

1 **Vertical Distribution Characteristics and Formation/Dissipation**
2 **Mechanisms of Air Pollutants in Xingtai, China Based on Multi-source**
3 **Data: A Case Study**

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9 **Abstract**

10 The Beijing-Tianjin-Hebei urban agglomeration is one of the regions in China with the
11 most severe air pollution. Using aircraft observations collected over Xingtai in May 2016
12 and multi-source data, such as aerosol chemical composition and lidar data, we analyzed
13 aerosol composition and optical properties, vertical pollution characteristics of gases
14 within the boundary layer, and their interactions with meteorological parameters. This
15 study focuses on investigating the transport and evolution mechanisms of pollutants during
16 transitions from polluted research flight No.7(RF7) to clean (RF8) periods in summertime
17 Xingtai. Results show that during RF7, the near-surface submicron aerosol (PM₁) mass
18 concentration was generally low (37.5 μg m⁻³), with the contribution of inorganic salts far
19 exceeding that of organic matter. During RF8, the near-surface wind speed slightly
20 increased, and the dominant wind direction remained southeasterly. Vertically, pollutant
21 concentrations showed notable decreasing trends, and the average near-surface PM₁ mass
22 concentration slightly decreased (30.6 μg m⁻³). However, the chemical components of PM₁
23 differed substantially from those during RF7. Aircraft observations indicated weak cold-
24 air activities above 1000 m during RF8, while the southeasterly wind still prevailed below
25 1000 m, with a slight increase in wind speed. From RF7 to RF8, the overall vertical
26 atmosphere gradually transitioned from polluted to clean conditions. During RF8, an
27 inversion layer appeared in the temperature profile between 1100 m and 1300 m, with
28 ~21.3% of this inversion coming from black carbon heating. During RF7, sulfur dioxide
29 (SO₂), nitrogen dioxide (NO₂), and carbon monoxide (CO) were strongly correlated with
30 the scattering coefficient, with correlation coefficients of 0.92 and 0.96 for SO₂ and NO₂,
31 respectively. By contrast, during RF8, the correlations between SO₂, NO₂, and CO and the
32 scattering coefficient decreased, with SO₂ having the highest correlation with the scattering
33 coefficient (coefficient of determination = 0.88). From RF7 to RF8, the concentrations of
34 various pollutants within the boundary layer, except for ozone (O₃), had decreasing trends.
35 When the relative humidity was below 50%, O₃ generally contributed positively to
36 extinction. There was no pronounced correlation between O₃ and relative humidity when
37 the relative humidity exceeded 50%.

38 **Keywords** Boundary layer · Pollutants · Vertical observations · Chemical composition ·
39 Meteorological factors

40 **1 Introduction**

41 The planetary boundary layer (PBL) serves as a pivotal hub for the exchange of energy,
42 water vapor, and matter between the surface and the atmosphere. In the PBL, physical
43 quantities, such as wind, temperature, humidity, pressure, and turbulent diffusion, as well
44 as chemical properties related to pollutant compositions and transformation mechanisms,
45 vary dramatically in time and space, particularly in the vertical direction [Haskins et al.,
46 2019; Jiang et al., 2021; Riedel et al., 2013; Sun et al., 2016b, 2020]. The formation and
47 dissipation of atmospheric pollutants primarily occur within the PBL [Qu et al., 2017]. The
48 physicochemical properties and evolution mechanisms of pollutants in the vertical
49 direction remain crucial factors affecting the accuracy of air quality forecasting models
50 [Brook et al., 2013]. Due to limitations in observation platforms and instrument resolution,
51 existing studies on atmospheric pollution have primarily focused on ground-level
52 measurements. It is still a complex and hot topic as to how the physical and chemical
53 properties of the urban boundary layer interact with each other to jointly drive the
54 formation, accumulation, and dissipation of atmospheric pollutants.

55 Vertical observation methods for atmospheric pollutants include comparisons of
56 observations at different altitudes [Levin et al., 2009], meteorological tower observations
57 (such as observations made at the Beijing 325-m Meteorological Tower [Wang et al.,
58 2018]), sounding balloon or tethered balloon observations [Han et al., 2018], ground-based
59 radar observations [Wang et al., 2012], aircraft measurements [Wang et al., 2024], and
60 model simulations [Wu et al., 2011]. Each of these approaches can provide valuable
61 insights into the vertical distribution and evolution of pollutants within the PBL. Friedrich
62 et al. [2012] found that considering the liquid-water mixing ratio derived from the vertical
63 humidity profile under moist-adiabatic conditions can provide a more precise
64 determination of atmospheric stability. Zhou et al. [2018] utilized multi-source
65 observations and numerical models to study the Yangtze River Delta region, revealing that
66 the lifting of a cold front and the transport of pollutants facilitate the accumulation of
67 pollutants at altitudes above 100 m rather than near the surface. Based on aircraft
68 observations, Dong et al. [2017] demonstrated that surface cooling effects and the uneven
69 changes in heating rates caused by absorbing aerosols altered the atmospheric temperature
70 profile, resulting in increased stability within the PBL and instability above the PBL. The

71 vertical variation characteristics of pollutants within the urban boundary layer are closely
72 related to the dynamic/thermodynamic structure and turbulence characteristics of the PBL.
73 Pollutants do not simply decrease with altitude, e.g., aerosol mass concentrations under
74 clean weather conditions decrease rapidly with altitude, whereas in polluted or stagnant
75 weather conditions, aerosol concentrations gradually decrease with altitude [Ding et al.,
76 2005]. The proportion of secondary organic carbon increases with altitude, as reported by
77 Ren et al. [2021]. Simulation studies focused on the Beijing region have indicated that local
78 sources contribute greatly to near-surface pollutants, with local contributions of nitrogen
79 dioxide (NO₂), sulfur dioxide (SO₂), and the mass concentration of particulate matter with
80 diameters less than 10 μm (PM₁₀) reaching up to 90%, 65% and 75%, respectively.
81 However, at an altitude of 1.1 km, the contributions from external sources (e.g., cities south
82 of Beijing) to SO₂ and PM₁₀ exceed 50% [Wu et al., 2011]. Studies on air pollution in
83 Beijing have suggested that the proportion of organic matter is the highest near the surface
84 due to surface emissions. Under polluted conditions, aerosols are dominated by nitrates
85 above 200 m [Sun et al., 2020]. At night, O₃ concentrations are higher in the upper
86 boundary layer, where the hydrolysis of dinitrogen pentoxide (N₂O₅) to form nitrates
87 becomes more significant. The formed nitrates contribute to increased ground-level nitrate
88 concentrations the following day as the boundary layer mixes and develops [Sun et al.,
89 2018].

90 The Beijing-Tianjin-Hebei region is located in the transition zone between mountainous
91 and plain regions. Due to the joint effects of blocking systems and the unique topography,
92 the atmospheric environment in this region highly sensitive and vulnerable [Xu et al., 2021],
93 making it one of the regions with the most severe atmospheric pollution in China [Cai et
94 al., 2020]. Xingtai, situated in southern Hebei Province, China, has a complex emission
95 environment and high-intensity pollution emissions, making it a hotspot of air pollution in
96 the Beijing-Tianjin-Hebei region. Air pollution is the result of the combined and interacting
97 effects of multiple boundary-layer factors [Ren et al., 2021].

