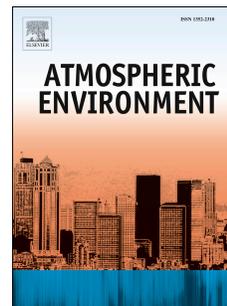


# Journal Pre-proof

Horizontal flux of ozone in the planetary boundary layer in Hong Kong using wind LiDAR measurements

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1 Horizontal flux of ozone in the planetary boundary layer in Hong Kong using wind

2 LiDAR measurements

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17 **Abstract.** While the crucial roles of ozone ( $O_3$ ) transport in the planetary boundary layer (PBL)  
18 have been acknowledged for some time, there is currently limited knowledge about this aspect  
19 primarily due to the limited availability of measurements to determine the characteristics of the  
20 PBL. In this study, measurements from a wind Light Detection and Ranging (LiDAR) system were  
21 taken to monitor vertical profile of wind pattern at an urban site in Hong Kong in September 2022,  
22 a period when the city was frequently impacted by tropical cyclones and experienced severe  $O_3$   
23 pollution levels. The PBL height was identified based on the vertical profile of wind speed shear.  
24 By combining information on the PBL height, vertical wind profile, and  $O_3$  concentration, we  
25 performed a cross-sectional analysis to explore the total horizontal flux (THF) of  $O_3$  across the  
26 PBL in Hong Kong. Throughout the entire study month, the THF of  $O_3$  exhibited a predominant  
27 easterly component. However, during the  $O_3$  pollution episodes, the THF of  $O_3$  exhibited a  
28 predominant westerly component, indicating an increased regional transport from Greater Bay  
29 Area (GBA). The westerly winds between  $240^\circ$  and  $300^\circ$  contributed 61.2% to the total flux of  $O_3$   
30 in the PBL during these episodes. In addition, clockwise veering winds were observed from the  
31 ground to the top of the PBL, which can be attributed to the Ekman spiral. As a result, during the  
32  $O_3$  pollution episodes, the wind with the peak  $O_3$  flux shifted from westerly to northwesterly as  
33 the height increased in the PBL. The northwesterly THF of  $O_3$  between  $290^\circ$  and  $300^\circ$  reached its  
34 peak at 600 m above ground level during these episodes. These findings enhance our understanding  
35 of the 3D pollutant transports for both long-term averages and short-term pollution episodes in the  
36 GBA and Hong Kong.

37 **Keywords:** LiDAR, Ozone, Pollution flux, Planetary boundary layer, Vertical variation.

## 38 1. Introduction

39 The Greater Bay Area (GBA) has become one of the largest city clusters in the world due  
40 to rapid urbanization and industrialization (Yu et al., 2023). However, in exchange for its  
41 tremendous economic development, the region has also experienced high levels of air pollution  
42 (Fan and Li, 2023). In recent years, ozone ( $O_3$ ) pollution has become prominent due to the non-  
43 linear response of  $O_3$  to the emission controls of its precursors (Guo et al., 2023; Tang et al., 2022).  
44 Because  $O_3$  is a secondary pollutant, regional transport can play a dominant role in the formation  
45 of severe  $O_3$  pollution episodes in the GBA region (Li et al., 2013; Shen et al., 2022). Although  
46 various efforts have been made to identify the causes of severe  $O_3$  pollution episodes, a complete  
47 picture of the three-dimensional (3D) transport of  $O_3$  in this region remains unclear due to a lack  
48 of upper-air observations.

49 The regional transport of  $O_3$  is largely determined by the 3D wind field in the planetary  
50 boundary layer (PBL), which, however, is highly variable (Zhang et al., 2023). The wind field in  
51 the PBL over the GBA region can be influenced by various factors, such as synoptic weather  
52 patterns, mesoscale atmospheric circulations, urban heat island effect, and complex terrain (He et  
53 al., 2021; Xia et al., 2023). For instance, confluence zones of wind fields are often found at  
54 different locations in Hong Kong due to atmospheric circulations (Fung et al., 2005). Yang et al.  
55 (2019) analyzed severe air pollution in Hong Kong and found that strong vertical wind shear within  
56 the PBL was positively correlated with surface pollutant concentrations during pollution episodes.  
57 Therefore, the characteristics of the wind field in the PBL can significantly influence the  
58 development of severe air pollution episodes (Cruz et al., 2023; Yan et al., 2022).

59 It is common to observe winds veering from the bottom to the top of the PBL, which is  
60 denoted as the Ekman spiral (Ekman and Kullenberg, 1905). This can be attributed to the decrease

61 in friction as a significant force with increasing height. The winds are geostrophic in the free  
62 atmosphere, resulting from a balance between the pressure gradient force and Coriolis effect. As  
63 a consequence, wind direction is parallel to the isobars. However, within the PBL, the pressure  
64 gradient force and Coriolis effect are augmented by the frictional force. Friction causes air to spiral  
65 into low pressure areas since it reduces the magnitude of the Coriolis force. Theoretically, the wind  
66 direction in the PBL rotates clockwise with altitude in the northern hemisphere. This phenomenon  
67 was reported in Beijing using a radar wind profiler (Wang et al., 2023). A similar spiral-shaped  
68 wind profile was observed in another study in Beijing using measurements from a meteorological  
69 tower (Zhang et al., 2021). Due to the opposite Coriolis effect in the southern hemisphere, the wind  
70 direction may rotate counterclockwise with altitude in the PBL (Potts et al., 2023). Considering  
71 the veering wind profiles, which may impose significant asymmetrical loadings on structures,  
72 becomes essential in the design of super-tall buildings or gigantic wind turbines (Tse et al., 2016).  
73 However, observational investigation of the impacts of the wind spiral on pollution transport  
74 remains limited.

75 In addition to the wind field in the PBL, the height of the PBL also plays an important role  
76 in governing the evolution of severe air pollution episodes (Paul and Das, 2022). Pollutants  
77 released from the ground are often trapped within the PBL (Jiang et al., 2022). This is because the  
78 PBL is characterized by a capping inversion layer at its top (Liu et al., 2022). As a result, the  
79 interactions of mass and momentum between the PBL and free atmosphere are limited. In specific  
80 meteorological conditions, the suppressed PBL can contribute significantly to the rapid increase  
81 in ground-level pollutant concentrations (Su et al., 2020). Thus, the majority of pollutant transport  
82 between different regions occurs within the PBL. Quantifying this transport within the PBL is  
83 crucial for understanding the regional transport of air pollutants.

84 While the crucial roles of O<sub>3</sub> transport in the PBL have been acknowledged for some time,  
85 there is currently limited knowledge about this aspect. This is primarily attributed to the limited  
86 availability of measurements to determine the characteristics of the PBL. Previous studies were  
87 mostly conducted based on ground-level measurements, which are insufficient to depict a complete  
88 picture of 3D wind and pollution fields. Vertical measurements have mainly relied on aircraft,  
89 meteorological tower, and balloon-borne monitoring (Zhou et al., 2022). These vertical  
90 measurements, however, either lack spatiotemporal resolution or are costly in terms of data  
91 collection (Li et al., 2022). The detailed variations in the wind field and pollutant transport in the  
92 PBL have yet to be fully understood. To overcome these limitations, a monitoring system capable  
93 of continuously collecting data throughout the PBL is necessary.

94 Recent developments in Light Detection and Ranging (LiDAR) technology have enabled  
95 accurate remote sensing of the vertical structure of PBL at a high spatiotemporal resolution,  
96 representing a substantial improvement over traditional ground-level measurements (Chen et al.,  
97 2023). By detecting the Doppler shift of laser signals, wind LiDAR can detect vertical distribution  
98 of wind speed and direction (Yang et al., 2022). In recent years, there has been a growing adoption  
99 of LiDAR technologies in air quality studies. For instance, Wang et al. (2019) applied a wind  
100 LiDAR system to monitor the vertical variation in wind velocity and the evolution of the urban  
101 boundary layer during a pollution episode in Beijing. Similarly, Park et al. (2022) employed wind  
102 LiDAR measurements to investigate the variations in the PBL height at an urban site in Seoul,  
103 Korea.

