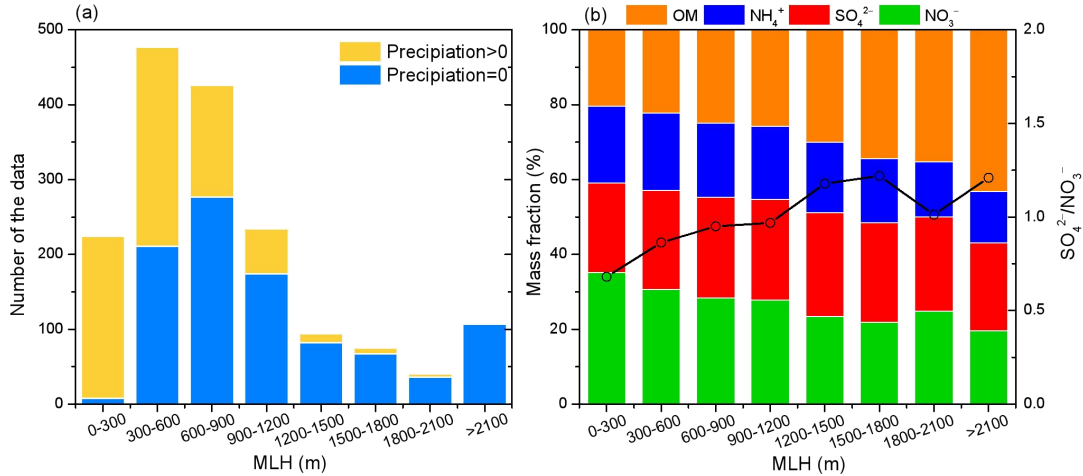
353 All aerosol species have wet deposition as a major sink, so precipitation is expected to have
354 significant effects on PM_{2.5} concentrations. As shown in Figure 6 (c), changes in the concentrations of
355 PM_{2.5} was sensitive to the rain events. When MLH fell in the range of 0–300 m, the concentration of
356 PM_{2.5} significantly decreased during rainfall period. Interestingly, when no rainfall occurred, even
357 though PM_{2.5} concentration kept stable under low MLH condition, the response of PM_{2.5} concentrations
358 to MLH still followed upward trend with MLH increasing from 300–600 to 900–1200 m. As for
359 specific aerosol species (Figure 7c), NO₃⁻ and NH₄⁺ concentration showed two obvious peaks, with one
360 in the range of 0–300 m, and the other in 900–1200 m. Under low MLH condition, the concentrations
361 of NO₃⁻ and NH₄⁺ were high, with NO₃⁻ as the dominant species in PM_{2.5} (Figure 8b). With the growth
362 of MLH, NO₃⁻ and NH₄⁺ initially decreased, but turned to increase again when MLH in the range of
363 900–1200 m. As for SO₄²⁻ and OC, the concentrations obviously increased with the elevation of MLH
364 and has exceeded that of NO₃⁻ when MLH higher than 1200 m. As shown in Figure 8 (a), low mixing
365 layer generally accompanied with cloudy and rainy conditions during summertime in the NCP in 2021,
366 and only small fraction of days without rainfall has been captured during this period. Therefore, despite
367 some high PM_{2.5} or major aerosol species values have been witnessed under low MLH condition, the
368 overall trend in Figure 3 (b) was still upwards along with the growth of mixing layer (MLH < 1200 m).
369 The increase of PM_{2.5} and its major chemical components under medium MLH condition was not only
370 associated with the weaker particle removal process by precipitation, but also related to the
371 enhancement of secondary aerosol formation due to appropriate chemical reaction environment.

372 WS can represent the atmospheric dissipation potential in the horizontal directions (Zhu et al.,
373 2018). Low WS generally suggested weak pressure gradients and potentially a more favorable
374 meteorological condition for PM_{2.5} enhancement (Ma et al., 2021). As expected, the concentrations of

375 PM_{2.5} (Figure 6d) and its aerosol species (Figure 7d) gradually decreased with the increase of WS. The
 376 response of these air pollutants to MLH followed similar upward trends under different WS conditions
 377 (MLH < 1200 m). Comparing with O₃, the impact of WD along with the increase of MLH seems
 378 different for PM_{2.5} and its dominant components. When MLH in the range of 600–1200 m, the NCP
 379 was dominated by southeast or south wind (Figure 3d). However, when southeast or south wind
 380 prevailed, the corresponding PM_{2.5} and its dominant components concentrations were comparable or
 381 even lower than other WD situations (Figure 6e and Figure 7e). This indicated that regional transport
 382 was not the dominant factor leading to the elevation of PM_{2.5} and its aerosol species along with the
 383 evolution of mixing layer (MLH < 1200 m).