98 Although previous studies have enhanced our understanding of the boundary layer
99 structure and pollution mechanisms in the Beijing-Tianjin-Hebei urban agglomeration,
100 further analysis and discussion of related issues are still needed to overcome some
101 limitations. Specifically, there is a lack of detailed spatiotemporal observations of the

102 boundary-layer structure, with most studies relying on low-resolution observations.
103 Moreover, the boundary-layer structure is highly complex, with substantial differences
104 under varying pollution conditions across different regions, thus requiring more extensive
105 data support. In this study, we utilize aircraft observations, in conjunction with near-surface
106 multi-source data, including submicron aerosol (PM₁) chemical composition, to analyze
107 the physicochemical and optical properties of aerosols, characteristics of the vertical
108 variation of pollution gases in the boundary layer and their interactions with meteorological
109 parameters. Furthermore, we focus on the transport and evolution mechanisms of a
110 pollution-to-clean process in Xingtai during the summer, aiming to provide insights into
111 the dynamic processes that occur during such transitions.

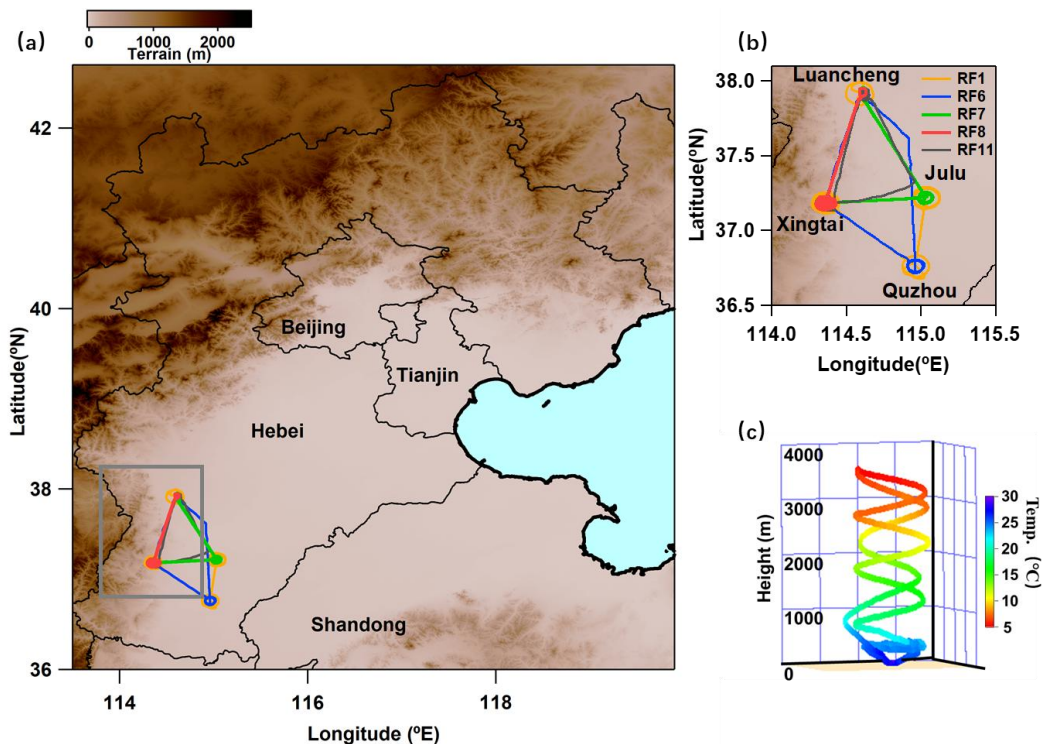
112 The remainder of this paper is organized as follows. Section 2 describes the sites and
113 campaigns. Section 3 analyzes the results. Finally, section 4 presents the main conclusions.

114 **2 Sites and campaigns**

115 To observe vertical profiles of aerosols and gases in the North China Plain, the Air
116 Chemistry Research in Asia campaign was conducted in Hebei Province, east of the
117 Taihang Mountains in the North China Plain, from May to June of 2016. During the
118 campaign, 11 vertical observation experiments were carried out using the Y-12 aircraft
119 flying out of four local airports. The Y-12 aircraft was equipped with various instruments
120 to measure aerosol optical properties, gas concentrations, and meteorological data.
121 Specifically, the 550-nm aerosol scattering coefficient was measured using an integrating
122 nephelometer (model 3565, TSI Inc.) at three wavelengths with a temporal resolution of
123 one second. The 550-nm absorption coefficient was measured by using a particle soot
124 absorption photometer (Radiance Research Inc., USA) with a temporal resolution of one
125 minute. The extinction coefficient is defined as the sum of the absorption and scattering
126 coefficients. Wang et al. [2018, 2024] and Benish et al. [2020] provide detailed information
127 about the observational instruments used in this experiment and flight trajectories. In the
128 Xingtai region, a total of five flight experiments were conducted, referred to as RF1, RF6,
129 RF7, RF8, and RF11. Fig.1 shows the flight trajectories of these experiments.

130

131 On 28 May 2016, two aircraft observation missions were conducted over the Xingtai region,
 132 one at ~0300 Coordinated Universal Time (UTC, experiment RF7, 11 am local time) and
 133 the other at 0900 UTC (experiment RF8, 5 pm local time). Sky conditions, as photographed
 134 by onboard observers, were different during each observation period (Fig. 2). During the
 135 RF7 flight, the sky at an altitude of 3 km appeared grayish-yellow, with low horizontal
 136 visibility. By contrast, during the RF8 flight, the horizontal visibility at high altitudes
 137 dramatically improved, and the sky appeared bright blue.



138 **Fig. 1.** (a) Five flight trajectories over Xingtai (colored lines) with terrain elevation (in m) as the background,
 139 (b) a zoomed-in view of the flight trajectories showing the locations of the airports that the aircraft flew from,
 140 and (c) vertical legs of each flight colored by the corresponding ambient air temperature.
 141



142 **Fig. 2.** Aerial photographs taken during the RF7 and RF8 flights.
 143

144 In conjunction with aircraft observations, various ground-based observations, including
145 aerosol chemical composition and meteorological parameters, were simultaneously
146 measured at the Xingtai supersite (114.36°E, 37.18°N; 182 m above sea level). Specifically,
147 the chemical components of non-refractory PM₁ (NR-PM₁), including sulfate (SO₄²⁻),
148 nitrate (NO₃⁻), ammonium (NH₄⁺), chloride (Cl⁻), and organics (Org), were measured in
149 situ using an Aerodyne Aerosol Chemical Speciation Monitor (ACSM) at temporal
150 resolutions of 5–8 minutes [Jiang et al., 2014; Sun et al., 2020b; Zhang et al., 2018]. Near-
151 surface black carbon (BC) measurements were conducted using a seven-wavelength
152 Aethalometer (model AE33, Magee Scientific, USA) [Drinovec et al., 2015; Wang et al.,
153 2018]. The BC data used in this study were measured specifically at the 880-nm
154 wavelength. Additionally, a MPL (Sigma Space Corporation, USA) was deployed at the
155 Xingtai supersite. Particle number concentrations were measured using a scanning mobility
156 particle sizer (model 3081/3085) with a resolution of five minutes. Ground-based
157 extinction coefficients of particulate matter were obtained from a Cavity Attenuated Phase
158 Shift-based extinction monitor. In addition, wind profiler radar data (114.65°E, 36.45°N;
159 58 m above sea level) with a temporal resolution of six minutes were provided by the
160 Meteorological Observation Center of the China Meteorological Administration, offering
161 parameters such as horizontal wind speed, wind direction, and vertical velocity.

