



Mixing layer transport flux of particulate matter in Beijing, China

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18 Abstract

19 Quantifying the transport flux of atmospheric pollutants plays an important role in understanding 20 the causes of atmospheric pollution and in making decisions regarding the prevention and control 21 of atmospheric pollution. In this study, the mixing layer height and wind profile of the mixing layer 22 were measured by ceilometer and doppler wind radar, respectively. The variation characteristics of 23 atmospheric transport capacity (TC) were analyzed using these two datasets. The research showed 24 that the TC appears to be strongest in spring $(3940 \pm 2110 \text{ m}^2 \text{ s}^{-1})$ and weakens in summer $(2953 \pm$ 25 $1322 \text{ m}^2 \text{ s}^{-1}$), autumn ($2580 \pm 1601 \text{ m}^2 \text{ s}^{-1}$) and winter ($2913 \pm 3323 \text{ m}^2 \text{ s}^{-1}$). Combined with the 26 near-surface fine particle concentration data, the TC influence on the PM2.5 concentration was 27 studied, and there is a strong inverse correlation between the PM2.5 and TC in spring, autumn and 28 winter (R = -0.66, -0.65 and -0.80, respectively) and a weak positive correlation in summer (R = 29 0.33). By calculating the transport flux of fine particles (TF), the TF in Beijing was found to be the 30 highest in spring at 226 ± 294 mg m⁻¹s⁻¹ and lower in the other three seasons at approximately 140 mg m⁻¹s⁻¹. Transport occurs between 14:00 and 18:00 LT. Except for during spring, the TF was large 31 32 in the pollution transition period (summer: 328 ± 280 mg m⁻¹s⁻¹, autumn: 280 ± 336 mg m⁻¹s⁻¹ and 33 winter: 240 ± 297 mg m⁻¹s⁻¹) and decreased during the heavy pollution period (summer: 295 ± 215 34 mg m⁻¹s⁻¹, autumn: 243 ± 238 mg m⁻¹s⁻¹ and winter: 212 ± 209 mg m⁻¹s⁻¹). Our results indicate that 35 the transportation influence in southern regions should receive more focus in the transition period 36 of pollution, while local emissions should receive more focus in the heavy pollution period.

37 1. Introduction

38 With the rapid development of the economy and industry, as well as the unique local topography, 39 Beijing has become one of the cities in the world that is most seriously affected by air pollution. As 40 early as before the 2008 Olympic Games, to fulfill the promise of "Green Olympics", Beijing's 41 industries were relocated to other provinces and cities. After the Olympic Games, with the 42 promulgation of the "Action Plan for Prevention and Control of Air Pollution", Beijing 43 implemented a series of measures to reduce pollutants, such as raising the emission standards of 44 motor vehicles and fuel standards for vehicles, changing coal to natural gas, coal to electricity and 45 so on. These measures have gradually improved the Beijing's air quality, with the concentration of fine particulate matter decreasing from 90 μ g m⁻³ in 2013 to 58 μ g m⁻³ in 2017 46 47 (http://www.cnemc.cn/).

48 Although Beijing's government has been dedicated in recent years to taking measures that could 49 ensure a steady decrease in poor air quality, there is still great pressure to ensure the continuous 50 decline in particulate matter concentration. Beijing is in the north of the North China Plain, the south 51 side and the west side are the Yanshan Mountains and the Taihang Mountains, respectively. Affected 52 by the mountains to the northwest, there are more subsiding airflows, a lower mixing layer height 53 and an extremely limited atmospheric diffusion capacity. In addition, pollutants tend to accumulate 54 in front of the mountains due to the influence of southerly winds and mountain obstruction. In 55 central and northern China, the increase in PM2.5 during winter is closely related to adverse 56 atmospheric transport conditions (Wang et al. 2016). Therefore, in addition to primary emissions 57 and secondary formation, weak atmospheric transport capacity (TC) is an important factor leading 58 to the frequent occurrence of serious air pollution in Beijing.