384 **Figure 7.** The distribution characteristics of NO₃⁻, SO₄²⁻, NH₄⁺, and OC concentrations with the
 385 evolution of MLH under different (a) temperature, (b) RH, (c) precipitation, (d) WS, and (e) WD
 386 conditions.



387

388 **Figure 8.** (a) The number distributions of the data when the daily precipitation larger than 0 mm or
 389 equal to 0 mm along with the evolution of MLH. (b) The mass fractions of major PM_{2.5} components
 390 and the mass ratio of SO₄²⁻ to NO₃⁻ along with the evolution of MLH when the daily precipitation
 391 equal to 0 mm.

392 3.4 Superposition-composite effects of PM_{2.5} and O₃ with the evolution of mixing layer

393 3.4.1 A case study of the typical PM_{2.5}-O₃ co-polluted episode

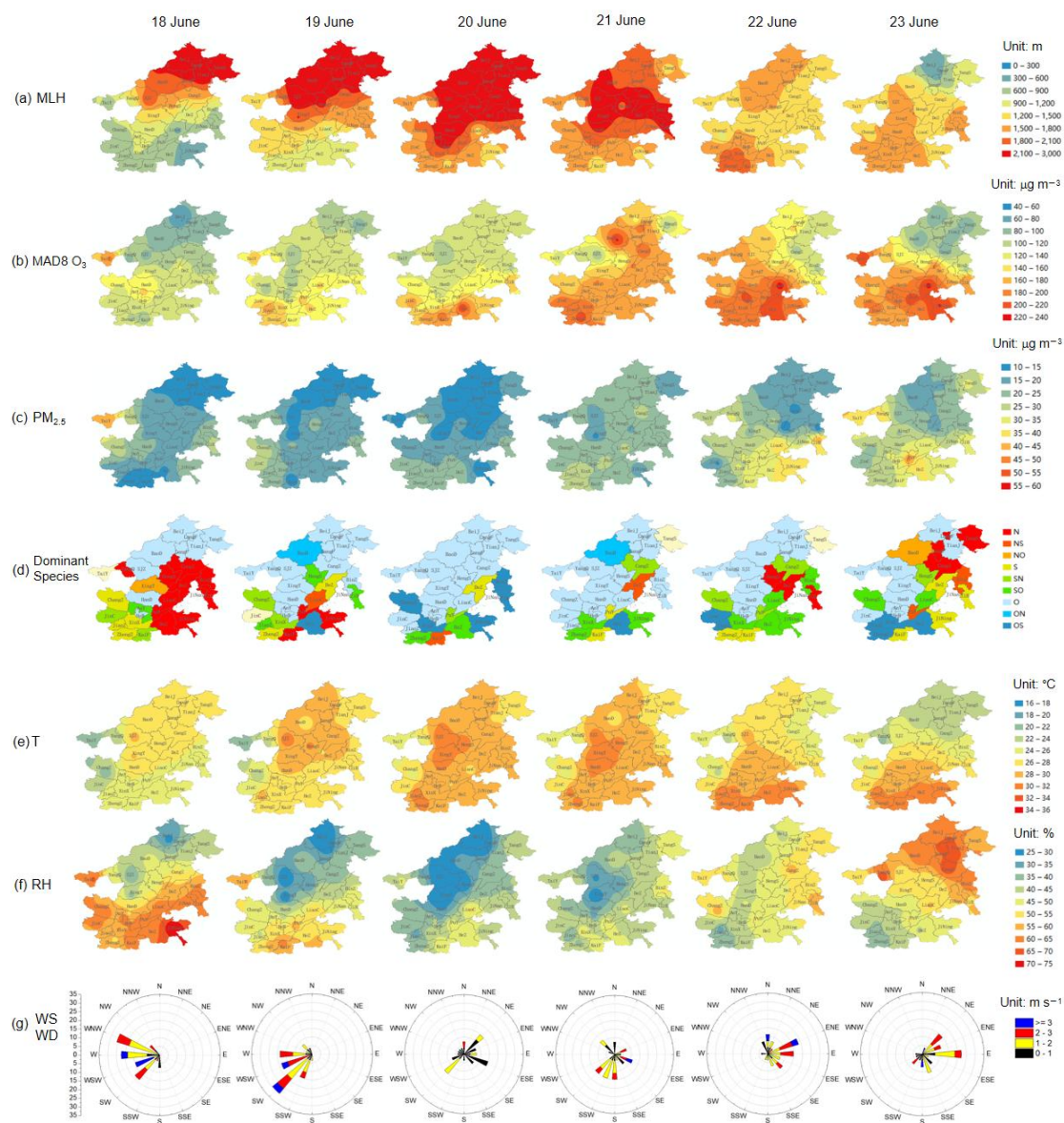
394 Previous results indicated that MDA8 O₃ and PM_{2.5} concentrations were closely related to the
 395 evolution of BLH. The increasing trend of PM_{2.5} concentration with the development of mixing layer
 396 under medium MLH condition discussed before indicated that the evolution of mixing layer was not a
 397 simple physical dilution process, and its influence on the enhanced secondary photochemical formation
 398 should be considered as well. Figure 9 and 10 demonstrated a typical PM_{2.5}-O₃ co-polluted episode
 399 (Episode II) during 18–29, 2021 to comprehensively present the relationship between the mixing layer
 400 and pollutants. On June 18–20, MLH gradually increased from 600–1200 m to 1500–3000 m in the
 401 southern and eastern part of the NCP, PM_{2.5} and MDA8 O₃ concentrations concurrently increased and
 402 showed similar spatial distributions. The wind speed dropped significantly on 20 June, and the value
 403 was lower than 1 m s⁻¹ in most cities. On 21–23 June, MLH started to decrease from 1500–3000 m to
 404 1200–1800 m, PM_{2.5} and MDA8 O₃ concentrations further increased, and the areas of high PM_{2.5}
 405 concentrations also coincided well with those of MDA8 O₃ concentrations. During 24–25 June, MLH
 406 continued to decrease, with some values even lower than 300 m. The MLH for the areas with high
 407 MDA8 O₃ was in the range of 900–1500 m. Interestingly, the synchronized spatial change
 408 characteristics of PM_{2.5} and MDA8 O₃ were consistent when MLH in the range of 900–1200 m, while