162 **3 Results**

163 **3.1 Ground-based observations**

164 Figure 3 presents PM₁ and its chemical components (SO₄²⁻, NO₃⁻, NH₄⁺, Cl⁻, organic
165 matter, and BC), ground-level organic sources derived from the source apportionment of
166 organic matter, meteorological elements, particle number concentrations, and the vertical
167 backscattering coefficient obtained from the MPL on 28 May 2016. The vertical
168 atmospheric evolution of pollutants on that day can be divided into four distinct stages,
169 denoted as S1, S2, S3, and S4. The RF7 and RF8 flight observations occurred during stages
170 S1 and S2, respectively. Based on MPL observations from the Xingtai supersite, we
171 analyzed the PBL heights (PBLHs) retrieved from the normalized relative backscatter
172 using the wavelet covariance transform method [Gamage and Hagelberg, 1993] after
173 eliminating the effects of clouds. The step size and threshold were set to 210 m and 0.05,

174 respectively. The first value from the lower layer exceeding this threshold or falling within
175 a given threshold interval is considered to be the micropulse lidar-derived PBLH. Li et al.
176 [2018] provide more details.

177 During stage S1, the surface wind was dominated by southeasterly wind, and the PBLH
178 gradually increased. Near the surface up to an altitude of 500 m, radar echoes were
179 relatively strong, indicating notable pollutant accumulation. However, ground-level PM₁
180 mass concentrations remained relatively low at 37.5 μg m⁻³. The contribution of inorganic
181 salts (NO₃⁻ + SO₄²⁻ + Cl⁻) to PM₁ was considerably higher than that of organic matter,
182 especially nitrates, which accounted for ~28% of the total PM₁ composition on average. In
183 general, high temperatures and an elevated PBLH facilitate nitrate volatilization and
184 concentration diffusion during the daytime, especially in summer, when nitrate
185 concentrations begin to decrease markedly after sunrise. However, a noticeable peak in
186 nitrate mass concentration was observed around 0200 UTC (10 am local time) on 28 May,
187 surpassing the concentration of organic components. Importantly, there were no notable
188 changes in wind direction and speed during this period, suggesting that the horizontal
189 transport conditions of pollutants remained relatively stable. Studies on airship
190 observations [Sun et al., 2018] and aircraft measurements [Pusede et al., 2016] have shown
191 that regardless of pollution levels, the incremental contribution of nitrates above 200 m and
192 higher altitudes is quite high during the nighttime. This is primarily attributed to the
193 hydrolysis of N₂O₅ (formed from the reaction between high concentrations of NO₂ and O₃)
194 in the upper boundary layer at night, generating substantial amounts of nitrates. Around
195 sunrise, especially from 0100 to 0200 UTC, when boundary layer activities become more
196 vigorous, the mixing effect becomes pronounced within the boundary layer, bringing high-
197 concentration nitrates in the upper boundary layer down to near the surface, thereby
198 resulting in a marked rise in the ground-level nitrate concentration. Therefore, the peak in
199 the ground-level nitrate concentration around 0200 UTC was likely primarily due to the
200 nitrates generated by the hydrolysis of N₂O₅ in the upper boundary layer during the
201 previous night.

202 Using the positive matrix factorization method (Zhang et al., 2018), three types of organic
203 aerosols were identified: two primary organic aerosols and one secondary organic aerosol,
204 namely, hydrocarbon-like organic aerosol, cooking-related organic aerosol and oxygenated

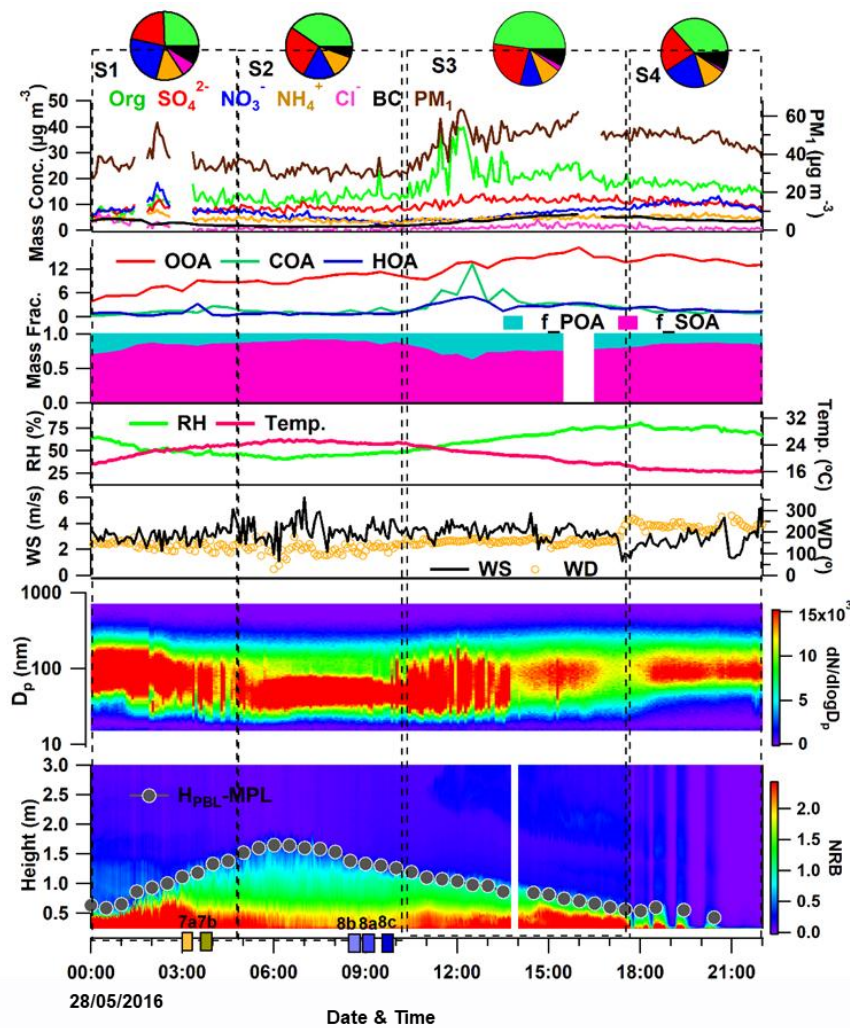
205 organic aerosol. During stage S1, the oxygenated organic aerosol concentration gradually
206 increased, and the proportion of secondary organic aerosol was over 65% of the organic
207 matter on average, with an increasing trend. During this stage, one flight mission (RF7)
208 with two vertical spirals (RF7a and RF7b) was conducted.

209 During stage S2, the near-surface wind speed increased slightly, and the wind direction
210 fluctuated, but the prevailing wind direction remained southeasterly. Micropulse lidar
211 (MPL) observations indicate that the thickness of the pollution layer decreased
212 substantially during stage S2, with a clear reduction in pollutant concentration in the
213 vertical direction. Although the average ground-level PM_{10} mass concentration ($30.6 \mu\text{g m}^{-3}$)
214 decreased slightly, the overall downward trend was not pronounced. However, the
215 PM_{10} chemical composition in stage S2 differed dramatically from that in stage S1. Notably,
216 the contribution of nitrates decreased markedly during stage S2 (16%), while the proportion
217 of organic matter increased notably, which became the dominant component of PM_{10} (40%).
218 Furthermore, the share of secondary organic aerosol in organic matter increased even more,
219 contributing over 90%. During this stage, one flight mission (RF8) with three sub-missions
220 (RF8a, RF8b, and RF8c) was conducted.