59 In recent decades, mixing layer height (MLH) and wind speed (WS) are two major factors that lead





60 to the annual increase in aerosol concentration and haze days during winter in China (Yang et al. 61 2016). Additionally, low MLH and low WS are important characteristics of weak TC (Song et al. 62 2014; Tang et al. 2015; Huang et al. 2018; Liu et al. 2018). The change in MLH represents the vertical TC of pollutants, and the change in WS represents the horizontal TC of pollutants. To 63 characterize the TC, the ventilation coefficient (VC) is usually used to evaluate the vertical and 64 65 horizontal transport capacity of the atmosphere (Nair et al. 2007; Tang et al. 2015; Zhu et al. 2018). 66 Thus, it is a good choice to use VC to evaluate the relationship between TC and air pollution in 67 Beijing. Although previous studies have analyzed the relationship between MLH and pollutants 68 (Schäfer et al. 2006; Geiß et al. 2017; Su et al. 2018; Miao and Liu 2019), studies on the effects of 69 VC on particle concentration are extremely rare. 70 In addition, with the reduction in local emission sources, the contribution of regional transport 71 becomes particularly important. There are three main transport routes affecting Beijing: the 72 northwest path, the southwest path and the southeast path (Chang et al. 2018; Li et al. 2018; Zhang 73 et al. 2018). The occurrence of heavy pollution in Beijing is closely related to the transportation of 74 pollutants in southern regions, mainly in southern Hebei, northern Henan and western Shandong, 75 while the high-speed northwest air mass is conducive to the removal of pollutants in Beijing (Zhang 76 et al. 2017; Li et al. 2018; Zhang et al. 2018; Ouyang et al. 2019). In recent years, the contribution 77 of regional transport to Beijing has been increasing annually, with a trend of 1.2% year-1, which 78 reached 31-73% in summer and 27-59% in winter (Wang et al. 2015; Chang et al. 2018; Cheng et 79 al. 2018). High PM_{2.5} concentrations are usually accompanied by high transport flux within a day 80 in Beijing (Tang et al. 2015; Zhu et al. 2016). As pollution worsens, the contribution of the 81 surrounding areas to the PM_{2.5} in Beijing has risen from 52% to 65% in a month on average (Zhao 82 et al. 2018). However, during heavy pollution, the transport flux decreased in Beijing (Tang et al. 83 2015; Zhu et al. 2016; Chang et al. 2018). Although many studies on regional transport have been carried out, most observational studies cannot easily quantify transport flux due to the lack of wind 84 profile data. Therefore, transport flux can only be obtained by models. When the model lacks 85 86 verification data, the reliability of the model will decrease. Thus, it is imperative to quantify the 87 transport flux through observations. 88 To solve the above two problems, we conducted 2 years of continuous observations on MLH and

wind profiles in the Beijing mixing layer and analyzed the mixing layer TC of pollutants and their relationship with particulate matter. Then, combined with the concentration of particulate matter, we analyzed fine particulate matter transport flux in the mixing layer (TF). Finally, using the PM_{2.5} concentration as an indicator to classify the air pollution degree, we analyzed the TF in Beijing during the transitional and heavily polluted period and illuminated the main controlling factors.

94 2. Methods

95 2.1 Observational station

96 To understand the TC characteristics in Beijing, two years of observations were conducted in Beijing 97 (2016.1.1-2017.12.31). The observational site (BJT) is in the Institute of Atmospheric Physics of 98 the Chinese Academy of Sciences, located west of the Jiande Bridge in the Haidian District, Beijing 99 (39.98° N, 116.38° W). The north and south sides of the station are the north third and north fourth 100 ring roads respectively, and the eastern side is Beijing-Tibet expressway. The altitude (a.s.l.) is about





101 60 m. There is no obvious emission source around the observational site except the highway.