409 inconsistent when MLH lower than 600 m. Significant rise of PM_{2.5} concentration was observed in
410 some cities with MLH lower than 300 m. It's noted that the dominant chemical composition of PM_{2.5} in
411 these areas was NO₃⁻. On 28 June, the rise in MLH was observed in the central and the southern part in
412 the NCP, and a surge of MDA8 O₃ and PM_{2.5} concentrations both occurred, with 160–220 μg m⁻³ and
413 40–50 μg m⁻³ respectively. In general, most cities were dominated by weak winds from the east and
414 southeast, which favored the formation of secondary pollutants from the gaseous precursors transported
415 from the southeast part and promoted the accumulation of air pollutants.

416 To better understand this PM_{2.5}-O₃ co-polluted event, here we classified the observations during
417 this typical event into four categories: O₃ polluted days (O₃PD; MDA8 O₃ concentration > 160 μg m⁻³
418 and PM_{2.5} < 35 μg m⁻³), PM_{2.5} polluted days (PM_{2.5}PD; MDA8 O₃ concentration < 160 μg m⁻³ and
419 PM_{2.5} > 35 μg m⁻³), O₃-PM_{2.5} co-pollution days (O₃-PM_{2.5}CPD; MDA8 O₃ concentration > 160 μg m⁻³
420 and PM_{2.5} > 35 μg m⁻³), and non-polluted days (NPD; MDA8 O₃ < 80 μg m⁻³ and PM_{2.5} < 35 μg m⁻³).
421 Figure 11 showed the meteorological and chemical characteristic of O₃-PM_{2.5}CPD, O₃PD, PM_{2.5}PD,
422 and NPD. The results indicated that the values of MLH on O₃-PM_{2.5}CPD were between those on O₃PD
423 and PM_{2.5}PD at around 900 m. On O₃-PM_{2.5}CPD, the oxidation ratio of sulfate (SOR, the molar ratio
424 of sulfate to the sum of sulfate and SO₂) and oxidation ratio of nitrate (NOR, the molar ratio of nitrate
425 to the sum of nitrate and NO₂) were the highest, with the values of 0.44 and 0.33, respectively, which
426 indicated the strong secondary formation of SO₄²⁻ and NO₃⁻ promoted by high O₃ concentration. The
427 PM_{2.5}PD occurred when MLH lower than 650 m, and the percentage of NO₃⁻ was the highest on
428 PM_{2.5}PD. The rise of PM_{2.5} in some cities under low MLH conditions may be attributed to three
429 mechanisms. The first one is the accumulation effect due to unfavorable diffusion condition when
430 MLH decreased. Second, these cities got little rain, and the effect of wet deposition was weak. In
431 addition, the corresponding low T and high RH can stimulate the formation of NO₃⁻ from gaseous state
432 (HNO₃). On O₃PD, the MLH was around 1300 m, and the NOR turned to decrease, demonstrating a
433 more significant role of partitioning process between gas and aerosol than the atmospheric oxidation
434 process under this stage. On NPD, the MLH was the highest, with the value of about 2400 m, and the
435 PM_{2.5} chemical composition was obviously dominated by OM.