104 In this study, measurements from a wind Light Detection and Ranging (LiDAR) system  
105 were taken to monitor vertical profile of wind pattern at an urban site in Hong Kong in September  
106 2022, a period when the city was frequently impacted by tropical cyclones and experienced severe

107 O<sub>3</sub> pollution levels. The PBL height was identified using the vertical profile of wind shear. By  
108 combining information on the PBL height, vertical wind profile, and O<sub>3</sub> concentration, we  
109 explored the horizontal flux of O<sub>3</sub> across the PBL in Hong Kong. The vertical variation in the O<sub>3</sub>  
110 flux was evaluated. The dependence of the horizontal flux of O<sub>3</sub> on the wind direction during the  
111 entire study period and the pollution episodes was analyzed. Cross-sectional analyses of the O<sub>3</sub>  
112 flux can help to provide answers to questions such as how many pollutants were transported from  
113 the central GBA into Hong Kong. Overall, this study aims to enhance our understanding of the 3D  
114 pollutant transports for both long-term averages and short-term pollution episodes in the GBA and  
115 Hong Kong.

## 116 **2. Data and methodology**

### 117 **2.1 Wind LiDAR measurements**

118 The wind LiDAR system, model WINDCUBE 100S, was produced by Leosphere, a  
119 Vaisala company. As shown in Figure S1, it was located at the Hong Kong Observatory (HKO)  
120 situated at King's Park in Hong Kong (22.3132°N, 114.1704°E), and operated in the Doppler  
121 Beam Swinging (DBS) mode. This mode detects the Doppler shifts of infrared laser signals that  
122 are backscattered by aerosols in the atmosphere. Measurements of hourly horizontal wind speed  
123 (W) and direction ( $\theta$ ) were taken from 50 m to 3.1 km above ground level (AGL), with a vertical  
124 resolution of 25 m. For further information on the algorithm used and the uncertainty of the wind  
125 LiDAR measurements, please refer to He et al. (2021). As shown in Figure S2, the quantity of  
126 valid samples decreases as the height increases, owing to the presence of clouds and the lack of  
127 aerosols in the free atmosphere. Nevertheless, the majority of wind data, specifically below 1 km,  
128 remains available.

### 129 **2.2 Meteorological and O<sub>3</sub> data**

130 To assess the accuracy of the wind LiDAR measurements during the study period, ground-  
131 level and upper-level wind data were obtained from collocated sources. Hourly ground-level wind  
132 data were obtained from the Automatic Weather Station (AWS) at King's Park, while upper-level  
133 wind data were collected through radiosonde measurements conducted at King's Park. The  
134 radiosonde measurements were available at 8:00 am and 20:00 pm (China's local time in UTC+8  
135 was used in this study) throughout the study period. In addition, hourly data on O<sub>3</sub> concentrations  
136 at Sham Shui Po (22.3315°N, 114.1567°E), which is situated adjacent to King's Park, were used  
137 to explore the O<sub>3</sub> flux. To gain a better understanding of the vertical variation in O<sub>3</sub> concentration  
138 in the PBL, we obtained the O<sub>3</sub> data at Tai Mo Shan (22.4102°N, 114.1245°E), which is situated  
139 at an elevation of 950 meters above sea level.

### 140 **2.3 PBL height**

141 The height of PBL can be identified by analyzing various profiles, such as vertical velocity  
142 variance, wind speed shear, and wind directional shear (Tucker et al., 2009). As a result of surface  
143 friction, winds within the PBL are typically weaker than those in the free atmosphere. In the layer  
144 above the PBL, the wind speed becomes more uniform as the effect of friction decreases  
145 significantly. Large wind shear can be present at the top of the PBL (Canut et al., 2012; Lindvall  
146 and Svensson, 2019). Therefore, the PBL height can be defined as the altitude of a transition layer  
147 where there is a marked change in wind behavior.

148 The identification of the PBL height in this study is based on the vertical profile of wind  
149 speed shear. To be specific, the gradient of the horizontal wind speed is estimated as a function of  
150 height in order to determine the vertical distribution of wind speed shear (unit: s<sup>-1</sup>):

$$151 \quad \eta = \frac{W(z_{i+1}) - W(z_i)}{z_{i+1} - z_i} \quad (1)$$

152 where  $z$  denotes the altitude (unit: m);  $z_{i+1} - z_i$  represents the vertical resolution of the LiDAR  
 153 measurement, which is 25 m; and  $W$  represents the horizontal wind speed (unit: m/s). Then, the  
 154 PBL height can be determined by identifying the height at which the gradient of the horizontal  
 155 wind speed reaches its maximum value with respect to altitude. It should be noted, however, that  
 156 the wind shear can also be significant near the ground. For this reason, the wind data collected  
 157 below 100 m near the ground were excluded from the PBL estimation process to ensure the  
 158 accuracy of the results.

#### 159 **2.4 Horizontal flux of O<sub>3</sub>**

160 Horizontal flux of O<sub>3</sub> was analyzed based on the wind LiDAR measurements to gain  
 161 insights into O<sub>3</sub> transport in the study region. The flux represents the mass flow per unit cross-  
 162 sectional area per unit time (unit: g/(m<sup>2</sup>·s)):

$$163 \quad f = c \cdot W \quad (2)$$

164 where  $c$  denotes the O<sub>3</sub> concentration (unit: μg/m<sup>3</sup>). The total horizontal flux (THF) of O<sub>3</sub> through  
 165 a cross-sectional area of 1 m<sup>2</sup> during the study period at a given direction  $\theta$  and altitude  $z$  can be  
 166 estimated as follows (unit: g/m<sup>2</sup>):

$$167 \quad THF(\theta, z) = \sum_{t=1}^N f(\theta, z, t) \cdot \Delta t = \sum_{t=1}^N c(z, t) \cdot W(\theta, z, t) \cdot \Delta t \quad (3)$$

168 where  $\theta$  is the direction;  $t$  represents time; and  $\Delta t$  (which is 1 hour) indicates the temporal  
 169 resolution of LiDAR measurements. For the purposes of this study, it was assumed that O<sub>3</sub> within  
 170 the PBL was well mixed. Consequently, the temporal variation in O<sub>3</sub> concentration was taken into  
 171 account. The dependence of the THF of O<sub>3</sub> on both direction ( $\theta$ ) and altitude ( $z$ ) was analyzed to  
 172 improve our understanding of the 3D transport of O<sub>3</sub> over Hong Kong.

173 The analysis then proceeded to examine the THF of O<sub>3</sub> through the entire PBL (Figure 1).

174 At a given direction  $\theta$ , the THF of O<sub>3</sub> through a cross-sectional area with a width of 1 m across the

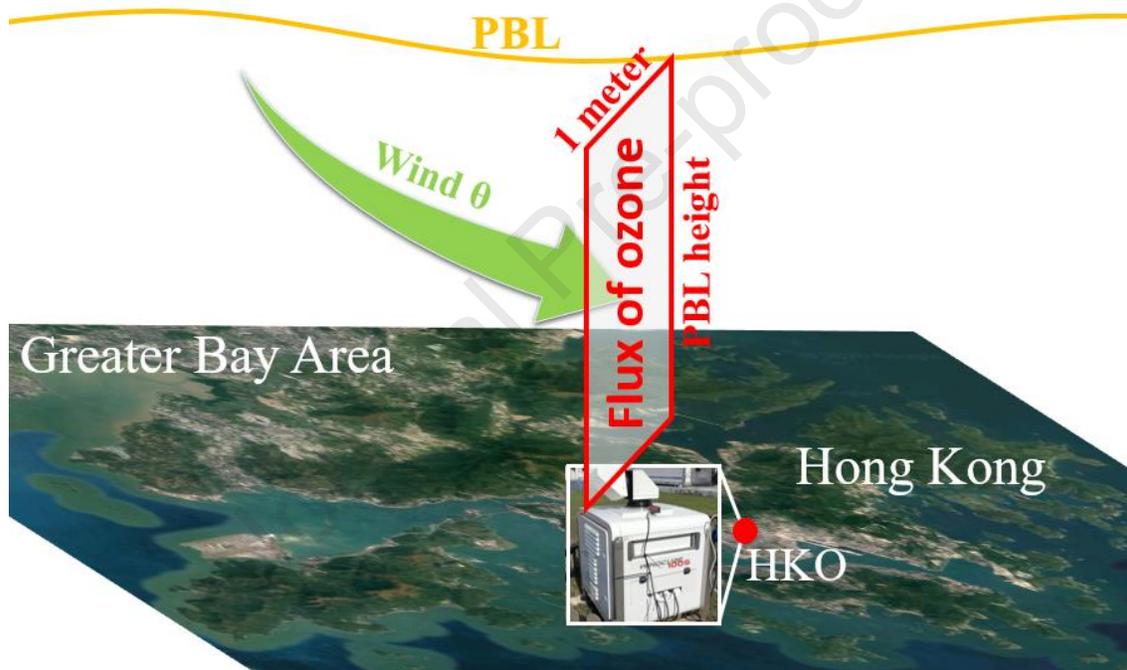
175 entire PBL during the study period can be estimated as follows (unit: kg):

$$176 \quad THF(\theta)_{PBL} = \sum_{t=1}^N \sum_{z=1}^{H(t)} c(z, t) \cdot W(\theta, z, t) \cdot \Delta z \cdot 1m \cdot \Delta t \quad (4)$$