102 2.2 Observations of MLH and wind profiles

To analyze TC, MLH was observed by a single-lens ceilometer (CL51, Vaisala, Finland), and the 103 104 wind profile in the mixing layer was simultaneously observed by doppler wind radar (Windcube 105 100s, Leosphere, France). A single-lens ceilometer measures the attenuated backscatter coefficient 106 profile of atmospheric aerosols by pulsed diode laser lidar technology (910 nm waveband) within a 107 7.7 km range, and determine the MLH through the position of abrupt changes in the backscattering 108 coefficient profile. In the actual measurement, the measurement interval was 16 s and the 109 measurement resolution was 10 m. More detail descriptions are presented in the published literature (Tang et al. 2016; Zhu et al. 2016). In this study, the gradient method (Steyn et al. 1999) is used to 110 111 determine the MLH; that is, the top of the mixing layer was determined by the maximum negative 112 gradient value ($-d\beta/dx$) in the profile of the atmosphere backscattering coefficient. Moreover, to 113 eliminate the interference of aerosol layer structure and the detection noise to data, the MLH was calculated by the improved gradient method after averaging the profile data (Münkel et al. 2007; 114 115 Tang et al. 2015).

116 Doppler wind radar uses the remote sensor method of laser detection and ranging technology and 117 measures the doppler frequency shift generated by the laser through the backscatter echo signal of 118 particles in the air. Windcube 100s can provide 3D wind field data within a 3 km range from the 119 system, including u, v and w vectors. In the actual measurement, starting from 100 m, the spatial 120 resolution is 50 m, the WS accuracy is $< 0.5 \text{ m s}^{-1}$, and the radial WS range is $-30 \text{ m s}^{-1} - 30 \text{ m s}^{-1}$.

121 2.3 Other data

During the observations, the hourly PM_{2.5} and ozone surface concentrations of the Beijing Olympic
 Sports Center (39.99° N, 116.40° W) were obtained from the Ministry of Environmental Protection
 of China (http://www.zhb.gov.cn/).

125 2.4 Analytical method

126 VC (m² s⁻¹) was obtained by combining MLH (m) and wind speed in the mixing layer (WS_{ML}, m s⁻¹), which can be used to characterize TC. A higher VC indicates a stronger TC, which is conducive 128 to the transport and diffusion of heavy air pollution. The VC calculation method is as follows: 120 $VC = ML V \times WS$ (1)

$$129 \quad VC = MLH \times WS_{ML},\tag{1}$$

$$130 \qquad WS_{ML} = \frac{1}{n} \sum_{i=1} WS_i, \tag{2}$$

131
$$WS = \sqrt{\overline{u}^2 + \overline{v}^2},$$
 (3)

where WS_{ML} is the average WS within the mixing layer, calculated by Eq. (2); WS_i is the WS
observed at all heights, calculated by the mean value of u and v in the wind profile according to Eq.
(3); and n is the number of measurement layers in the mixing layer (Nair et al. 2007).

135 TF (mg m⁻¹s⁻¹) is determined by TC and the $PM_{2.5}$ concentration in the area under analysis. The 136 calculation method for a certain height is shown in Eq. (4):

137
$$TF_{u_1} = u_1 \times C_{PM_{2,F}} \times MLH$$

(4)





138 It is extremely difficult to observe the $PM_{2.5}$ concentration in the mixing layer by height, but 139 previous observations have shown that the backscattering coefficient profile in the mixing layer is 140 relatively uniform (Tang et al. 2015). Assuming that the particle concentration in the mixing layer 141 is uniform, the TFs are calculated as follows:

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$$TF_{u} = \frac{1}{n} \sum_{i=1}^{n} u_{i} \times C_{PM_{2,5}} \times MLH$$

143
$$TF_{v} = \frac{1}{n} \sum_{i=1}^{n} v_{i} \times C_{PM_{2,5}} \times MLH$$
(5)

144 Through the above method, radial and zonal transport fluxes can be obtained, and vector synthesis 145 in two directions can be conducted to obtain the main transport direction to find the transport source 146 area.

