1 Self-organized classification of boundary layer meteorology and associated

2 characteristics of air quality in Beijing

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- 16 Abstract

17 Self-organizing maps (SOMs; a feature-extracting technique based on an unsupervised machine learning 18 algorithm) are used to classify atmospheric boundary layer (ABL) meteorology over Beijing through detecting topological relationships among the 5-year (2013-2017) radiosonde-based virtual potential temperature profiles. 19 20 The classified ABL types are then examined in relation to near-surface pollutant concentrations to understand the 21 modulation effects of the changing ABL meteorology on Beijing's air quality. Nine ABL types (i.e., SOM nodes) 22 are obtained through SOM classification technique, and each is characterized with distinct dynamic and 23 thermodynamic conditions. In general, the self-organized ABL types are able to distinguish between high and low 24 loadings of near-surface pollutants. The average concentrations of PM2.5, NO2 and CO dramatically increased 25 from the near neutral (i.e., Node 1) to strong stable conditions (i.e., Node 9) during all seasons except for summer. Since extremely strong stability can isolate the near-surface observations from the influence of elevated SO_2 26 27 pollution layers, the highest average SO_2 concentrations are typically observed in Node 3 (a layer with strong stability in the upper ABL) rather than Node 9. In contrast, near-surface O₃ shows an opposite dependence on 28 atmospheric stability, with the lowest average concentration in Node 9. Analysis of three typical pollution months 29 (i.e., January 2013, December 2015 and December 2016) suggests that the ABL types are the primary drivers of 30 day-to-day variations in Beijing's air quality. Assuming a fixed relationship between ABL type and PM_{2.5} loading 31

for different years, the relative (absolute) contributions of the ABL anomaly to elevated $PM_{2.5}$ levels are estimated to be 58.3 % (44.4 µg/m³) in January 2013, 46.4 % (22.2 µg/m³) in December 2015, and 73.3 % (34.6 µg/m³) in December 2016.

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36 1 Introduction

37 The atmospheric boundary layer (ABL) is the section of atmosphere that responds directly to the flows of mass, energy and momentum from the earth's surface (Stull, 1988). Since most air pollutants are emitted or chemically 38 produced within this layer, its evolution plays an important role in transport, dispersion and deposition of air 39 40 pollutants (Chen et al., 2012;Fan et al., 2008;Whiteman et al., 2014;Platis et al., 2016;Wolf et al., 2014;Wu et al., 2013). The ABL structure is determined by complex interactions between atmosphere static stability and those 41 mechanical processes (such as wind shear from synoptic or terrain-induced flows) (Stull, 1988). These processes 42 43 can operate at a variety of different heights and temporal scales, and their dominance may vary considerably with height and time at any given location (Salmond and McKendry, 2005). This makes it very difficult to observe and 44 predict the transport and diffusion of air pollutants within the ABL (Chambers et al., 2015b;Chambers et al., 45 2015a), particularly in those complex-terrain regions such as Beijing. 46

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Beijing, the capital of China, is geographically located at the northwestern border of the Great North China 48 49 Plain. This city is surrounded by the Yan Mountains to the north and the Taihang Mountains to the west, with the Bohai Sea to the 160 km southeast (Fig. 1). Under favorable weather conditions (e.g., stagnant weather), 50 51 terrain-related circulations can be well developed over Beijing and its surroundings, leading to a complex ABL structure, which is thought to substantially affect Beijing's air quality (Hu et al., 2014; Miao et al., 2017; Ye et al., 52 2016;Gao et al., 2016;Xu et al., 2016). With high emissions of air pollutants from anthropogenic sources, Beijing 53 54 is suffering serious air pollution problems and the pollution can be even more severe when southwesterly and southeasterly winds prevail within the ABL (Chen et al., 2008;Ye et al., 2016;Zhang et al., 2014;Zhang et al., 55 2012). 56

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58 Several studies investigated the interactions between ABL meteorology and air quality in Beijing using 59 tower-based observations (Sun et al., 2013;Sun et al., 2015;Guinot et al., 2006;Guo et al., 2014). However, the 60 results are not ideal because the tower-based observations have a low observational height (325 m). In addition, 61 numerous intensive ABL measures were conducted using other approaches such as mooring boats, airplane, and 62 ground-based remote sensing (Tang et al., 2015;Zhu et al., 2016;Zhang et al., 2009;Hua et al., 2016). Since these approaches are complex, expensive and labor intensive, they are often restricted to the duration of specific research campaigns and hence their results may be considered 'unrepresentative'. Overall, the existing knowledge of linkages between ABL meteorology and air quality in Beijing is drawn largely from either low observational height or short observational duration; therefore, the common patterns of the influence of the changing ABL structures on Beijing's air quality remains unclear and need to be further studied (Quan et al., 2013;Miao et al., 2017;Guo et al., 2014).