221 Upon entering stage S3, surface humidity gradually increased, and micropulse-lidar-
222 derived radar echoes began to strengthen progressively in the vertical direction.
223 Concurrently, the near-surface PM_{10} mass concentration also rose notably. In particular, the
224 mass concentration of organic matter increased from below $15 \mu\text{g m}^{-3}$ to $40 \mu\text{g m}^{-3}$ within
225 two hours, with a growth rate exceeding $13 \mu\text{g m}^{-3}$. Studies have shown that [Sun et al.,
226 2016a, b; Wang et al., 2018] as pollution intensifies, the proportion of inorganic salts tends
227 to increase remarkably, while the contribution of organic matter decreases. However,
228 during stage S3, although the mass concentration of inorganic matter increased with
229 increasing pollution, its growth rate was much lower than that of organic matter, with the
230 average contribution of organic matter exceeding 48% of PM_{10} . According to the source
231 apportionment results of organic matter during this period from Zhang et al. [2018], the
232 mass concentration of cooking-related organic aerosols began to increase considerably
233 around 1000 UTC, reaching a peak at 1200 UTC, coinciding with the local dinner cooking
234 time in Xingtai. The rapidly increasing cooking-related organic aerosols contributed
235 greatly to organic matter, with a proportion of up to 40%. During this time, cooking-related

236 organic aerosols played a crucial role in the occurrence and development of pollution in
 237 stage S3.

238 After entering stage S4, the wind direction shifted to the west, and the wind speed
 239 decreased markedly. In addition, the near-surface relative humidity remained around 70%,
 240 and the ground-level PM_{10} mass concentration was slightly lower compared with stage S3,
 241 but the proportion of inorganic salts, particularly nitrates, increased rapidly. In terms of
 242 mass concentration variations, after nightfall, the mass concentrations of all chemical
 243 components either decreased or remained stable, except for nitrates, which slightly
 244 increased. Notably, a small peak in nitrate mass concentration was observed from 2000 to
 245 2100 UTC, corresponding mainly to the shift in wind direction and the increase in wind
 246 speed, suggesting that the primary influencing factor was the alteration of the pollution
 247 transport pathway.



248

249 **Fig. 3.** Time series of (from top to bottom) ground-level concentrations of aerosol chemical components,
250 source apportionment of organic matter, meteorological elements, particle number concentrations, and
251 normalized relative lidar backscatter (NRB) observations derived from the micropulse lidar (grey dots show
252 boundary-layer heights) on 28 May 2016. Time of vertical spirals over Xingtai from RF7 and RF8 are labeled
253 as 7a/b and 8a/b/c. Vertical dashed lines cutting through all panels represent the division of each stage
254 (S1–S4).

255 **3.2 Aircraft-based observations of optical properties**

256 The formation mechanisms of pollution can be considered from two aspects. One is the
257 chemical reactions of internal factors, such as the formation of secondary aerosols, and the
258 other is the impact of boundary-layer development on pollution, i.e., the external
259 environment associated meteorological conditions for pollution [Sun et al., 2018]. During
260 the RF7b (0400 UTC) airborne sub-mission over Xingtai, the vertical distribution of the
261 extinction coefficient clearly shows a pollution profile, with the maximum exceeding 300
262 M m^{-1} throughout the entire layer (Fig. 4). The wind profile (Fig. 5) reveals that from the
263 near-surface up to 850 hPa (1500 m), southerly winds prevailed, with low wind speeds ($<$
264 4 m s^{-1}) and relative humidity levels of 50% to 60%, all favoring the development of air
265 pollutants. Under the combined effects of terrain, emissions, and meteorological factors,
266 the persistent southerly winds in a weak pressure field constitute a typical weather
267 background for pollution formation in the North China Plain [Sun et al., 2018]. Near the
268 surface at that time, Xingtai was located between two high-pressure systems, which led to
269 weak pressure gradients and horizontal diffusion. Thus, the visibility ranged from 18 km
270 to 20 km. There was no obvious inversion in the atmosphere. At 500 hPa, a cyclone
271 positioned over eastern Inner Mongolia guided weak cold air southward. By 0200 UTC,
272 the southern edge of the cold air had reached northern Hebei, but Xingtai was still under
273 stagnant atmospheric conditions. Additionally, the formation of a convergence line before
274 the arrival of cold air further contributed to pollutant accumulation in Xingtai. At around
275 0900 UTC (RF8), the pressure gradient at 500 hPa gradually increased as the cold vortex
276 began to affect the Xingtai region. However, the weather system was manifested as a
277 forward-tilting trough structure, with the wind direction shifting to northeasterly and wind
278 speed increasing above 850 hPa. The weak cold air (northwesterly wind) primarily
279 impacted areas above 1250 m, while southeasterly wind still prevailed below this level,
280 with a slight increase in wind speed (including near the ground). Consistent with MPL

281 observations, the vertical atmosphere in the aircraft profile from RF7 (stage S1) to RF8
282 (stage S2) showed a transition from polluted to clean conditions, with a notable reduction
283 in aerosol extinction. Note that the degree of reduction in the extinction coefficient varied
284 considerably across different altitudes.

285 The RF8 temperature profile showed a pronounced inversion layer from 1100 m to 1250
286 m, with a maximum inversion strength of $0.02^{\circ}\text{C m}^{-1}$. Assuming that the heating rate of
287 BC absorption in a given layer is roughly proportional to the radiation reaching that layer,
288 the BC heating rate between 1000–1250 m was estimated to be $0.049^{\circ}\text{C h}^{-1}$ based on the
289 BC distribution during RF8a. This estimation was made using the method proposed by Lu
290 et al. [2020]. Assuming that the maximum heating duration of BC absorption is 4 hours,
291 the maximum temperature increases in this layer due to BC heating was estimated to be
292 0.20°C . The maximum inversion strength during RF8 was $0.94^{\circ}\text{C m}^{-1}$, with warming due
293 to BC heating accounting for $\sim 21.3\%$ of this inversion. Consequently, warm air mass
294 activities contributed to $\sim 78.7\%$ of the inversion at this altitude. The presence of the
295 temperature inversion facilitated the accumulation of pollutants such as BC at this altitude
296 [Deng et al., 2016; Han et al., 2017]. The accumulation of BC, in turn, promoted the
297 development of the inversion layer, indicating a bidirectional positive feedback effect. Due
298 to the inversion layer, the area with large values of extinction coefficient was also
299 concentrated in this layer. For instance, at an altitude of 1120 m, the extinction coefficient
300 was 186.5 M m^{-1} during RF7b and 184.8 M m^{-1} during RF8a, indicating no noticeable
301 decrease in the extinction coefficient.