147 3. Results and discussions

148 **3.1 Mixing layer transport capacity (TC)**

149 3.1.1 Seasonal variation

To understand the variations of TC, we carried out continuously measured MLH and wind profiles within the mixing layer over a 2-year period (2016.1.1-2017.12.31). The availability was verified after MLH elimination by Tang et al. (Tang et al. 2016). After the exclusion of the data of MLH under rainy, sandstorm and windy conditions, data availability was 95% over the 2-year period, higher than that of previous studies (Tang et al. 2016; Mues et al. 2017). The availability was lowest in February at 86% and highest in July at 99%.

156 The seasonal variation in MLH was higher in spring $(781 \pm 229 \text{ m})$ and summer $(767 \pm 219 \text{ m})$ and lower in autumn (612 \pm 166 m) and winter (584 \pm 221 m) (Fig. 1). However, WS_{ML} was quite 157 different from MLH in terms of seasonal variation, with the largest value at 4.6 ± 1.6 m s⁻¹ in spring, 158 followed by winter $(4.1 \pm 2.7 \text{ m s}^{-1})$ and autumn $(3.7 \pm 1.6 \text{ m s}^{-1})$, and the smallest value at 3.6 ± 1.1 159 160 m s⁻¹ in summer. VC was calculated by the MLH and wind profile, and the seasonal variation in TC over 2 years was analyzed (Fig. 1). The results demonstrate that the TC was strongest in spring, as 161 the VC reached as high as 3940 \pm 2110 m² s⁻¹. The TC differences among summer, winter and 162 163 autumn were small when the VC values were 2953 ± 1322 m² s⁻¹, 2913 ± 3323 m² s⁻¹ and $2580 \pm$ 1601 m² s⁻¹, respectively. A monthly analysis shows that the TC was the strongest in May, the VC 164 165 was as high as 5161 ± 2085 m² s⁻¹, the TC was the worst in December, and the VC was only 1690 \pm 1072 m² s⁻¹. The VC value in May was 3.1 times higher than that in December. The seasonal 166 variation in the PM_{2.5} concentration was the highest in winter ($80 \pm 87 \ \mu g \, m^{-3}$), followed by autumn 167 $(68 \pm 54 \ \mu g \ m^{-3})$ and spring $(67 \pm 60 \ \mu g \ m^{-3})$, and the seasonal variation was the lowest in summer 168 169 $(51 \pm 29 \ \mu g \ m^{-3})$. The lowest monthly average PM_{2.5} concentration was $42 \pm 26 \ \mu g \ m^{-3}$ in August. 170 The highest monthly average was in January at $94 \pm 100 \ \mu g \ m^{-3}$, 2.2 times higher than that in August 171 (Fig. 1). Thus, the vertical and horizontal diffusion capacities are strong in spring and weak in autumn and winter. In summer, the vertical diffusion capacity is strong, while the horizontal 172 173 diffusion capacity is weak. Overall, high PM2.5 concentrations are associated with poor TC.