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70 Meanwhile, the routine radiosondes are not being fully utilized to investigate urban pollution issues. The 71 advantage of radiosondes over the other approaches seems to be their length, which usually spans several decades. 72 For a long time, it was challenging to reduce the wealth of radiosonde data to characterize the ABL structure, and 73 therefore routine radiosondes were remained a very limited use in case studies (Ji et al., 2012;Zhao et al., 74 2013;Gao et al., 2016). Recently, self-organizing maps (SOMs; a feature-extracting technique based on an unsupervised machine learning algorithm) (Kohonen, 2001) were introduced to investigate the ABL 75 thermodynamic structure, indicating the capabilities of SOMs in feature extraction from a large dataset of the 76 77 ABL measurements (Katurji et al., 2015). In fact, the SOM has an increasing application in atmospheric and 78 environmental sciences during the past several years (Jensen et al., 2012; Jiang et al., 2017; Gibson et al., 79 2016; Pearce et al., 2014; Stauffer et al., 2016), including a radiosonde-based application in South Africa (Dyson, 80 2015). However, there is thus far no SOM application in air pollution-related ABL structure research. It is expected such a new analytical approach can tap the potential of routine radiosondes to reveal the ABL 81 82 mechanism of air pollution in Beijing.

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This study investigates the influence of ABL meteorology on Beijing's air quality based on the SOM application to 5 years (2013–2017) of routine meteorological radiosondes. First, we use the SOM technique to classify the state of ABL through detecting topological relationships among the radiosonde-based virtual potential temperature profiles (see section 3.1). Then, we provide a visual insight into near-surface pollutant variations under various ABL types and discuss the potential physical mechanisms behind their relationships (see section 3.2–3.3). It is expected that such an association between air quality and ABL type could provide local policy makers with useful information for improving the predictions of urban air quality.

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92 2 Materials and methods

93 2.1 Data preparation and preprocessing

The recent 5-year (2013–2017) radiosonde data observed at the Beijing Observatory (39.81 N, 116.48 E, WMO station number 54511) were collected from the University of Wyoming (http://weather.uwyo.edu/). The radiosondes were launched twice a day (08:00 and 20:00 Local Time, corresponding to the morning and evening, respectively) and provided atmospheric sounding data (profiles of temperature, relative humidity, wind speed, etc.) at the mandatory pressure levels (e.g., surface, 1000, 925, 850, 700 hPa) and additional significant levels. In addition, the hourly near-surface meteorological parameters (including temperature, wind speed and relative humidity, etc.) in 2013–2016 were collected from the Beijing Meteorological Bureau.

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102 We chose the 2000 m above ground level as the upper limit of the ABL based on a number studies investigating 103 the ABL height over Beijing or North China (Tang et al., 2016;Guo et al., 2016;Miao et al., 2017). This height exceeded the top of the ABL in most cases, and therefore, most ABL processes influencing the near-surface air 104 105 quality were included in the analysis herein. In our study period, the average number of data points in radiosonde profiles was 3.7 below 500 m and 10.1 below 2000 m. Despite the coarse resolution, the profile shapes were 106 enough for SOM technique to classify the state of ABL. Previous radiosonde-based study indicated that surface 107 108 temperature inversions occur frequently in the eastern China (Li et al., 2012), suggesting that all of the two-time 109 radiosondes mainly represent the nocturnal stable ABL. To keep a whole night, the daily ABL profiles were 110 composited from the radiosondes at 20:00 and 08:00 of the next day.

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The mass concentrations of atmospheric pollutants (including PM2.5, O3, NO2, SO2 and CO) over Beijing during 112 the period from 2013 to 2017 are obtained from the Ministry of Environmental Protection of the People's 113 Republic of China (http://datacenter.mep.gov.cn/). In addition, hourly PM_{2.5} measured at the Beijing US Embassy 114 (http://www.stateair.net/) are also used in this study. The PM2.5 values in the two datasets show a well consistence 115 116 with a mean correlation coefficient of 0.94. The mean hourly standard error of PM_{2.5} across sites changes little from 12.6 to 12.9 after the inclusion of US Embassy. Hourly concentrations are calculated for the Beijing urban 117 area by averaging concentrations from nine urban sites (including Dongsi, Guanyuan, Tiantan, Wanshouxigong, 118 Aotizhongxin, Nongzhanguan, Gucheng, Haidianwanliu and US Embassy). The daily pollutant concentration is 119 120 then performed afternoon-to-afternoon (15:00 h–15:00 h), in order to include one whole night in each 24-h period.

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122 **2.2 Self-organizing maps technique**

123 The SOM is thought to be an ideal tool for feature extraction because the input data are treated as a continuum 124 without relying on correlation, cluster or eigenfunction analysis (Liu et al., 2006). Since Kohonen (1982) first proposed SOM, it has been widely used for data downscaling and visualization in various disciplines (Jensen et al., 2012;Katurji et al., 2015;Dyson, 2015;Stauffer et al., 2016;Pearce et al., 2014;Jiang et al., 2017). In this study, the SOM is introduced to classify the ABL types through detecting topological relationships among the 5-year (2013–2017) radiosonde-based virtual potential temperature profiles. Since the SOM is sensitive to the virtual potential temperature value, the deviation profiles, which are determined by subtracting the mean virtual potential temperature of each profile from each level, are used as the SOM input.