436 To explore the relevance of hourly O₃, PM_{2.5}, its components and MLH, we have taken PuY and
437 HeZ as examples. Figure S4 plotted the day-to-day variations along with the diurnal variations of O₃,
438 PM_{2.5}, its components and MLH in PuY and HeZ during Episode II (June 18–29, 2021). The results

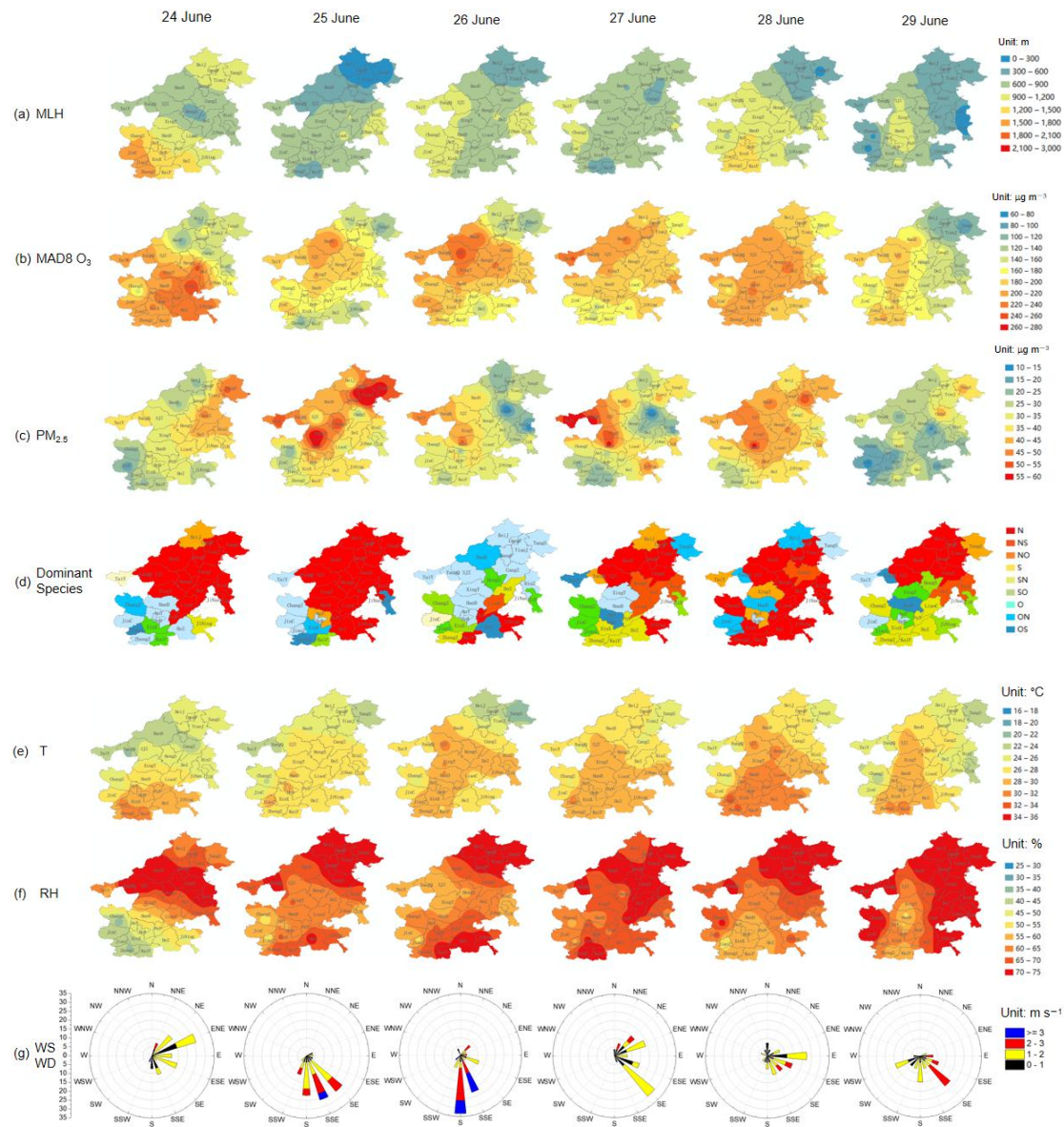
439 showed that there were large diurnal as well as day-to-day variability in the O₃ and PM_{2.5} levels. The
440 diurnal variations of MLH were clearly visible (Figure S5), with the rise in MLH during the daytime
441 and the decrease in MLH at night. The concentration of PM_{2.5} increased with the decrease of MLH at
442 night, but the concentration of O₃ increased with the rise of MLH at daytime. Interestingly, we observed
443 noontime soar of SO₄²⁻ and OC concentrations in PuY, and the values of SOR kept stable or even
444 increase at noon. Besides, it's noted that O₃ and PM_{2.5} both gradually accumulated with the
445 development of mixing layer during June 18–21 and 26–28, which can be attributed to the O₃ and PM_{2.5}
446 superposition composite effects. The decrease in PM_{2.5} at daytime with the rise of MLH can be offset
447 partly by an increment in secondary pollutants formation derived from O₃ growth. Then with the
448 decrease of MLH at night, the concentration of the original existing PM_{2.5} increased due to unfavorable
449 diffusion. In general, the conclusions in this work was only suitable to the day-to-day relationship
450 between air pollutants and MLH. The hourly relationships were much more complicated and need more
451 further analysis.
452