176 3.1.2 Diurnal variation

177 Moreover, the diurnal variations in meteorological factors during different seasons were analyzed 178 to reveal the diurnal evolution characteristics of atmospheric TC. The peak and trough values of 179 MLH and VC appeared simultaneously at approximately 15:30 LT and 05:30 LT, respectively. 180 Generally, the daily variation in MLH is characterized by a low value at night, which increases rapidly after sunrise and reaches the maximum value in the afternoon (Fig. 2a). The daily maximum 181 182 value of MLH is seasonal, where it is higher in spring and summer and lower in autumn and winter. 183 The daily minimum value of MLH generally occurs when the mixing layer is stable and is closely related to WS. The diurnal variation in WS_{ML} is stable, with a peak at approximately 19:30 LT and 184 185 a trough at approximately 10:00 LT, which is 4 h later than the peak valley of MLH (Fig. 2b). The 186 diurnal variation in VC is similar to MLH, showing that the TC is strong before sunset, gradually 187 weakens after sunset and remains stable at night. The TC in spring was significantly stronger than 188 that during other seasons, and the maximum daily value reached 8678 m² s⁻¹ (Fig. 2c). In addition 189 to spring, the daily maximum values of VC in summer, autumn and winter were close at 190 approximately 5000 m² s⁻¹ (Fig. 2c). The TC growth rate in spring was significantly higher than that 191 in other seasons, reaching a maximum at approximately 09:00 LT. Late in autumn, the TC growth rate peaked at approximately 10:00 LT. Summer and winter peaked at approximately 11:00 LT. 192 Throughout the year, VC began to increase during winter at the latest, at approximately 09:00 LT, 193 194 indicating that the weaker TC remained for a longer period during winter. TC was weakened most 195 rapidly in spring; however, the TC was still higher than the VC of other seasons after declining. In 196 addition to spring, the TC in autumn and winter weakened the most rapidly and the slowest in 197 summer. In general, vertical and horizontal diffusion is very strong in the spring during both day 198 and night. In winter, vertical diffusion is weak during the day, and horizontal transportation during 199 the night is the main transportation. In summer, vertical diffusion during the day is dominant.



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Fig. 2. Diurnal variations and growth rates of MLH (a), WS_{ML} (b) and VC (c) in spring, summer, autumn and winter in Beijing. Diurnal variations are represented by lines and scatters. Growth rates are represented by columns, and only positive values are shown in the figure.





204 3.1.3 Frequency distribution

205 Although there is little difference in TC between summer, autumn and winter, there is serious 206 pollution in autumn and winter. To analyze this problem, the VC frequency distribution was studied. 207 The results show that VC had a high frequency in the range of 1000-4000 m² s⁻¹ from 2016 to 2017, but the frequency distribution was different in different seasons (Fig. 3). The VC showed a strong 208 209 TC in spring, mainly in the range of 2000-5000 m² s⁻¹, with the highest frequency (24%) in the range of 2000-3000 m² s⁻¹. In summer, the high frequency of VC occurred in the range of 1000-4000 m² 210 s⁻¹, which was slightly lower than that in spring, and the highest frequency (27%) occurred in the 211 212 range of 3000-4000 m² s⁻¹. Additionally, the VC high frequency appeared in a lower range in autumn 213 and winter. The VC occurred at a high frequency of 1000-3000 m² s⁻¹ in autumn, and the highest 214 frequency occurred within the range of 2000-3000 m² s⁻¹, accounting for 33%. In winter, VC appeared more frequently in the range of 0-2000 m² s⁻¹ and was the highest in the range of 1000-215 2000 m² s⁻¹, which was 28%. However, the VC frequency of 0-1000 m² s⁻¹ in winter was 216 217 significantly higher than that of the other seasons, up to 22%, which was 7 times higher than that of 218 spring, 5 times higher than that of summer and 2 times higher than that of autumn. According to the 219 seasonal variation in PM_{2.5} concentration, heavy pollution in autumn and winter is related to the 220 high frequency of poor TC.





Fig. 3. Frequency distribution of the daily VC from January 2016 December 2017 in Beijing.