302 Under the influence of cold air aloft, the relative humidity above the inversion layer at 1250
303 m during RF8a dramatically decreased. However, below the top of the inversion layer,
304 particularly within the inversion layer, there was no remarkable decrease in water vapor
305 content. This situation would still be conducive to the hygroscopic growth of particles and
306 liquid-phase chemical reactions, thereby inhibiting the reduction of pollutant
307 concentrations. Within the 1250–1500 m altitude layer above the inversion layer, the
308 extinction coefficient decreased considerably from RF7b to RF8a due to the disturbance
309 caused by weak cold-air activities, with the average extinction coefficient dropping from
310 approximately 190 M m^{-1} to 19 M m^{-1} . Below the inversion layer, there was no obvious
311 cold-air influence, and the southeasterly wind was still the prevailing wind direction. Note

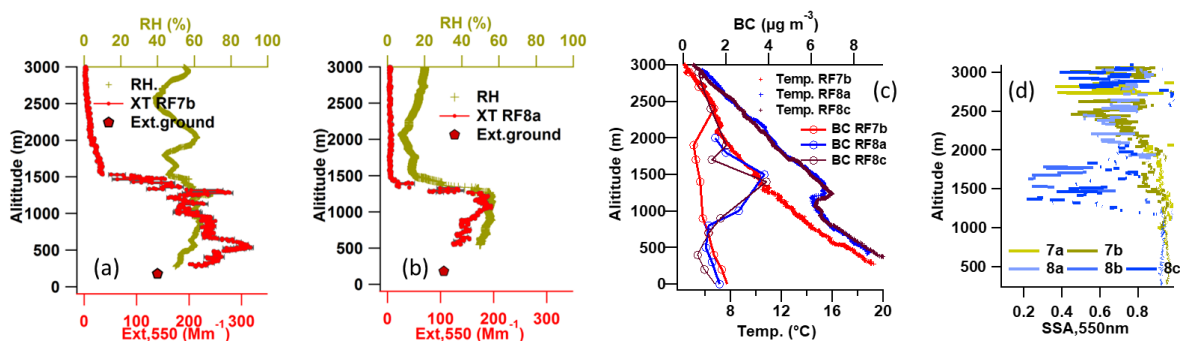
312 that observations were unavailable below 500 m in RF8a, so the 180–500 m layer was not
313 included in the analysis. On the one hand, the slight increase in wind speed at this altitude
314 slightly enhanced the horizontal diffusion of pollutants. On the other hand, the notable
315 differences in the chemical composition of near-surface PM_{10} compared with RF7 (stage
316 S1) and the fluctuations in wind direction (Fig. 3) may have caused a shift in the air mass
317 direction during RF8 (stage S2). The section discussing the backward trajectory analysis
318 will examine this further. As a result of these combined effects, there was also a certain
319 decreasing trend in pollutant concentrations, but the decreasing rate was lower than that in
320 the 1250–1500 m layer affected by weak cold-air activities. Specifically, in the 500–950-
321 m altitude range, the aircraft-measured extinction coefficient decreased from $\sim 271 M m^{-1}$
322 to $150 M m^{-1}$, while the ground-based observations showed a decrease in the extinction
323 coefficient from $140.1 M m^{-1}$ to $84.9 M m^{-1}$.

324 Table 1 presents the mean aerosol optical depth (AOD) values for different layers of the
325 atmosphere over Xingtai during the five flights from RF7a to RF8c, calculated by
326 integrating extinction coefficients over height [Wang et al., 2018]. Due to the influence of
327 the PBL, AOD values greater than 0.1 were mainly found within the boundary layer below
328 2000 m. Overall AOD values recorded during RF7a and RF7b both exceeded 0.3, with
329 values of 0.16 and 0.18 in the 0–1000 m layer, respectively, contributing over 53% to the
330 overall AOD. During RF8, the overall AOD value was noticeably lower compared with
331 RF7, with an average decrease of $\sim 51\%$. The single-scattering albedo, defined as the ratio
332 of the scattering coefficient to the total extinction coefficient at a specific wavelength
333 [Wang et al., 2018a], also varied. As the atmosphere transitioned from polluted (RF7) to
334 relatively clean (RF8) conditions, the single-scattering albedo within the altitude range of
335 1250–1800 m decreased. Specifically, the average single-scattering albedo in this layer
336 dropped from 0.85 during RF7 to 0.43 during RF8, indicating a prominent increase in the
337 contribution of aerosol absorption to total radiation in the altitude range of 1250–1800 m.
338 Correspondingly, the concentration of BC aerosols also increased markedly in this layer.

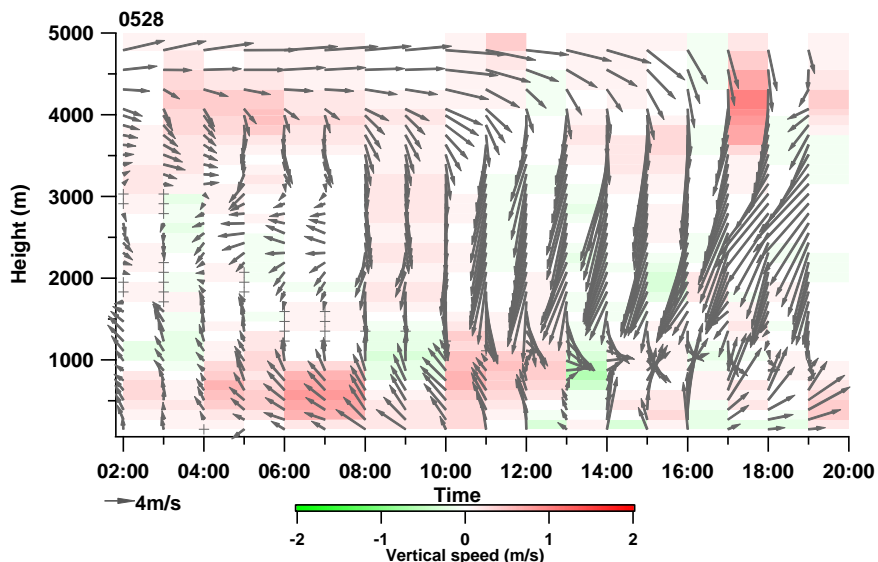
339 The bottom of the PBLH is defined as the point where the mean decreasing rate of the
340 aerosol scattering coefficient from aircraft measurements reaches $\sim 0.81 M m^{-1}$ [F. Wang
341 et al., 2018a]. PBLHs during RF7b and RF8a were thus identified as 1280 m and 1211 m,
342 respectively. However, PBLHs determined by the MPL wavelet method were 1333 m and

343 1288 m, respectively (Fig. 3). Although the PBLH estimates obtained from the wavelet
 344 method are relatively close to aircraft observations, they are slightly overestimated. Due to
 345 the pronounced diurnal cycles of the PBL, the airborne-derived PBLH in RF7b (0200 UTC)
 346 was higher than that in RF8a (0900 UTC), as shown in Fig. 4. During heavy pollution
 347 episodes, the PBLH and meteorological conditions in the PBL play a vital role in the
 348 cumulative explosive growth and diffusion of pollutants. High aerosol concentrations can
 349 enhance the stability of the urban PBL, thereby lowering the PBLH and further
 350 exacerbating pollution, which is known as the “two-way feedback mechanism” [Ding et
 351 al., 2016; Liu et al., 2019]. As the atmosphere gradually became cleaner after 0900 UTC,
 352 the changing meteorological conditions and the weakening feedback mechanism of
 353 pollutants on the PBLH notably slowed down the decreasing rate of the PBLH caused by
 354 the diurnal cycle (Fig. 3).