453

454

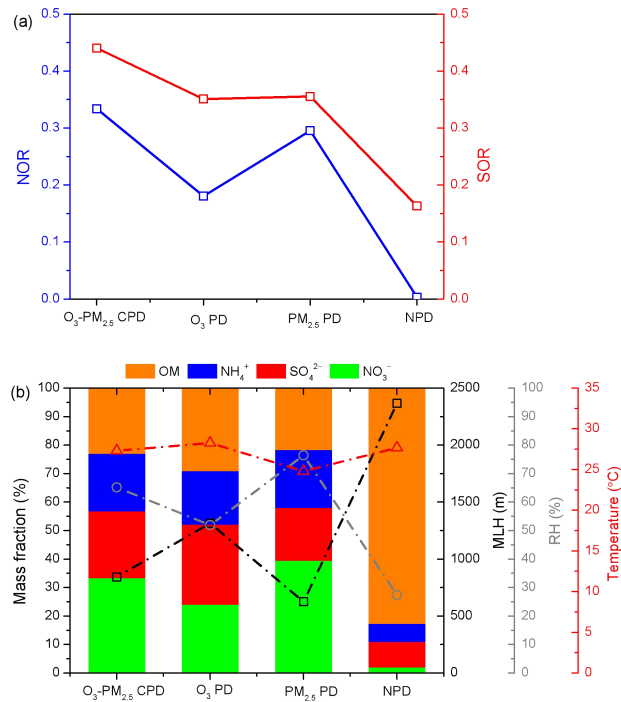
455 **Figure 9.** The spatial distribution of (a) MLH, (b) MDA8 O₃, (c) PM_{2.5}, (d) the dominant PM_{2.5}
 456 chemical component (N: NO₃⁻ dominant, NS: NO₃⁻ and SO₄²⁻ dominant, NO: NO₃⁻ and OM dominant,
 457 S: SO₄²⁻ dominant, SN: SO₄²⁻ and NO₃⁻ dominant, SO: SO₄²⁻ and OM dominant, O: OM dominant, ON:
 458 OM and NO₃⁻ dominant, OS: OM and SO₄²⁻ dominant), (e) T, and (f) RH, (g) the overall change
 459 characteristics of WS and WD in the NCP from June 18 to 23, 2021. The dominant PM_{2.5} chemical
 460 component type was identified as the method proposed by Wang et al. (2022b): if the mass fraction of
 461 the maximum component was 1.2 times higher than that of the secondary one, the former was
 462 considered as the dominant factor, otherwise both dominated PM_{2.5} formation.