177 where  $\Delta z$  (which is 25 m) represents the vertical resolution of LiDAR measurements. The

178 dependence of the THF of O<sub>3</sub> across the entire PBL on direction ( $\theta$ ) was then analyzed to help

179 understand the O<sub>3</sub> transport in the study region.



180

181 **Figure 1.** Experimental setup for estimating the total horizontal flux (THF) of O<sub>3</sub> through a cross-  
182 sectional area across the PBL in Hong Kong.

### 183 3. Results

#### 184 3.1 O<sub>3</sub> variation

185 Time series of O<sub>3</sub> concentration at Sham Shui Po in September 2022 is presented in Figure

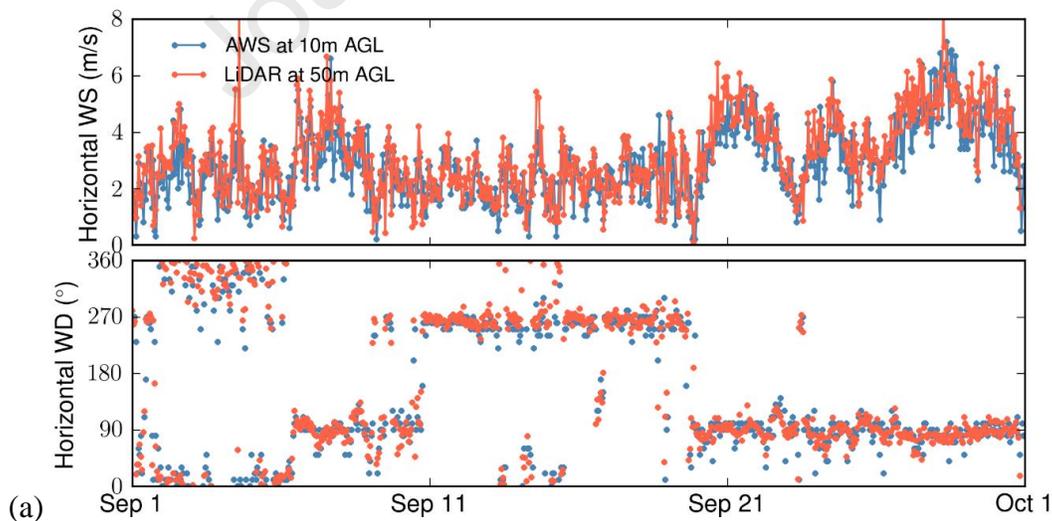
186 S3. Based on the monthly average, the O<sub>3</sub> concentration is estimated to be  $101.47 \pm 51.98 \mu\text{g}/\text{m}^3$ .

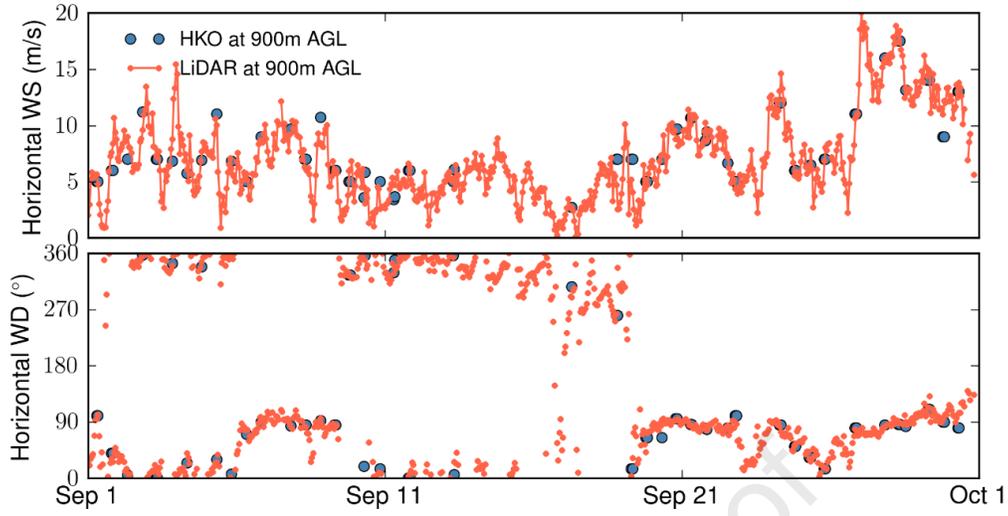
187 The grey line in the figure marks the threshold of  $160 \mu\text{g}/\text{m}^3$ , which was used to identify O<sub>3</sub>

188 pollution episodes in this study. During the study period, there were frequent O<sub>3</sub> pollution episodes,  
 189 and the maximum concentration of over 300 µg/m<sup>3</sup> was recorded on September 13. The hourly O<sub>3</sub>  
 190 concentration exceeded the threshold for 78 hours, which is equivalent to 10.8% of the study month.

### 191 3.2 Wind variation

192 Ground-level and upper-level wind data were used to assess the accuracy of the wind  
 193 LiDAR measurements during the study period. Panel (a) of Figure 2 shows time series of wind  
 194 speed and direction at 50 m AGL from LiDAR measurements and at 10 m AGL from ground-  
 195 based AWS measurements in September 2022. The wind LiDAR measurements were found to be  
 196 in good agreement with the ground-based measurements. The average wind speed at 10 m AGL  
 197 was slightly lower than that at 50 m AGL. Based on monthly averages, the estimated wind speed  
 198 at 10 m AGL and 50 m AGL at King's Park were  $2.9 \pm 1.3$  m/s and  $3.2 \pm 1.4$  m/s, respectively.  
 199 Panel (b) shows the time series of wind speed and direction at 900 m AGL as measured by the  
 200 wind LiDAR and radiosonde instruments. Once again, the results showed good agreement between  
 201 the LiDAR and sounding measurements.



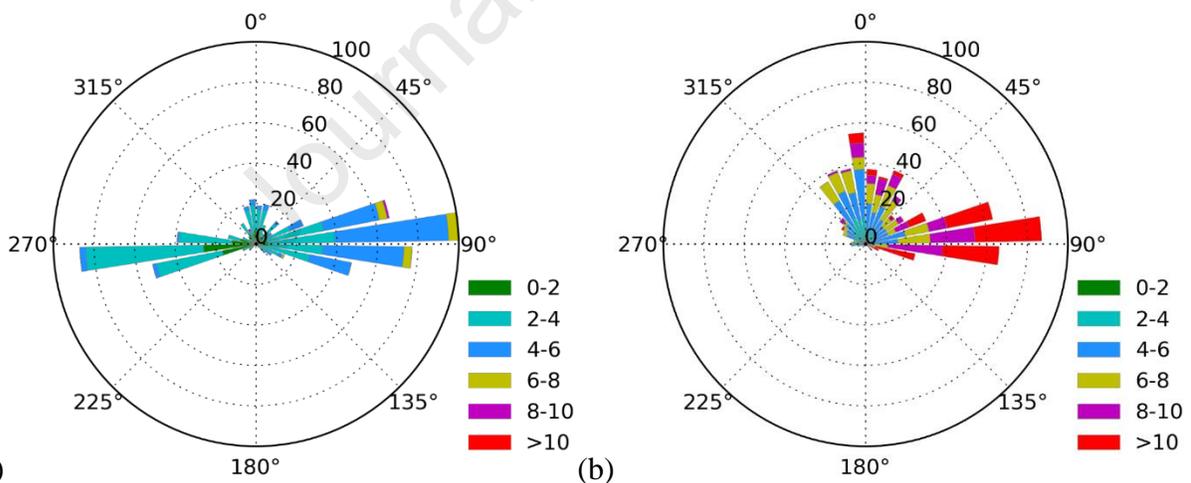


203 (b) Sep 1 Sep 11 Sep 21 Oct 1

204 **Figure 2.** (a) Time series of wind speed and direction at 50 m AGL from LiDAR measurements  
 205 and at 10 m AGL from ground-based AWS measurements in September 2022. (b) Time series of  
 206 wind speed and direction at 900 m AGL obtained from LiDAR and radiosonde measurements in  
 207 September 2022.