224 **3.2 Response of particulate matter to TC**

225 Studies have found that air pollution worsens when TC weakens (Tang et al. 2015; Liu et al. 2018; 226 Sun et al. 2018). To further understand the response of fine particles to TC in different seasons, the correlations between meteorological factors and PM2.5 concentration were analyzed (Fig. 4). From 227 228 2016 to 2017, the annual average PM_{2.5} concentration was $66 \pm 62 \mu g m^3$, the maximum concentration was 898 µg m⁻³, and the minimum concentration was only 1 µg m⁻³, which showed 229 230 high concentrations in autumn and winter. As shown in Fig. 4, PM2.5 concentrations increased 231 exponentially with decreases in MLH, WS_{ML} and VC, indicating that the concentration of fine particles was highly sensitive to these meteorological factors. When MLH, WS_{ML} and VC were 232 233 lower than 400 m, 2.5 m s⁻¹ and 1500 m² s⁻¹, respectively, the air pollution declines sharply. VC had 234 a better correlation with the $PM_{2.5}$ concentration than MLH and WS_{ML} , indicating that VC can better 235 characterize pollution dissipation. The PM_{2.5} concentration in winter had a better response to TC 236 than the other seasons, with the correlation coefficient with VC reaching -0.80, followed by spring





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and autumn, with correlation coefficients of -0.66 and -0.65, respectively (Fig. 4). The correlation in spring and autumn may decrease due to dust. In summer, $PM_{2.5}$ had a poor relationship with WS_{ML} and even had weak positive correlations with MLH (R = 0.42) and VC (R = 0.33). A high ozone concentration existed in the high MLH (Fig. 4), which will promote the transformation of gaseous precursors to secondary particles. Therefore, the weak positive correlation in summer was related to a strong photochemical reaction.

- Thus, MLH, WS_{ML} and VC can be used as indicator factors for the formation of air pollution, but
 the particle concentration responds best to VC. Additionally, the response of particle concentration
- to VC showed obvious seasonal differences, with the best in winter, followed by autumn and spring,
- and a weak positive correlation in summer.





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250 3.3 Mixing layer transport flux of particulate matter

251 To quantify the transport of pollutants in Beijing, the Beijing TF was analyzed, and the transport 252 direction of fine particles was characterized by the wind direction in the mixing layer. As shown in 253 Fig. 5, the TF in spring was the largest, reaching 226 ± 294 mg m⁻¹s⁻¹, and there was no significant 254 difference in summer, autumn or winter, when the TF values were 147 ± 182 mg m⁻¹s⁻¹, 143 ± 194 mg m⁻¹s⁻¹ and 134 ± 179 mg m⁻¹s⁻¹, respectively. The northwesterly and westerly directions were 255 256 the main transport sources of the cold period in Beijing. With temperature warming, the transport 257 direction gradually increased from west to south, mainly as a southwesterly in spring and southerly 258 in summer. The monthly average maximum value of TF occurred in May, as high as 269 ± 328 mg 259 m⁻¹s⁻¹ and mainly originated from the southwest direction, which was accompanied by a strong wind. The minimum value appeared in August, as low as 106 ± 145 mg m⁻¹s⁻¹, which was mainly 260 261 transported from western regions, with low WS values. The TF in May was 2.5 times higher than 262 that in August (Fig. 5). Therefore, the change in transport direction leads to an obvious seasonal 263 variation in TF. Overall, the regional transport contributes the most to the particulate matter 264 concentration in spring, which is mainly related to increased dust activities; regional transport has the least contribution in winter, which indicates that more focus should be given to local emission 265 source control; in summer and autumn, the southwest airflow transportation influence on Beijing 266 267 should receive more focus.