463

464

465 **Figure 10.** The spatial distribution of (a) MLH, (b) MDA8 O₃, (c) PM_{2.5}, (d) the dominant PM_{2.5}
 466 chemical component (N: NO₃⁻ dominant, NS: NO₃⁻ and SO₄²⁻ dominant, NO: NO₃⁻ and OM dominant,
 467 S: SO₄²⁻ dominant, SN: SO₄²⁻ and NO₃⁻ dominant, SO: SO₄²⁻ and OM dominant, O: OM dominant, ON:
 468 OM and NO₃⁻ dominant, OS: OM and SO₄²⁻ dominant), (e) T, and (f) RH, (g) the overall change
 469 characteristics of WS and WD in the NCP from June 24 to 29, 2021. The dominant PM_{2.5} chemical
 470 component type was identified as the method proposed by Wang et al. (2022b): if the mass fraction of
 471 the maximum component was 1.2 times higher than that of the secondary one, the former was
 472 considered as the dominant factor, otherwise both dominated PM_{2.5} formation.



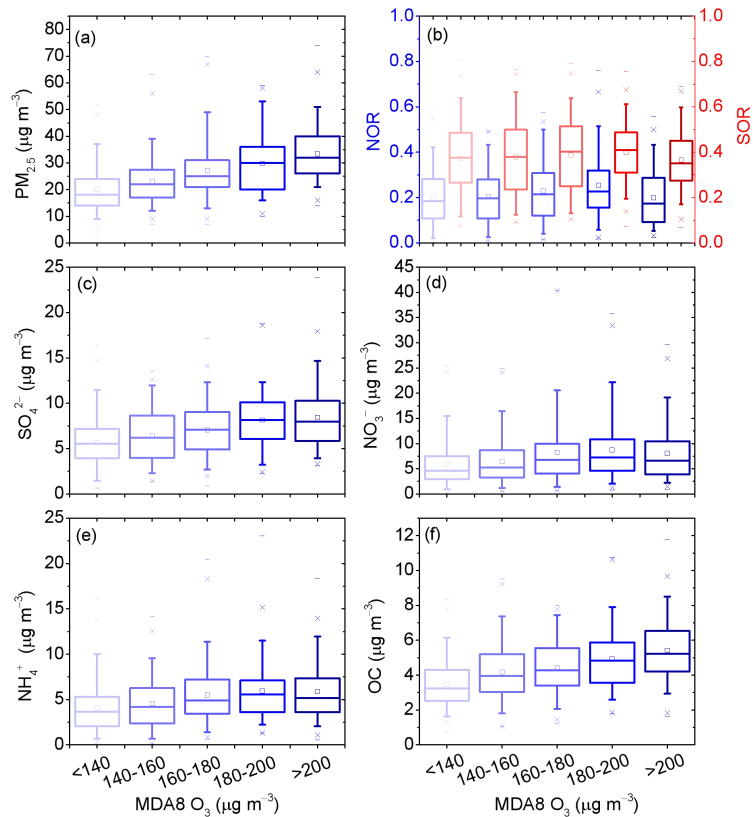
473

474 **Figure 11.** The distribution characteristics of (a) NOR and SOR, and (b) the mass fractions of major

475 PM_{2.5} components, MLH, RH, and temperature under O₃-PM_{2.5} CPD, O₃ PD, PM_{2.5} PD, and NPD

476 conditions from June 24 to 29, 2021.

477 **3.4.2 Interaction between PM_{2.5} and O₃ along with the evolution of MLH**