208 Since wind is a vector, two components were evaluated separately: the east-west (U) and  
 209 north-south (V) wind speeds. Positive U and V wind speeds correspond to westerly and southerly  
 210 winds, respectively, while negative U and V wind speeds correspond to easterly and northerly  
 211 winds, respectively. Panels (a) and (b) of Figure S4 present a comparison of the U and V wind  
 212 velocities at 50 m AGL as measured by the wind LiDAR and at 10 m AGL by the ground-based  
 213 AWS monitoring at King's Park during the study month. The comparison revealed good agreement,  
 214 with correlation coefficients of 0.94 and 0.71 (N = 698) for the U and V wind speeds, respectively.  
 215 Panels (c) and (d) depict a comparison of the U and V wind speeds at 900 m AGL as measured by  
 216 the wind LiDAR and radiosonde instruments at King's Park. The comparison showed good  
 217 agreement, with correlation coefficients of 0.98 and 0.88 (N = 68) for the U and V wind speeds,  
 218 respectively.

219 The time-series evaluations indicated that wind behavior varied with altitude. Figure 3  
 220 compares the wind roses at 50 m and 900 m AGL as measured by the wind LiDAR in September  
 221 2022. At 50 m AGL, the winds were mainly composed of easterly and westerly winds. As the  
 222 altitude increased, the westerly components shifted to northerly and northwesterly. The clockwise  
 223 veering winds from the bottom to the top of the PBL can be attributed to the Ekman spiral, resulting  
 224 from the decrease in friction as a significant force with increasing height. The winds are  
 225 geostrophic in the free atmosphere. As a consequence, wind direction is parallel to the isobars.  
 226 When a tropical cyclone is located to the east of Hong Kong, the dominant wind in the free  
 227 atmosphere becomes northerly. However, within the PBL, friction causes air to spiral into low  
 228 pressure areas since it reduces the magnitude of the Coriolis force. Therefore, when the tropical  
 229 cyclone is located to the east of Hong Kong, the dominant winds near the ground turn westerly or  
 230 northwesterly.

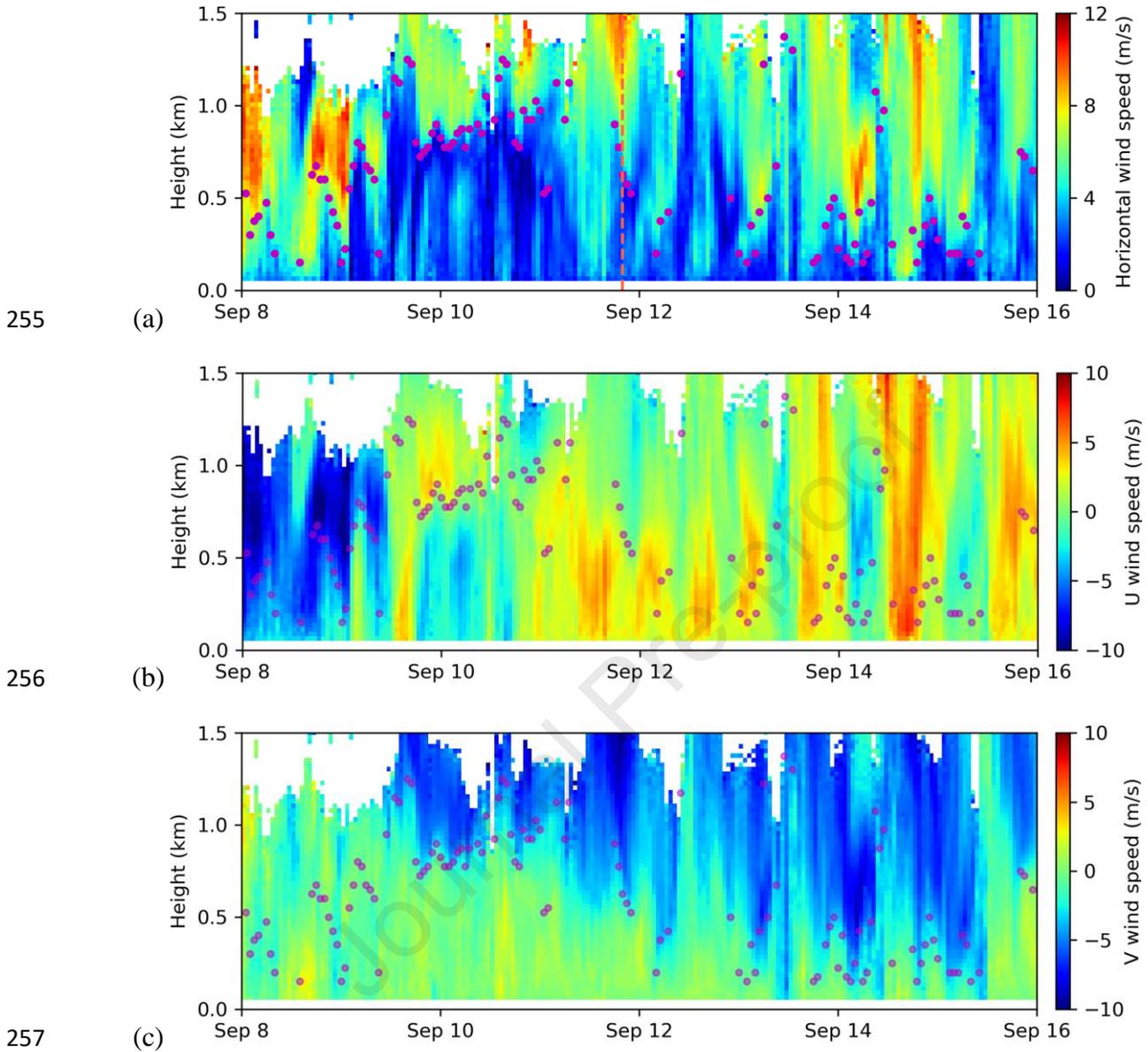


231 (a) 232 **Figure 3.** Wind roses at (a) 50 m and (b) 900 m AGL as measured by the wind LiDAR in  
 233 September 2022.

### 234 3.3 PBL height

235           The vertical profile of wind speed shear was used to identify the PBL height. An example  
236 is provided in Figure S5. Panel (a) of it shows the vertical distribution of horizontal wind speed  
237 (W) at 20:00 pm on September 11, 2022. Due to surface friction, wind speeds within the PBL are  
238 usually lower than those in the free atmosphere. The corresponding vertical profile of the wind  
239 speed shear is presented in panel (b). The PBL height can be determined by identifying the height  
240 at which the gradient of the horizontal wind speed reaches its maximum value with respect to  
241 altitude. Based on this criterion, the PBL height was estimated to be 0.62 km.

242           Panel (a) of Figure 4 shows the time series of the PBL height, along with the vertical  
243 distribution of horizontal wind speed (W) from September 8 to 15, 2022. The corresponding  
244 vertical distributions of U and V wind speeds are displayed in panels (b) and (c), respectively. As  
245 depicted in Figure S6, the tropical cyclone "Muifa" moved towards the Asian continent and was  
246 situated near Taiwan on September 12. When Hong Kong was impacted by the air subsidence at  
247 the outer edge of the tropical cyclone, the atmosphere became more stable, and the PBL height  
248 dropped. As a result, the PBL height decreased from around 1 km on September 10 to  
249 approximately 200 m on September 14. The suppressed PBL can hinder the vertical dispersion of  
250 air pollutants. In addition, when the tropical cyclone affected Hong Kong, the dominant winds  
251 changed from easterly to westerly. These westerly winds transported air pollutants from the central  
252 GBA into Hong Kong, thereby contributing to a rapid increase in pollutant concentrations in Hong  
253 Kong. Moreover, strong northerly geostrophic winds were present above the PBL due to the  
254 balance between the pressure gradient force and Coriolis effect.