Fig. 5. Seasonal variations in the mixing layer transport flux of PM and transportation directions. 269 To understand the regional transport influence on the Beijing area, the diurnal variation 270 271 characteristics of TF were analyzed during different seasons in Beijing. The daily minimum value of TF appeared at approximately 07:00 LT and was accompanied by a northerly wind. As the wind 272 273 direction gradually turned south, the daily minimum value of TF continued to rise until the daily 274 maximum value appeared at approximately 16:00 LT (Fig. 6). Transportation mainly occurred between 14:00 and 18:00 LT, which was consistent with the results of a previous study (Ge et al. 275 276 2018). In spring, the WS was higher, so the peak TF duration was shorter, at approximately 2 h. The 277 maximum daily value was 494 mg m⁻¹s⁻¹, and the minimum was 87 mg m⁻¹s⁻¹ in spring. Therefore, the diurnal variation in TF during spring showed the characteristics of a rapid rise and rapid decline. 278 279 The peak duration was approximately 4 h for a long time in summer and autumn, where the daily maximum values were 259 mg m⁻¹s⁻¹ and 240 mg m⁻¹s⁻¹, and the minimum values were 53 mg m⁻¹ 280 281 ¹s⁻¹ and 66 mg m⁻¹s⁻¹, respectively. The diurnal variation in TF during summer and autumn showed 282 the characteristics of a slow rise and slow decline. Specifically, the daily variation had a strong fluctuation in winter, which peaked at only 16:00 LT (215 mg m⁻¹s⁻¹), then dropped sharply to 193 283 284 mg m⁻¹s⁻¹, plateaued from 17:00 to 22:00 LT for approximately 5 h, maintained at approximately 285 176 mg m⁻¹s⁻¹, and then quickly dropped to 78 mg m⁻¹s⁻¹





286 The TF variation rules can be summarized as a high TF corresponds to a southerly wind and a low 287 TF corresponds to a northerly wind. When the wind direction in the mixing layer changed from north to south, the wind gradually increased from the daily minimum to the daily maximum. The 288 289 TF increased by 6 times in spring, 5 times in summer, 4 times in autumn and 3 times in winter. The 290 current pattern is because areas south of Beijing are heavily polluted and southerly winds help 291 transport pollutants into the city, leading to high transport flux in spring, summer and autumn 292 afternoons (Fig. 6). The results further confirm the conclusion that the northwest wind in Beijing is 293 a clean wind (Wang et al. 2015; Zhang et al. 2018). Thus, the north wind is conducive to the outward 294 transport of pollutants from Beijing, which helps to alleviate pollution. As a result, there was no 295 high TF in winter when the westerly wind and northerly wind prevailed. This finding also proves the important influence of local emissions on heavy pollution occurrence during winter in Beijing. 296 297 In summary, there are 4 main transport routes that affect Beijing, including the northwest path, 298 southwest path, west path and south path. The TF in winter is low, local emissions play an important 299 role, and we must pay attention to local pollutant emission control.



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Fig. 6. Diurnal variations in the mixing layer transport flux of PM and transportation directions
 during different seasons in Beijing.

303 3.4 TF under different degrees of air pollution

Previous studies have demonstrated that transportation occurs only at the transition period of 304 305 pollution, and transportation is weak at the peak of pollution (Tang et al. 2015; Zhu et al. 2016). To 306 quantify the transport impact of different pollution levels, the PM2.5 concentration was divided into five levels according to the "Technical Regulation on Ambient Air Quality Index (on trial)" 307 (HJ 633-2012): $PM_{2.5} \le 35 \ \mu g \ m^{-3}$ (clear days), $35 < PM_{2.5} \le 75 \ \mu g \ m^{-3}$ (slight haze), $75 < PM_{2.5} \le 10^{-3}$ 308 115 μ g m⁻³ (light haze), 115 < PM_{2.5} \leq 150 μ g m⁻³ (medium haze) and PM_{2.5} > 150 μ g m⁻³ (heavy 309 310 haze). With pollution aggravation, the TF in Beijing increased by varying degrees during different seasons, and the transportation direction gradually shifted from northwest to south (except during 311 312 winter) (Fig. 7). In particular, the TF continued to increase only in spring, from 93 ± 124 mg m⁻¹s⁻¹ 313 on clear days to 382 ± 438 mg m⁻¹s⁻¹ on heavily polluted days, which may be caused by more dust during spring. With the except of during spring, with pollution deterioration, the TF showed an 314 increasing trend at the initial stage of pollution and decreasing trend during the heavy pollution 315 period. From medium haze to heavy haze, the TF decreased from 328 ± 280 mg m⁻¹s⁻¹ to 295 ± 215 316 317 mg m⁻¹s⁻¹in summer, from 280 ± 336 mg m⁻¹s⁻¹ to 243 ± 238 mg m⁻¹s⁻¹ in autumn, and from $240 \pm$ 297 mg m⁻¹s⁻¹ to 212 ± 209 mg m⁻¹s⁻¹ in winter. These results indicate that although the region south 318 319 of Beijing is the main transport source during summer and autumn in Beijing, this contribution is 320 significantly reduced during the severe pollution period. In winter, with pollution aggravation, the 321 transportation direction changed from northwest to southwest and finally to the north. In contrast to