478

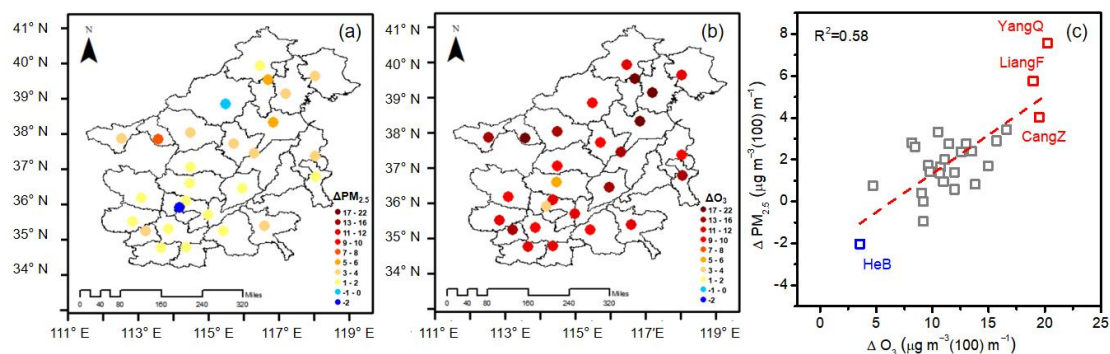
479 **Figure 12.** Box plots showing the statistics of (a) PM_{2.5}, (b) NOR and SOR, (c) SO₄²⁻, (d) NO₃⁻, (e)
480 NH₄⁺, and (f) OC for different MDA8 O₃ conditions (< 140 μg m⁻³, 140–160 μg m⁻³, 160–180 μg m⁻³,
481 180–200 μg m⁻³, > 200 μg m⁻³). The distance between the bottom and the top of the box reflects the
482 inter quartile range; the line and square in between are the median and mean values, respectively. The
483 whiskers above and below the box refer the 95 % confidence interval of the data. Note that rainy days
484 were excluded.

485 Figure 12 displays the box-and-whisker plots of PM_{2.5} and its major components for different
486 MDA8 O₃ conditions. To isolate the impacts of precipitation on PM_{2.5} concentration, these rainy days
487 when the daily rainfall amount greater than 0 mm were excluded in this section. Here the
488 concentrations of PM_{2.5} and its major components were found to increase synchronously with elevated
489 MDA8 O₃ concentration, especially when MDA8 O₃ increased from < 140 to 180–200 μg m⁻³. This
490 summertime collaborative growth process of PM_{2.5}-O₃ has also been observed in other works (Wang et
491 al., 2022a; Wu et al., 2022). With elevated MDA8 O₃ concentration, SOR and NOR both slightly
492 increased, and reached the maximum when MDA8 O₃ at around 160–200 μg m⁻³, which indicated the
493 strong secondary formation of SO₄²⁻ and NO₃⁻ promoted by high O₃ concentration. When MDA8 O₃
494 increased from 180–200 to > 200 μg m⁻³, the concentrations of NO₃⁻, NH₄⁺, and SO₄²⁻ kept stable or
495 started to decrease, and the values of SOR and NOR decreased synchronously. During this stage, the
496 high O₃ concentration often accompanied with dry and hot meteorological conditions, which was not
497 beneficial to the aqueous chemical production and was conducive to the partitioning of nitrate to the
498 gas phase.

499 To verify the potential impact of photochemical oxidation to the increase of PM_{2.5} concentration
500 with mixing layer development, the changes in PM_{2.5} and MDA8 O₃ along with the increase of MLH
501 were quantified in the “2+26” cities in the NCP. Linear regression equations between air pollutants and
502 MLH were fitted during the initial increasing stage (300 < MLH < 1200 m) and their slopes were given
503 in Figure 13. The slopes indicated the rates of the maximum changes in air pollutant concentration for a
504 unit change in MLH (100 m). The slopes of PM_{2.5} and O₃ were expressed as ΔPM_{2.5} and ΔO₃ (μg m⁻³
505 (100) m⁻¹). It was found that ΔPM_{2.5} was closely related to ΔO₃ (R²=0.58), and obvious spatial
506 difference in ΔPM_{2.5} and ΔO₃ was witnessed in the NCP during the observation period. ΔPM_{2.5} and ΔO₃
507 both showed high values in YangQ, , LangF and CangZ, with values of 7.56 and 20.24 μg m⁻³ (100)
508 m⁻¹ in YangQ, 5.75 and 18.97 μg m⁻³ (100) m⁻¹ in LangF, and 4.02 and 19.49 μg m⁻³ (100) m⁻¹ in

509 CangZ, respectively. Comparing with these cities, $\Delta PM_{2.5}$ and ΔO_3 were lowest in HeB, with the value
 510 of 3.54 and $-2.02 \mu g m^{-3} (100) m^{-1}$, respectively, which implied that the secondary formation here was
 511 weak and the surface $PM_{2.5}$ change characteristic was dominantly controlled by local emissions or
 512 vertical diffusion effect.