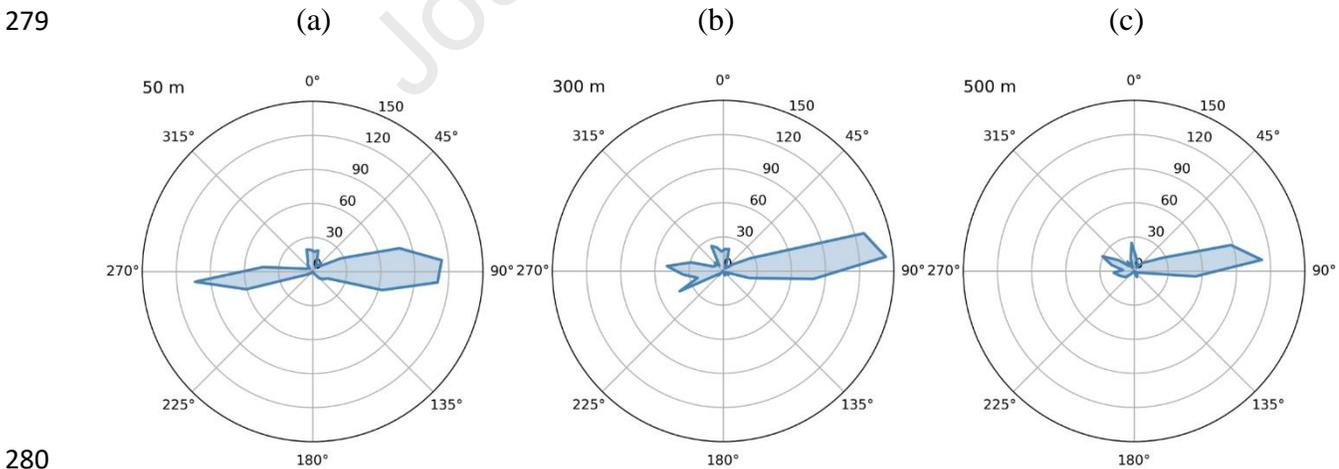
258 **Figure 4.** Time series of the PBL height (purple dots), along with the vertical distributions of (a)  
 259 horizontal wind speed, (b) U wind speed, and (c) V wind speed from September 8 to 15, 2022. The  
 260 red dashed line marks the example data used in Figure S5.

261 Based on the estimated PBL throughout the study month, Figure S7 shows the diurnal  
 262 variation in the monthly average PBL height, along with the vertical distribution of horizontal  
 263 wind speed in September 2022. During the study month, the average PBL height was  $0.56 \pm 0.34$   
 264 km. A significant diurnal variation was found, with the PBL height increasing in the morning and

265 reaching its peak at noontime and early afternoon (e.g.,  $0.74 \pm 0.38$  km at 12:00 pm). This diurnal  
 266 pattern is attributable to the variation in solar heating throughout the day, which, in turn, affects  
 267 the convection of the atmosphere.

### 268 3.4 Vertical variance in O<sub>3</sub> flux

269 By combining information on the PBL height, vertical wind profile, and O<sub>3</sub> concentration,  
 270 we explored the vertical variance in the THF of O<sub>3</sub> across the PBL. Figure 5 shows the THF of O<sub>3</sub>  
 271 through a unit cross-sectional area for various directions, with a resolution of 10°, at three altitudes  
 272 during the entire month of September 2022. Throughout the study month, the THF of O<sub>3</sub> exhibited  
 273 a predominant easterly component at all three altitudes. Table 1 summarizes the THF of O<sub>3</sub> through  
 274 a unit cross-sectional area for six primary groups of directions: 0-60°, 60-120°, 120-180°, 180-  
 275 240°, 240-300°, and 300-360°. The easterly winds between 60° and 120° contributed 50.8%,  
 276 52.3%, and 52.5% to the total flux of O<sub>3</sub> at 50 m, 300 m, and 500 m, respectively. The predominant  
 277 easterly component of O<sub>3</sub> flux can be attributed to the dominant easterly wind pattern during the  
 278 study month.



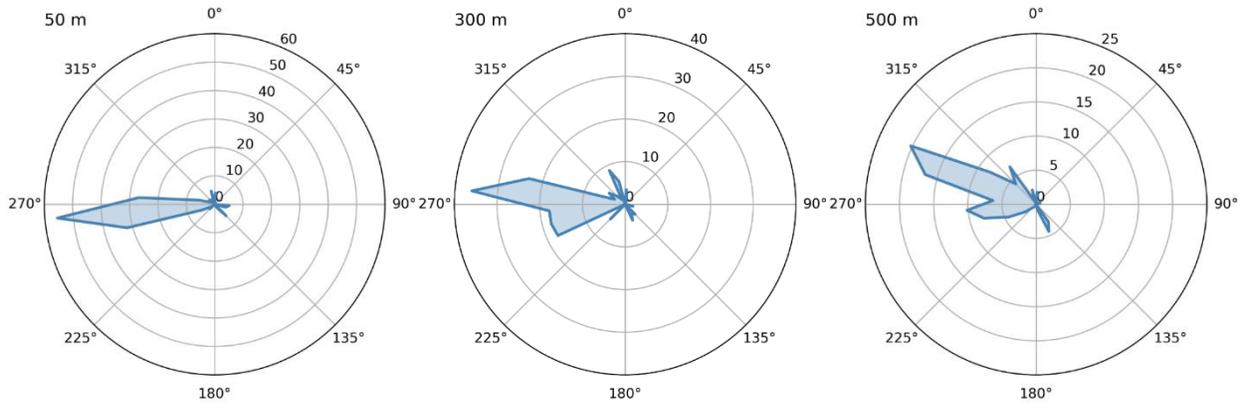
280  
 281 **Figure 5.** The THF of O<sub>3</sub> through a unit cross-sectional area for various directions at (a) 50 m, (b)  
 282 300 m, and (c) 500m AGL during the entire month of September 2022 (unit: g/m<sup>2</sup>).

283 **Table 1.** The THF of O<sub>3</sub> through a unit cross-sectional area for six primary groups of directions at  
 284 50 m, 300 m, and 500 m AGL during the entire study month. The percentage contributions of each  
 285 component are indicated in parentheses.

Altitudes	THF of O <sub>3</sub> through a unit cross-sectional area (unit: g/m <sup>2</sup> )					
	0-60°	60-120°	120-180°	180-240°	240-300°	300-360°
50 m	67.93 (8.4%)	408.92 (50.8%)	23.32 (2.9%)	6.54 (0.8%)	231.75 (28.8%)	65.83 (8.2%)
300 m	63.26 (8.2%)	404.18 (52.3%)	18.05 (2.3%)	11.15 (1.4%)	188.66 (24.4%)	87.25 (11.3%)
500 m	59.20 (10.6%)	291.82 (52.5%)	13.90 (2.5%)	14.90 (2.7%)	104.28 (18.7%)	72.18 (13.0%)

286  
 287 Similar analyses were performed to investigate the horizontal flux of O<sub>3</sub> during the O<sub>3</sub>  
 288 pollution episodes. Figure 6 shows the THF of O<sub>3</sub> through a unit cross-sectional area for various  
 289 directions, with a resolution of 10°, at three altitudes during the O<sub>3</sub> pollution episodes in September  
 290 2022. During the O<sub>3</sub> pollution episodes, the THF of O<sub>3</sub> exhibited a predominant westerly  
 291 component at all three altitudes. Table 2 summarizes the THF of O<sub>3</sub> through a unit cross-sectional  
 292 area for six primary groups of directions. The westerly winds between 240° and 300° contributed  
 293 78.1%, 71.9%, and 64.7% to the total flux of O<sub>3</sub> at 50 m, 300 m, and 500 m, respectively. The  
 294 predominant westerly component of O<sub>3</sub> flux can be attributed to the change in the dominant wind  
 295 pattern when the tropical cyclone affected Hong Kong.