355 The wind field represents the characteristics of dynamic factors within the PBL. As shown
 356 in the wind profiler radar chart (Fig. 5), after entering stage S3 at 1000 UTC, the upper-
 357 level cold air continued to descend, reaching an altitude of ~900 m around 1300 UTC. This
 358 altitude was also the lowest level of this weak cold-air intrusion. Above 900 m, atmospheric
 359 diffusion conditions improved under the influence of the northerly wind. However, below
 360 900 m, the southerly wind still prevailed during stage S3. After 1000 UTC, the near-surface
 361 wind speed gradually decreased, and the PBLH further decreased, causing pollutants to
 362 accumulate once again.



363
 364 **Fig. 4.** Vertical distributions of the extinction coefficient and relative humidity during (a) RF7b and (b) RF8a,
 365 (c) vertical distributions of the black carbon (BC) concentration and temperature during RF7b, RF8a, and
 366 RF8c, and (d) the vertical distribution of the single-scattering albedo during the RF7 and RF8 flights.



367

368

Fig. 5. Vertical wind field measured by a wind profiler radar near Xingtai on 28 May 2016.

369

Table 1. Mean aerosol optical depths in different layers above Xingtai during the RF7 and RF8 flights.

Flights	Aerosol optical depths			
	0–1000 m	1000–2000 m	2000–3000 m	3000–4000 m
RF7a	0.16	0.14	6.23×10^{-3}	n/a
RF7b	0.18	0.15	6.81×10^{-3}	4.10×10^{-4}
RF8a	0.07	0.06	4.80×10^{-3}	2.22×10^{-4}
RF8b	0.11	0.07	n/a	n/a
RF8c	0.10	0.05	3.32×10^{-3}	5.09×10^{-4}

370

3.3 Air mass trajectory analysis

371

The 72-hour backward trajectories of air masses from 0000 UTC to 2300 UTC on 28 May

372

2016 were analyzed (Fig. 6). Results reveal a shift in the direction of air masses at an

373

altitude of 200 m between stages S2 (RF8) and S1 (RF7). During stage S2, the air masses

374

veered easterly, originating from the heavily polluted areas of eastern Hebei and

375

northwestern Shandong. After passing over the sea, the air masses traveled through Jiangsu,

376

Anhui, and Henan before reaching Xingtai. The water vapor contents of the air masses

377

increased during this journey. Compared with stage S1, the air masses during stage S2

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moved noticeably faster, corresponding to a slight increase in surface wind speed during

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this period. In stage S2, the direction of the air masses changed, and the faster movement

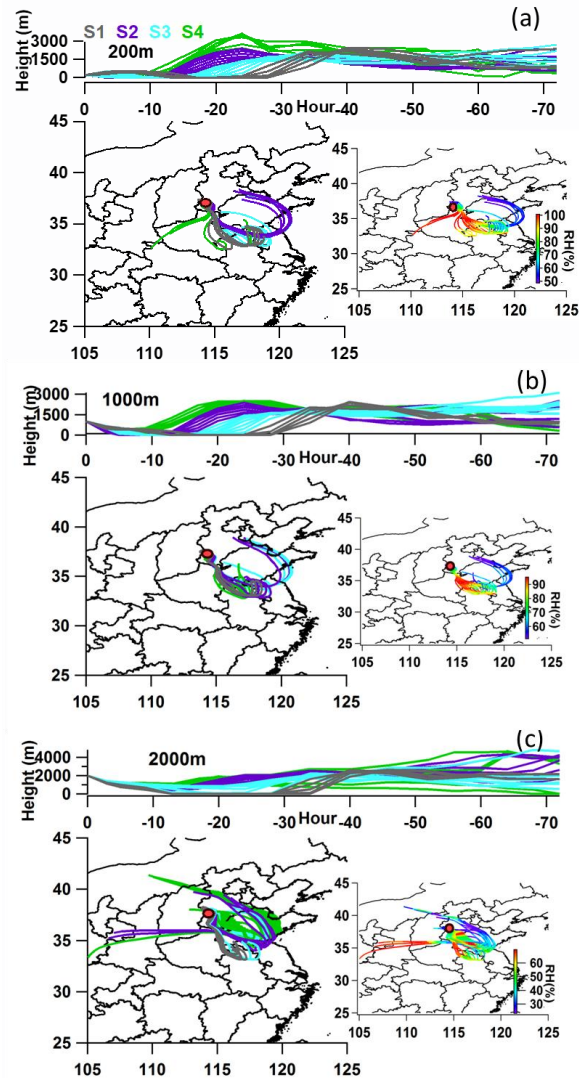
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of air masses improved atmospheric diffusion, resulting in a slight decrease in near-surface

381

pollutant concentrations compared with stage S1. In stage S3, the movement of air masses

382 slowed down again, with a direction similar to that in stage S1, leading to further pollution
 383 accumulation. During stage S4, at an altitude of 200 m, there was a pronounced adjustment
 384 in the direction of pollutant transport. Specifically, air masses originated from Henan in the
 385 southwest direction, and the relative humidity of these air masses was relatively high,
 386 facilitating the continued development of pollution. At an altitude of 1000 m within the
 387 PBL, the air masses consistently came from the southeast across all periods, with no
 388 marked difference in pollution sources between stages S1 and S2. However, at an altitude
 389 of 2000 m above the PBL, the air mass direction in stage S1 showed no notable difference
 390 from that at lower altitudes. By contrast, during stage S2, the air masses came from the
 391 relatively clean northwestern region and moved faster, leading to a cleaner atmosphere at
 392 higher altitudes.



393

394 **Fig. 6.** Seventy-two-hour backward trajectories at altitudes of (a) 200 m, (b) 1000 m, and (c) 2000 m from
395 0000 UTC to 2300 UTC on 28 May 2016. The upper figures in panels (a–c) show 72-hour backward
396 trajectory height maps of S1–S4, the lower left figures show the origins of the backward trajectories of S1–
397 S4, and the lower right figures show the relative humidity (unit: %) of trajectories from different origins.

398 **3.4 Aircraft-based observations of pollutant gases**

399 As showed in Fig.7, consistent with the changes in the extinction coefficient, the
400 concentrations of various pollutants (including SO₂, NO₂, and CO) in RF8 were noticeably
401 lower than those in RF7, primarily concentrated below 1500 m. Above 1500 m, the
402 concentrations of these pollutants quickly dropped to lower values as the background of
403 the free troposphere. The trend for O₃ was the opposite. Below 1500 m, the O₃
404 concentration in RF8 was higher than that in RF7. Between 1500 m and 2300 m, the O₃
405 concentration rapidly decreased from above 110 μg m⁻³ to ~75–82 μg m⁻³, lower than the
406 O₃ concentration at the same altitude in RF7. Above 2300 m, there was little difference in
407 the O₃ concentration between RF7 and RF8. Notably, the O₃ profile in RF7 did not rapidly
408 decrease at any particular altitude, remaining in the range of ~80 to 102 μg m⁻³ throughout
409 the entire layer.