513 Comparing with winter, the photochemistry in summer is quite active due to the strong solar
 514 radiation. Even though deep MLH favors the dilution of air pollutants, higher MLH can also promote
 515 secondary chemical feedback through enhancing the availability of atmospheric oxidation capacity
 516 (such as changes in O_3) along with appropriate meteorological condition. This conclusion corresponded
 517 well to the finding based on chemical transport model (Dai et al., 2023), which proposed strong
 518 chemical production of secondary aerosols when planetary boundary layer height was about 946.1m on
 519 O_3 - $PM_{2.5}$ co-pollution days. The strong chemical productions in the oxidative atmosphere at medium
 520 MLH condition may overcome the dilution effect on $PM_{2.5}$ induced by mixing layer development,
 521 leading to higher $PM_{2.5}$ level at the ground level. However, it should be noted that the conclusions in
 522 this work were only suitable to summertime regional observations, especially for warm and humid
 523 seasons. The conditions would be different in wintertime (much lower O_3 level). More extended
 524 observations in time and space should be needed in the future to further examine and better understand
 525 the complex interactions between MLH, air pollution, and chemical processing.



526
 527 **Figure 13.** The spatial distribution of (a) $\Delta PM_{2.5}$ and (b) ΔO_3 . (c) The relationships between $\Delta PM_{2.5}$ and
 528 ΔO_3 in the NCP during summertime. The corresponding correlation coefficients (R^2) was given at the
 529 top of the panel.

530 4 Conclusions

531 Mixing layer height (MLH) was generally considered as a critical physical parameter in

532 atmospheric environmental evaluation. It is assumed that extended mixing layer may lead to the
533 dilution of air pollutants and thus tend to decrease surface concentrations. Several publications have
534 indeed reported such anti-correlations in cold seasons. However, the understanding of the interaction
535 between near surface O₃ and PM_{2.5} (including its major components) along with the evolution of
536 mixing layer during warm seasons remained poor. Furthermore, previous observational studies were
537 mostly limited to a specific city. This paper is devoted to these topics by examining the response of
538 MDA8 O₃, PM_{2.5}, and its major components to the changes in mixing layer meteorology in the North
539 China Plain (NCP) during summertime. We showed that MDA8 O₃ initially increased and then
540 decreased with the growth of MLH. The maximum turning point of MLH was around 900–1800 m. As
541 for near-ground PM_{2.5}, similar non-linear change profile was found, with the maximum value of 31.65
542 μg m⁻³ under medium MLH condition (900–1200 m), which was quite different from the results
543 conducted in cold seasons. Compared with winter, the occurrence of low MLH during summertime in
544 the NCP was mostly accompanied with cloudy or rainy conditions, which promoted wet deposition and
545 led to low concentrations of PM_{2.5} at the ground level. Under medium MLH condition, strong chemical
546 productions of SO₄²⁻ and OC occurred along with appropriate mixing layer meteorology, where RH
547 was around 50–70 %, and the availability of atmospheric oxidants (i.e., O₃) increased. The strong
548 chemical productions at medium MLH conditions may offset the diffusion effect on PM_{2.5} induced by
549 mixing layer development, resulting in higher PM_{2.5} levels. The chemical characteristics of PM_{2.5}
550 significantly changed along with the growth of MLH. The composited concentration of NO₃⁻ was the
551 highest under low MLH condition, while the composited concentrations of SO₄²⁻ and OC obviously
552 increased under medium MLH condition. Temperature was the key factor controlling the opposite
553 changes in NO₃⁻ and SO₄²⁻ concentrations in PM_{2.5}. We conclude that the MLH can be an indicator of
554 air pollutants in cold seasons, but the correlation between MLH and air pollutants, such as O₃ and
555 PM_{2.5}, should be treated with care in hot seasons. At least for the observation period in the NCP this
556 was not the case. Although several studies have examined the change characteristics of MLH and its
557 influence on ground-level O₃ and PM_{2.5}, it remains challenging to elucidate the mechanism underlying
558 the complex relationships. In this work, we did not quantify the sensitivity of O₃ and PM_{2.5} to different
559 meteorological factors and chemical processes. To better understand the complex interactions between
560 MLH, air pollution, and chemical processing, a more detailed consideration the aids of explicit models
561 should be needed in the future. We also note that the present study was only confined to summertime

562 conditions (including two summer months) in the NCP, and the conclusions is likely to be different in
563 other seasons and regions. Thus, more extended observations in time and space should be needed in the
564 future.

565

566 **Data availability.** The data used in this paper can be provided upon request from the corresponding
567 author.

568

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570 **Y, Y Y, L L and Y X** analyzed the data. **J W** prepared the manuscript and all the coauthors helped
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577

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