296 (a) (b) (c)



297

298 **Figure 6.** The THF of O<sub>3</sub> through a unit cross-sectional area for various directions at (a) 50 m, (b)  
 299 300 m, and (c) 500m AGL during the O<sub>3</sub> pollution episodes in September 2022 (unit: g/m<sup>2</sup>).

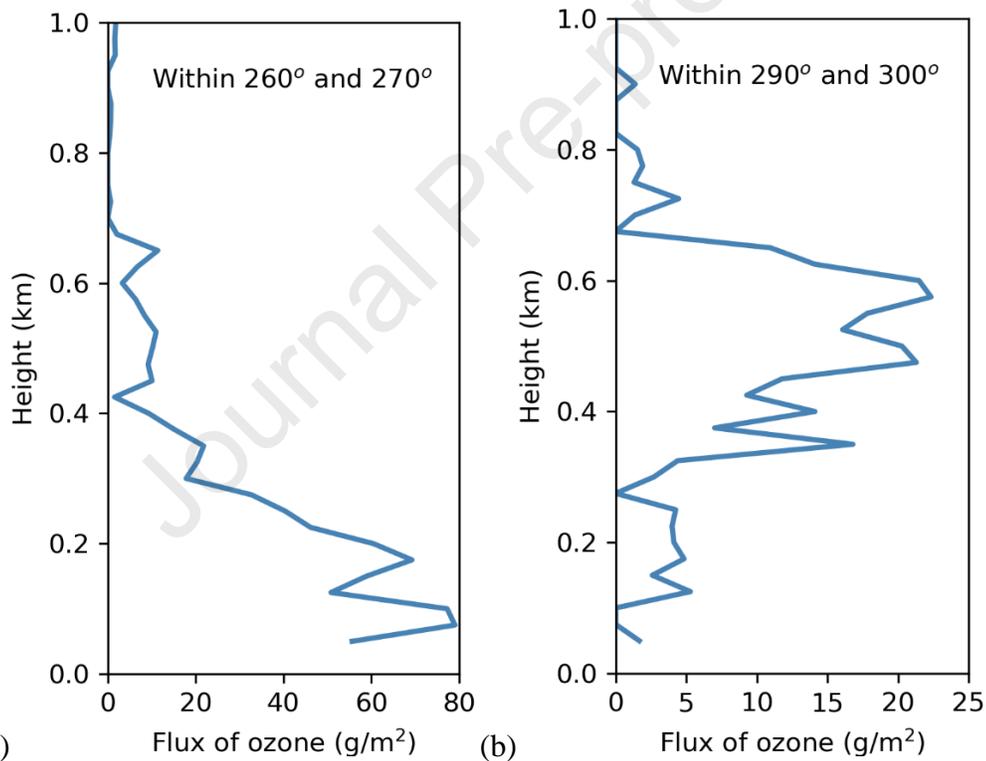
300 **Table 2.** The THF of O<sub>3</sub> through a unit cross-sectional area for six primary groups of directions at  
 301 50 m, 300 m, and 500 m AGL during the O<sub>3</sub> pollution episodes in September 2022. The percentage  
 302 contributions of each component are indicated in parentheses.

Altitudes	THF of O <sub>3</sub> through a unit cross-sectional area (unit: g/m <sup>2</sup> )					
	0-60°	60-120°	120-180°	180-240°	240-300°	300-360°
50 m	3.90 (2.4%)	9.22 (5.7%)	7.48 (4.6%)	4.17 (2.6%)	126.33 (78.1%)	10.63 (6.6%)
300 m	4.77 (3.0%)	1.87 (1.2%)	10.16 (6.3%)	5.22 (3.3%)	115.14 (71.9%)	22.96 (14.3%)
500 m	2.08 (2.0%)	0.61 (0.6%)	8.41 (8.3%)	3.10 (3.0%)	65.89 (64.7%)	21.81 (21.4%)

303

304 It is worth noting that, as the height increased in the PBL, the peak O<sub>3</sub> flux shifted from  
 305 westerly (260-270°) at 50 m to northwesterly (290-300°) at 500 m. This rotation can be attributed

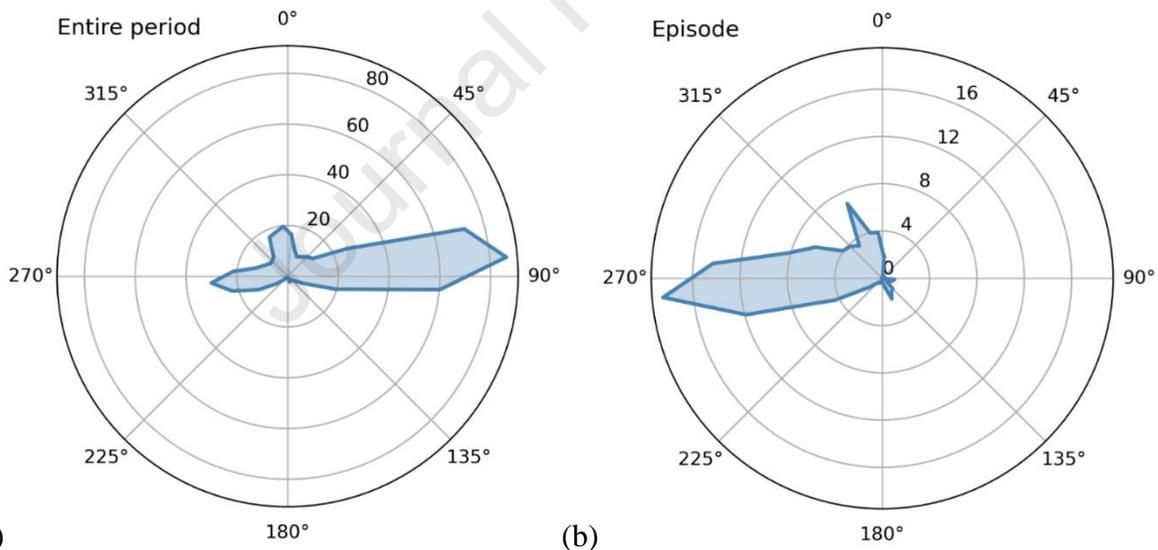
306 to the veering wind from the ground to the top of boundary layer. To gain further insights into the  
 307 vertical variation in the  $O_3$  flux, Figure 7 compares the vertical distribution of the THF of  $O_3$   
 308 through a unit cross-sectional area for the directions of (a)  $260\text{-}270^\circ$  and (b)  $290\text{-}300^\circ$  during the  
 309  $O_3$  pollution episodes in September 2022. For the directions between  $260^\circ$  and  $270^\circ$ , the THF of  
 310  $O_3$  shows a maximum near the ground, reaching a level of approximately  $80\text{ g/m}^2$  during the  $O_3$   
 311 pollution episodes. However, for the directions between  $290^\circ$  and  $300^\circ$ , the THF of  $O_3$  increased  
 312 with altitude within the PBL, with the total  $O_3$  flux reaching a peak exceeding  $20\text{ g/m}^2$  at  $600\text{ m}$   
 313 AGL.



314 (a) (b)  
 315 **Figure 7.** Vertical distributions of the THF of  $O_3$  through a unit cross-sectional area for the  
 316 directions of (a)  $260\text{-}270^\circ$  and (b)  $290\text{-}300^\circ$  during the  $O_3$  pollution episodes in September 2022  
 317 (unit:  $\text{g/m}^2$ ).