322 other seasons, the north wind with a low WS was the main wind during heavy pollution in winter,

323 indicating that regional transport contributed less to heavy pollution during winter in Beijing. In

324 general, the transport of pollutants from the southwest is the main controlling factor for pollution

325 occurrence during spring in Beijing. In other seasons, regional transport plays an important role in 326 the initial period of pollution, while local emissions during the period of heavy pollution are the

327 main controlling factor.



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Fig. 7. The mixing layer transport flux levels of PM and transportation directions under different degrees of pollution.

331 4. Conclusions

To understand the characteristics of fine particulate matter transport flux in Beijing, the height of
 the atmospheric mixing layer and wind profile within the mixing layer in Beijing were observed for
 a 2-year period. The main conclusions are as follows:

335 (1) By analyzing the variation characteristics of VC, the TC in Beijing is strongest in spring and 336 weaker in summer, autumn and winter. In spring, vertical and horizontal diffusion capacities are 337 strong; in autumn and winter, vertical and horizontal diffusion capacities are weak; in summer, 338 vertical diffusion capacity is strong and horizontal diffusion capacity is weak. The diurnal variation 339 in VC is consistent with MLH, which shows that the TC is strongest before sunset, gradually 340 weakens after sunset and remains stable at night. In spring, vertical and horizontal diffusion are very 341 strong during both day and night. In winter, vertical diffusion is weak during the day, and horizontal 342 transportation during the night is the main means of transportation. In summer, vertical diffusion during 343 the day is dominant. Although there is little difference in diffusivity between summer, autumn and 344 winter, poor TC occurs more frequently in autumn and winter.

(2) PM_{2.5} concentrations during different seasons have different responses to MLH, WS_{ML} and VC.
During the three dry seasons of winter, spring and autumn, the concentration of pollutants has a
good relationship with VC, indicating that the main dissipation method of pollutants is diffusion. In
summer, there is a weak positive correlation between pollutant concentration and VC, which is
related to strong photochemical reactions.

350 (3) TF is largest in spring and smaller in summer, autumn and winter in Beijing. The high TF mainly

351 comes from southward transport, while the low TF is accompanied by northwest transport. Using





the PM_{2.5} concentration as a classified index of atmospheric pollution, the results show that the regional transport of pollutants from the southwest is the main controlling factor of pollution during spring in Beijing, while during the other seasons, the regional transport from the southern area plays an important role in the initial period of pollution, and local emissions are the main controlling factors in the heavy pollution period, especially in winter.
In this study, the response of particulate matter to meteorological conditions in the mixing layer was

study, the response of particulate matter to intercorological conditions in the mixing layer was studied, and the difference in the seasonal response was found. The transport capacity during different seasons and the transport flux during different pollution periods were also discussed. The research results are of great significance to the early warning, prevention and control of atmospheric particulate pollution. However, due to the limitation of observational data, the near-surface particle concentration was used to replace the concentration column for discussion purposes, resulting in uncertainty in the result. In the future, this issue will be further discussed in combination with ground-based telemetry lidar.

365 Data availability

366 The data in this study are available from the corresponding author upon request (tgq@dq.cern.ac.cn).

367 Author contribution

GT and YW designed the research, LZ, BH, BL and YunL conducted the measurements. YusL andGT wrote the paper. SL reviewed and commented on the paper.

370 **Competing interests**

371 The authors declare that they have no conflict of interest.

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