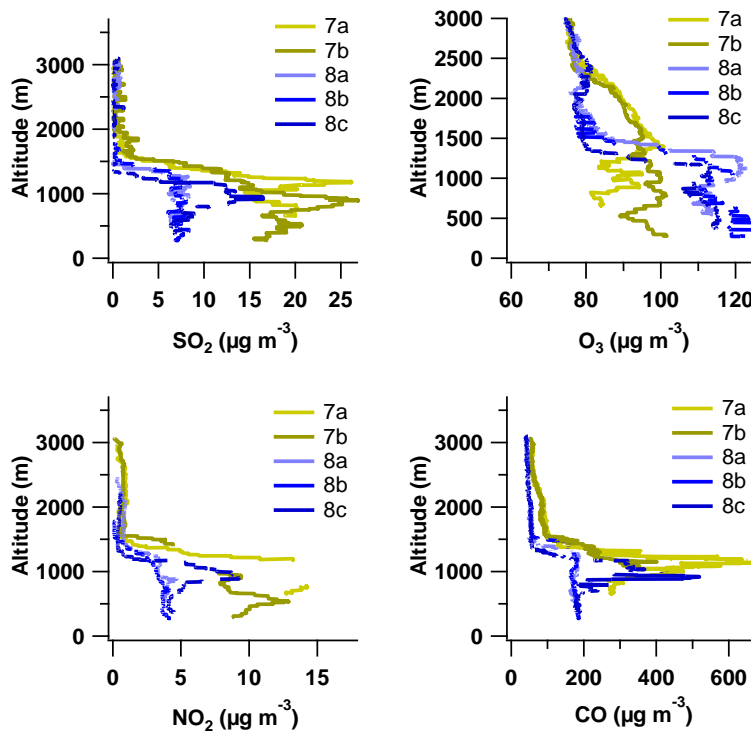
410 Gaseous and particulate pollutants from power plants, industrial emissions, and
411 transportation contribute substantially to atmospheric extinction in the vertical direction.
412 The correlations between pollutant gases and extinction (Figs. 8 and 9) reveal that in RF7
413 (representing a relatively polluted atmospheric background), SO₂, NO₂, and CO
414 concentrations were strongly correlated with the scattering coefficient. The correlation
415 coefficients(R^2) of SO₂ and NO₂ with the scattering coefficient were particularly high,
416 reaching 0.92 and 0.96, respectively. However, in RF8 (representing a relatively clean
417 atmosphere), these correlations decreased markedly. However, SO₂ was still highly
418 correlated with the scattering coefficient (coefficient of determination, $R^2 = 0.88$). Unlike
419 other pollutants, the correlation between O₃ and the scattering coefficient in RF7 differed
420 depending on the PBLH observed during the flight (with an average PBLH of 1211 m).
421 Above the boundary layer (PBLH > 1211 m), O₃ was positively correlated with the
422 scattering coefficient ($R^2 = 0.86$), with the corresponding scattering coefficients mainly
423 ranging from 0 to 100 M m⁻¹. This suggests that in the upper atmosphere, where the
424 influence of the boundary layer is reduced, the O₃ concentration and atmospheric scattering

425 properties were closely linked, potentially due to photochemical reactions and transport
426 processes affecting both O₃ and particulate matter. However, within the boundary layer
427 (PBLH ≤ 1211 m) in RF7, as the scattering coefficient increased, the O₃ concentration
428 gradually decreased. By contrast, in the relatively clean atmosphere in RF8, the O₃
429 concentration and the scattering coefficient showed a consistent positive correlation ($R^2 =$
430 0.90), even higher than the correlations between other gases and the scattering coefficient
431 during RF8. It shows O₃ and PM_{2.5} (both secondary pollutants) are formed from the same
432 emissions.

433 In RF7, O₃ generally contributed positively to extinction at relative humidity levels below
434 50%. However, when the relative humidity exceeded 50%, the contribution of O₃ to
435 extinction was either positive or negative. In RF8, a similar pattern was seen, with 50%
436 relative humidity serving as the threshold. Below this threshold, an increase in relative
437 humidity favored O₃ formation, leading to a higher O₃ concentration and a corresponding
438 rise in the scattering coefficient. Nevertheless, when the relative humidity exceeded 50%,
439 the O₃ concentration tended to fall in the 105–125 μg m⁻³ range. There was no apparent
440 correlation of O₃ and the scattering coefficient with relative humidity.

441 NO₂ and CO can be regarded as tracers for O₃ pollution [Kim et al., 2013]. NO₂ has a
442 relatively short lifespan and primarily influences O₃ through photochemical reactions. The
443 RF7 flight occurred in the morning (Beijing time), and solar ultraviolet radiation during
444 this period is generally notably weaker compared with noon and the afternoon hours.
445 Atmospheric pollution within the boundary layer also further weakens the amount of solar
446 ultraviolet radiation to some extent, inhibiting the photochemical cycle of O₃ formation,
447 thereby leading to the reduction and consumption of O₃. This phenomenon is known as the
448 titration effect. This is seen during the RF7 and RF8 flights, i.e., a decrease in the O₃
449 concentration accompanied by an increase in the NO₂ concentration (Fig. 9). Due to the
450 strong correlation between NO₂ and the scattering coefficient, the extinction coefficient
451 also showed a noticeable upward trend. CO is another important precursor of tropospheric
452 O₃. Above the boundary layer, where nitrogen oxide concentrations are lower, and titration
453 reactions are weaker, O₃ was not noticeably consumed. This maintained the relatively high
454 O₃ concentration above the boundary layer, where the CO mass concentration in RF7 was
455 also notably higher than that in RF8. During RF8, due to the influence of weak cold-air

456 activities, the concentrations of all pollutants within the boundary layer, except for O₃,
 457 decreased compared with RF7. The main reason for this may be that the RF8 flight took
 458 place in the afternoon (Beijing time) when sunlight was stronger. The atmosphere during
 459 RF8 was cleaner than that during RF7, leading to a marked reduction in the atmospheric
 460 extinction capacity of pollutants. At this time, enhanced solar radiation, particularly the
 461 intense ultraviolet radiation, intensified the photochemical cycling of nitrogen oxides,
 462 thereby actively promoting O₃ formation. Consequently, within the boundary layer, a
 463 positive correlation between O₃ and the scattering coefficient was found. Above the
 464 boundary layer, due to the influence of weak cold-air activities, the concentrations of O₃
 465 and other gaseous pollutants decreased to relatively lower levels. However, the correlation
 466 exhibited by different gases and extinction may involve complex physicochemical
 467 mechanisms, and further corroboration with a large amount of sample data is still required.
 468



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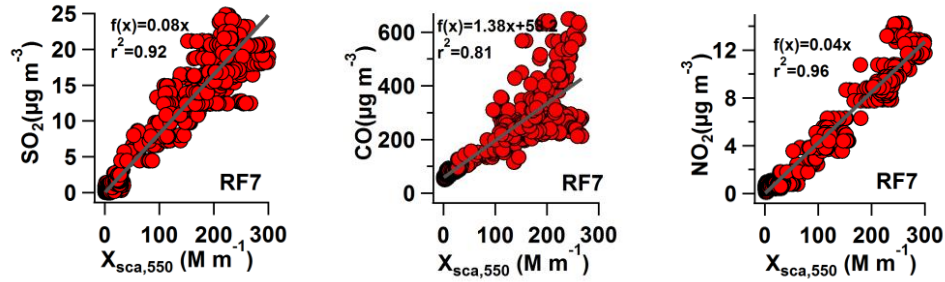
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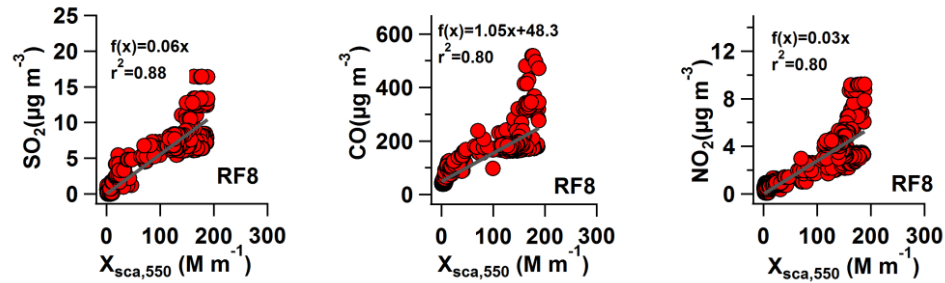
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Fig. 7. Vertical profile distributions of SO₂, O₃, NO₂, and CO concentrations during the RF7 and RF8 flights.