### 318 3.5 $O_3$ flux across the PBL

319 In the last section, the THF of O<sub>3</sub> across the entire PBL was analyzed. Panel (a) of Figure  
 320 8 shows the THF of O<sub>3</sub> through an area with a width of 1 m across the entire PBL for various  
 321 directions during the entire study month. For the entire month, the THF of O<sub>3</sub> across the PBL  
 322 exhibited a predominant easterly component. As summarized in Table 3, the easterly winds  
 323 between 60° and 120° contributed 49.3% to the total flux of O<sub>3</sub> in the PBL. Panel (b) shows the  
 324 THF of O<sub>3</sub> through an area with a width of 1 m across the PBL for various directions during the  
 325 O<sub>3</sub> pollution episodes. During the episodes, the THF of O<sub>3</sub> across the PBL exhibited a predominant  
 326 westerly component. The westerly winds within 240° and 300° contributed 61.2% to the total flux  
 327 of O<sub>3</sub> in the PBL. These results indicate that during the pollution episodes, there can be substantial  
 328 differences in O<sub>3</sub> fluxes compared to the average conditions. These analyses enhanced our  
 329 understanding of both the short-term variations and long-term average levels of air pollution.



330 (a) (b)  
 331 **Figure 8.** The THF of O<sub>3</sub> through an area with a width of 1 m across the entire PBL for various  
 332 directions during (a) the entire study month and (b) O<sub>3</sub> pollution episodes in September 2022 (unit:  
 333 kg).

334 **Table 3.** The THF of O<sub>3</sub> through an area with a width of 1 m across the entire PBL for six primary  
 335 groups of directions during the entire study month and the O<sub>3</sub> pollution episodes in September

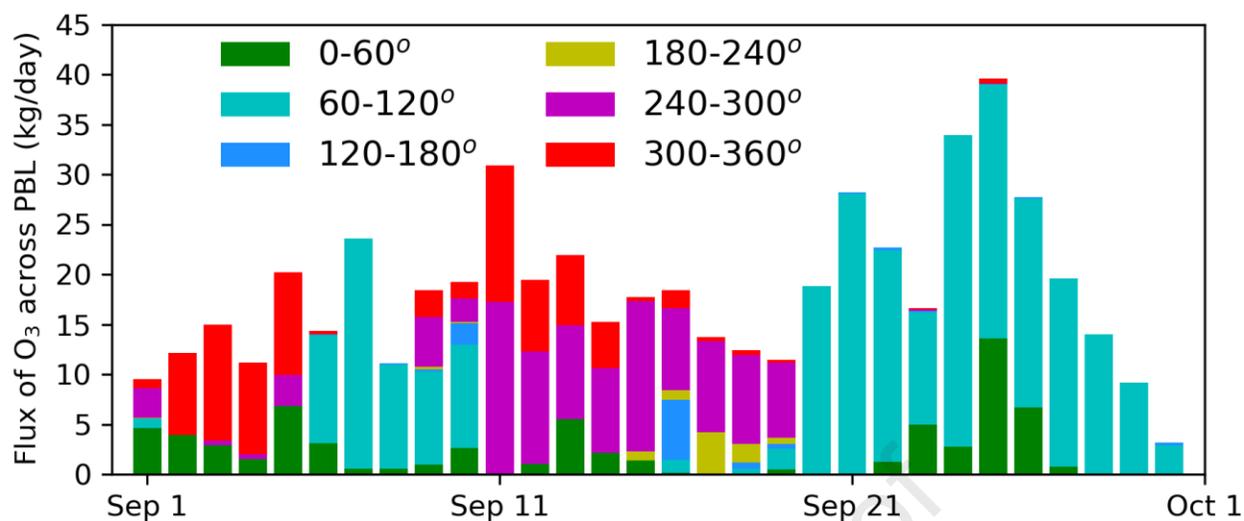
336 2022. Results for daytime (between 8:00 am and 20:00 pm) and nighttime (before 8:00 am or after  
 337 20:00 pm) are also presented. The percentage contributions of each component are indicated in  
 338 parentheses.

Periods	THF of O <sub>3</sub> across the entire PBL for various directions (unit: kg)						
	0-60°	60-120°	120-180°	180-240°	240-300°	300-360°	All
Entire	68.26 (12.4%)	270.74 (49.3%)	10.65 (1.9%)	8.88 (1.6%)	109.60 (19.9%)	81.31 (14.8%)	549.4
Entire - night	23.71 (9.8%)	149.28 (62.0%)	0.90 (0.38%)	5.78 (2.40%)	34.96 (14.5%)	26.10 (10.8%)	240.7
Entire - day	44.56 (14.4%)	121.47 (39.3%)	9.75 (3.2%)	3.10 (1.0%)	74.64 (24.2%)	55.22 (17.9%)	308.7
Episodes	2.27 (2.2%)	2.94 (2.8%)	6.08 (5.8%)	3.03 (2.9%)	63.64 (61.2%)	26.05 (25.0%)	104.0
Episodes - night	0.03 (0.2%)	1.42 (11.8%)	0.33 (2.8%)	1.32 (11.0%)	8.55 (70.9%)	0.41 (3.4%)	12.1
Episodes - day	2.24 (2.4%)	1.52 (1.7%)	5.74 (6.2%)	1.72 (1.9%)	55.09 (59.9%)	25.64 (27.9%)	91.9

339  
 340 Results for the THF of O<sub>3</sub> in daytime (between 8:00 am and 20:00 pm) and nighttime  
 341 (before 8:00 am or after 20:00 pm) are also presented in Table 3. Throughout the entire month, the  
 342 THF of O<sub>3</sub> across the PBL in nighttime was approximately 20% lower than that in daytime. For

343 both the daytime and nighttime throughout the month, the THF of O<sub>3</sub> across the PBL showed a  
344 predominant easterly component. During the O<sub>3</sub> episodes, the THF of O<sub>3</sub> across the PBL in  
345 daytime was substantially higher than that in the nighttime. These results indicate enhanced  
346 photochemical formation during the daytime episodes. For both the daytime and nighttime  
347 episodes, the THF of O<sub>3</sub> across the PBL exhibited a predominant westerly component. The  
348 westerly winds within 240° and 300° contributed 70.9% and 59.9% to the total flux of O<sub>3</sub> in the  
349 PBL for nighttime and daytime, respectively.

350         Figure 9 shows the daily variation in the THF of O<sub>3</sub> through an area with a width of 1 m  
351 across the entire PBL for six primary groups of directions in September 2022. The impacts of the  
352 wind from different directions greatly varied. In early September, the northwesterly winds between  
353 300° and 360° played a dominant role in the O<sub>3</sub> transport in Hong Kong. From September 6 to 10,  
354 the easterly winds between 60° and 120° governed the O<sub>3</sub> transport. From September 11 to 19, the  
355 westerly winds between 240° and 300° played a dominant role in the O<sub>3</sub> transport. After September  
356 20, the O<sub>3</sub> transport was determined by easterly winds between 60° and 120°. As shown in Figure  
357 S8, we separated the daily variations in the THF of O<sub>3</sub> across the PBL into nighttime and daytime.  
358 For the non-episode periods, the O<sub>3</sub> fluxes in daytime and nighttime were comparable. However,  
359 during the O<sub>3</sub> episodes, the O<sub>3</sub> flux greatly increased during the daytime due to enhanced  
360 photochemical formation of O<sub>3</sub>. These results demonstrate that the source of O<sub>3</sub> transported over  
361 Hong Kong varied significantly from day to day. The association between the wind conditions and  
362 O<sub>3</sub> transport is apparent.



363  
364 **Figure 9.** Daily variations in the THF of O<sub>3</sub> through an area with a width of 1 m across the entire  
365 PBL for six primary groups of directions in September 2022.

#### 366 4. Discussion

367 Ground-level air quality monitoring provides pollutant concentrations at specific locations.  
368 However, cross-sectional analyses are necessary to understand pollutant transports across different  
369 regions. Analyzing the O<sub>3</sub> flux can help provide answers to questions such as how many pollutants  
370 were transported from a specific region (e.g., the central GBA area) into Hong Kong. In the cross-  
371 sectional analyses, information on the PBL height is essential because most air pollutants are  
372 mixed within the PBL. As a capping inversion layer forms between the PBL and the free  
373 atmosphere, we can assume that the interaction between the PBL and the free atmosphere is limited.  
374 In this study, cross-sectional analyses were performed at a specific location. However, with more  
375 LiDAR systems available at different locations, it is possible to analyze the pollutant flux across  
376 the entire boundary between two cities.