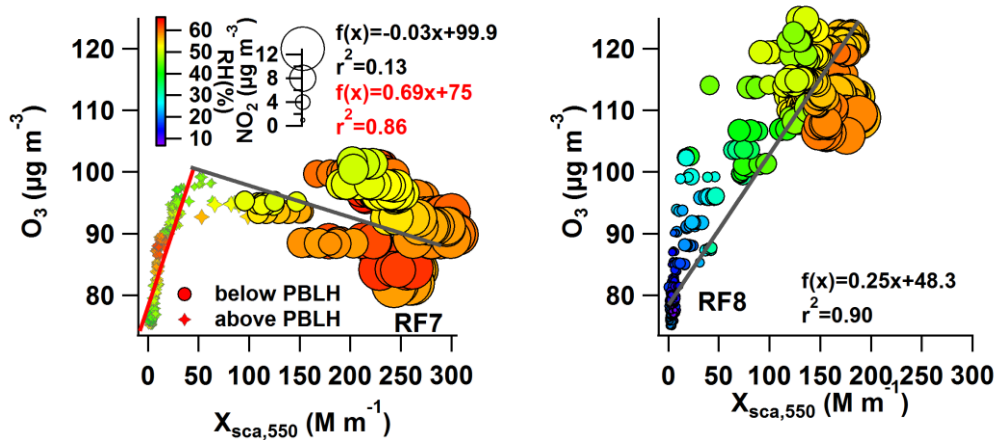
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474



475 **Fig. 8.** Correlations of SO₂, NO₂, and CO concentrations with the scattering coefficient during the RF7 (top
476 row) and RF8 (bottom row) flights.



477

478 **Fig. 9.** Correlations between the O₃ concentration and the scattering coefficient during the RF7 and RF8
479 flights. The circle color and size denote the relative humidity and the NO₂ concentration, respectively.

480

481 **4 Conclusions**

482 Air pollution remains a significant environmental issue in China, especially in the Beijing-
483 Tianjin-Hebei region. Within the boundary layer, various meteorological elements and
484 multiple boundary-layer components are complex and highly variable, playing a crucial
485 role in the formation and dissipation of pollution. Detailed observations within the
486 boundary layer are essential for accurately capturing the thermal and dynamic evolution of
487 boundary-layer structures during pollution processes, which in turn enhances our

488 understanding of the pollution mechanisms in the Beijing-Tianjin-Hebei region. The
489 factors influencing pollution development can be categorized into two aspects. One
490 involves the reactions related to the physicochemical properties of pollutants, such as the
491 formation of secondary aerosols. The other concerns the impact of external environmental
492 factors, such as the role of boundary-layer structures in pollution processes. In this study,
493 based on aircraft observations from Xingtai in Hebei made in May 2016, combined with
494 ground-level multi-source data, including PM₁ chemical composition, we investigate
495 aerosol composition and optical properties, vertical pollution characteristics of gases
496 within the boundary layer, and their interactions with meteorological parameters. Special
497 emphasis is placed on exploring the transport and evolution mechanisms of a pollution-to-
498 clean transition (RF7–RF8) in the Xingtai region during the summer.

499

500 From ground-level observations, the overall PM₁ mass concentration in stage S1 (RF7) was
501 relatively low at 37.5 μg m⁻³, and the contribution of inorganic salts was considerably
502 greater than that of organics. Upon entering stage S2 (RF8), the near-surface wind speed
503 slightly increased, and the wind direction fluctuated to some extent, but the prevailing wind
504 direction remained southeasterly. During stage S2, the thickness of the pollution layer
505 noticeably decreased, and pollutant concentrations showed distinct weakening trends in the
506 vertical direction. Although the average ground-level PM₁ mass concentration (30.6 μg
507 m⁻³) slightly decreased, the chemical composition of PM₁ during stage S2 differed
508 markedly from that during stage S1. The contribution of nitrates decreased to around 16%,
509 while the proportion of organics gradually increased to about 40%. The contribution of
510 secondary organic aerosols (over 90%) was greater than that of the organics. Aircraft
511 observations revealed that at ~0900 UTC (RF8), the pressure gradient at 500 hPa gradually
512 increased as a cold vortex began to influence the Xingtai region. The primary impact of
513 weak cold-air activities was observed above 1250 m, while below this height, the prevailing
514 wind direction remained southeasterly, and the wind speed increased. From RF7 (stage S1)
515 to RF8 (stage S2), the vertical atmosphere gradually transitioned from polluted to clean. In
516 RF8, the temperature profile had a pronounced inversion at an altitude of approximately
517 1100–1250 m, with a maximum inversion strength of 0.94°C m⁻¹. The warming from BC
518 heating contributed about 21.3% of this inversion. The presence of the inversion facilitated

519 pollutant accumulation, and the BC accumulation further promoted the development of the
520 inversion layer, indicating a bidirectional positive feedback effect between the two. The
521 backward trajectory analysis shows that air masses at both 2000 m and 1000 m during stage
522 S1 came from the southeast direction. However, during stage S2, the air masses at 2000 m
523 shifted toward the cleaner northwestern region compared with those at the same height in
524 stage S1, moving at a faster speed, which corresponds to the transition of the upper
525 atmosphere to a cleaner state.

526 In RF7, SO₂, NO₂, and CO concentrations were strongly correlated with the scattering
527 coefficient, with correlation coefficients of 0.92 and 0.96 for SO₂ and NO₂, respectively.
528 However, in RF8, these correlations dramatically decreased. However, the SO₂
529 concentration was still relatively highly correlated with the scattering coefficient ($R^2 =$
530 0.88). From RF7 to RF8, the concentrations of all pollutants within the boundary layer,
531 except for O₃, had decreasing trends. Within the boundary layer, O₃ concentrations
532 gradually decreased as the scattering coefficient increased. Above the boundary layer, the
533 O₃ concentration was positively correlated with the scattering coefficient ($R^2 = 0.86$), with
534 corresponding scattering coefficients mainly ranging from 0 to 100 M m⁻¹. Whether under
535 clean (RF8) or polluted (RF7) weather conditions, O₃ generally contributed positively to
536 extinction when the relative humidity was below 50%. However, when the relative
537 humidity exceeded 50%, the contribution of O₃ to extinction was either positive or negative.
538 There was no apparent correlation of O₃ and the scattering coefficient with relative
539 humidity.

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