377 This study identified significant impacts of weather systems on O<sub>3</sub> transport. Synoptic  
378 patterns featuring a tropical cyclone were found to be conducive to the occurrence of air pollution  
379 episodes in Hong Kong. Other synoptic patterns, such as a high pressure system located to the

380 north or a trough located to the south, can significantly increase O<sub>3</sub> concentrations in Hong Kong  
381 (Lin et al., 2021). Among these weather patterns, the pollutant flux over Hong Kong can vary  
382 significantly. It is worthwhile to extend our study to cover various O<sub>3</sub> episodes under different  
383 synoptic patterns over a more extended period. Based on the long-term dataset, the main features  
384 of the pollutant transports under different synoptic patterns can be compared.

385 This study identified significant vertical variance in the wind pattern from the ground up  
386 to the top of the PBL. For instance, when the ground-level winds were westerly, the winds at the  
387 upper PBL could shift to northwesterly. The veering winds from the bottom to the top of the PBL  
388 (i.e., Ekman spiral) can be attributed to the decrease in friction as the altitude increases. In the free  
389 atmosphere, friction has a negligible effect, and the wind becomes geostrophic, following the  
390 isobars. In the PBL, however, friction causes air to spiral into low-pressure areas. Given the  
391 significant vertical variation in the wind pattern, the pollution transports can vary greatly at  
392 different altitudes in the PBL. This underscores the importance of using LiDAR techniques to  
393 improve our understanding of the 3D transport of air pollutants.

394 Control of O<sub>3</sub> pollution is a complicated and slow process due to the non-linear response  
395 of O<sub>3</sub> to its precursors. Since the implementation of clean air plan in 2013, the O<sub>3</sub> pollution has  
396 become a prominent environmental issue in China. Therefore, the transport of O<sub>3</sub> across the GBA  
397 and Hong Kong was analyzed in this study. In the GBA and Hong Kong, the annual concentration  
398 levels of other pollutants, such as fine particulate matter, are still much higher than the air quality  
399 guidelines as recommended by the World Health Organization. Future studies can apply the cross-  
400 sectional analyses to understand the transport of other air pollutants, such as particulate matter.

401 LiDAR measurements provide a unique opportunity to identify the variation in the PBL  
402 height. Previous studies have employed various algorithms to estimate the PBL height. In this

403 study, we adopted the method based on the vertical profile of wind speed shear. Reasonable results  
404 were obtained, as indicated by the decreased PBL height due to the effect of subsidence at the  
405 outer edge of a tropical cyclone. In addition, the PBL height reached its maximum at noontime.  
406 These results are consistent with our understanding of the typical variation in the PBL height. This  
407 method has some limitations. For example, super-geostrophic phenomena may occur in the  
408 nighttime boundary layer (Wang et al., 2023). As a result, wind speeds in the boundary layer can  
409 surpass those in the free atmosphere. When the super-geostrophic phenomena occur, our algorithm  
410 cannot estimate the PBL height. In such cases, the PBL height will be filled by interpolating  
411 adjacent values. In the future, a comprehensive evaluation of the PBL height can be conducted by  
412 comparing it with other datasets, such as aerosol LiDAR and meteorological sounding data.

413         The wind fields in Hong Kong are influenced by complex terrain and architectural features.  
414 Firstly, the increased roughness of the urban surface can amplify friction, thereby augmenting the  
415 Ekman spiral. Consequently, this can lead to an increased rotation in the wind pattern in relation  
416 to altitude. Secondly, the complex architectural features in urban areas increase the thermal  
417 contrast between the land and ocean in Hong Kong. Consequently, this enhancement can  
418 strengthen the land-sea breeze circulation, which is a ubiquitous mesoscale meteorological  
419 phenomenon occurring along the coastal regions of Hong Kong. The complex wind fields, in turn,  
420 impact the transport of pollutants in Hong Kong. In this study, wind LiDAR measurements were  
421 taken at a single station located in an urban area. The presence of complex wind fields emphasizes  
422 the significance of utilizing LiDAR measurements from multiple stations across the territory to  
423 track the 3D transport of air pollutants.

424         Uncertainty in estimating the  $O_3$  flux is determined by several factors, including the PBL  
425 height and the vertical profiles of wind and pollutant concentration. Firstly, the PBL height was

426 estimated using wind LiDAR measurements taken on King's Park Hill, which has an elevation of  
427 90 m. The estimation results are slightly lower than those of some other studies, which reported  
428 daytime mixing layer heights ranging from 0.6 km to 1.1 km in Hong Kong (Yang et al., 2013).  
429 Secondly, uncertainties exist in the wind LiDAR measurements of the upper-level winds. In this  
430 study, collocated ground measurements and upper-air radiosonde measurements were used to  
431 evaluate the performance of the wind LiDAR. Overall, the wind LiDAR demonstrated a good  
432 capability for measuring the vertical profile of wind, exhibiting a mean absolute deviation ranging  
433 from 0.5 m/s to 0.7 m/s compared to the collocated measurements (Figure S4). Considering the  
434 average wind speeds ranging from approximately 3 m/s to 10 m/s within PBL, the uncertainties  
435 arising from the wind measurements remained within 20%.

436 Vertical variations in pollutant concentrations may exist within the PBL. In this study, we  
437 made an assumption that air pollutants are well mixed within the PBL. In a convective mixing  
438 layer during the daytime, pollutants can be easily dispersed upward and mixed within the PBL. To  
439 gain a deeper understanding of the uncertainty stemming from this assumption, we compare the  
440 concentrations of  $O_3$  between Sham Shui Po and Tai Mo Shan. The former is situated at an  
441 elevation of 17 meters above sea level, whereas the latter stands at an elevation of 950 meters. As  
442 shown in Figure S9, we separate the comparisons into two parts: daytime (between 8:00 am and  
443 20:00 pm) and nighttime (before 8:00 am or after 20:00 pm). Correlation coefficients between  $O_3$   
444 concentrations at the two stations increased from 0.61 for nighttime to 0.87 for daytime, indicating  
445 the impacts of enhanced vertical mixing during the daytime. In addition, the mean absolute  
446 percentage deviations in the  $O_3$  concentration for the nighttime and daytime were estimated to be  
447 18.9% and 14.0%, respectively. Both of these values fall within the range of 20%. Future analyses  
448 can take into account the vertical distribution of pollutant concentrations when the data are

449 available. For instance, the ozone LiDAR system can be used to detect the vertical profile of O<sub>3</sub>  
450 concentrations. Synergistic measurements from wind and pollution LiDARs would greatly help to  
451 minimize the bias in the estimation of pollution flux in the upper PBL.

## 452 **5. Conclusion**

453 In this study, measurements from a wind LiDAR system were taken to monitor the vertical  
454 profile of wind pattern at an urban site in Hong Kong in September 2022. The PBL height was  
455 identified based on the vertical profile of wind speed shear. A cross-sectional analysis was  
456 performed to explore the THF of O<sub>3</sub> across the PBL in Hong Kong. Clockwise veering winds were  
457 observed from the ground to the top of the PBL, which can be attributed to the decreased influence  
458 of friction with increasing height. As a result, the wind with the peak O<sub>3</sub> flux shifted from westerly  
459 to northwesterly as the height increased in the PBL. Throughout the study month, the THF of O<sub>3</sub>  
460 exhibited a predominant easterly component. However, during the O<sub>3</sub> pollution episodes, the THF  
461 of O<sub>3</sub> exhibited a predominant westerly component, indicating an increased regional transport of  
462 pollutants from the GBA. These analyses provide insights into both the short-term variations and  
463 long-term average levels of air pollution. The findings enhance our understanding of the 3D  
464 pollutant transports in the GBA and Hong Kong. Future studies can combine wind and pollution  
465 LiDAR to minimize the bias in the estimation of pollution flux in the upper PBL.

## 466 **Supplement**

467 Additional figures (Figures S1–S9).

## 468 **Competing interests**

469 The authors declare that they have no actual or potential competing financial interests.

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**Highlights**

- Total horizontal flux (THF) of O<sub>3</sub> across the PBL was explored using a wind LiDAR
- The PBL height was identified based on the vertical profile of wind speed shear
- The THF of O<sub>3</sub> exhibited a predominant westerly component during O<sub>3</sub> episodes
- Clockwise veering flux of O<sub>3</sub> were observed from the ground to the top of the PBL
- Our results enhance our understanding of the 3D pollutant transports in the GBA

**Declaration of interests**

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

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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