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The training of SOM is an unsupervised, iterative procedure, and the result is a matrix of nodes (i.e., types) that 132 133 represent the input data. The following provides a simple introduction about the SOM algorithm, and the details 134 can be found in Kohonen (2001). To learn from the input data, every SOM node has a parametric reference vector with which it is associated, and these reference vectors are randomly generated. After initialization of the 135 136 reference vectors, a stochastic input vector is compared to every reference vector, and the closest match, named the best-matching unit, is determined by the smallest Euclidean distance. Each reference vector is then updated so 137 that the best-matching unit and its neighbors become more like the input vector. Whether or not a reference vector 138 learns from the input vector is determined by the neighborhood function. Only reference vectors that are 139 140 topologically close enough to the best-matching unit will be updated according to the SOM learning algorithm.

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142 The first step of SOM training is to determine a matrix size of nodes for initializing the reference vectors. This step is performed subjectively and depends on the degree of generation required (Lennard and Hegerl, 2015). We 143 test several SOM matrixes, and finally select a 3×3 matrix, because it captured unique profiles without the 144 profiles being too general as with a smaller matrix, or being too similar as with a larger matrix. In addition, the 145 batch mode is chosen to execute the SOM algorithm, because it is much more computationally efficient compared 146 147 to the sequence mode. The other user-defined settings in the SOM software are set at the default, such as the hexagon topology, Gaussian neighborhood function, etc. The SOM code used in this study is sourced from the 148 149 MATLAB SOM Toolbox, which is freely available from http://www.cis.hut.fi/projects/somtoolbox/.

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151 **2.3** Measuring the discriminative power of SOM technique for pollution assessment

The Kruskal-Wallis one-way analysis of variance is used as a non-parametric method to test the difference of pollutant concentrations among the various ABL types. A 1% significance level is used and hereafter denoted as *KW* in Sect. 3.2. Furthermore, the coefficients of variation (*CV*) of pollutant means across the various ABL types are also used to examine the discriminative power of the SOM technique for pollution assessment.

157 **3 Results and discussion**

158 3.1 Self-organized boundary layer meteorology

We construct a 3×3 SOM matrix for daily virtual potential temperature deviation profiles, and the 159 self-organized output shown in Fig. 1 represents nine ABL types (i.e., SOM nodes). On the SOM plane, the most 160 161 notable feature is adjacency of like types (e.g., Nodes 1 and 2) and the separation of contrasting types (e.g., Nodes 1 and 9). Such ordering is a feature of the SOM algorithm (i.e., 'self-organized'), which allows us to distinguish 162 163 the unique characteristics of nodes through the variation of specific features across the SOM plane. According to 164 the ordering feature, the SOM nodes in the four corners (i.e., Nodes 1, 3, 7 and 9) can be thought of as the typical types and the others can be considered as transitional types. The four typical ABL types have a relatively higher 165 166 occurrence frequency (> 10%), with the highest frequency associated to Node 1 (23%). Furthermore, the seasonal 167 statistical results (Fig. 2) reveal that these self-organized ABL types exhibit a strong seasonality. For example, Node 3 occurs more frequently in winter and autumn, while Node 1 has a relatively higher occurrence in spring 168 169 and summer.





Figure 1. The 3 × 3 SOM output for radiosonde-based virtual potential temperature (θ_v) deviation profiles observed at the Beijing Observatory. SOM nodes are shown in red, with the corresponding individual profiles in grey. For reference, the overall average θ_v deviation profile and 25th and 75th percentile profiles

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Figure 2. Relative frequency of individual ABL types (i.e., SOM nodes) in all four seasons. Winter (DJF);
Spring (MAM); Summer (JJA); Autumn (SON).

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Figure 3 displays the average profiles of wind speed, relative humidity, virtual potential temperature gradient 181 182 according to the ABL types. Clearly, each of the self-organized types features distinct dynamic and 183 thermodynamic conditions within the ABL, suggesting the SOM technique is feasible to classify the boundary 184 layer meteorology. Since the classification is based on the twice-daily radiosondes, the resulting ABL types are 185 dominated by near neutral to strong stable conditions, and none of the types fall within the unstable category (i.e., $\Delta \theta_v / \Delta z < 0$). While Node 3 features the strong static stability in the upper ABL (the large $\Delta \theta_v / \Delta z$ values), Node 1 186 represents a near neutral ABL condition with the lowest $\Delta \theta_{v}/\Delta z$ values and the highest wind shears in the lower 187 188 ABL. In contrast, Node 7 corresponds to a moderate static stability in the lower ABL, and Node 9 relates to a strong static stability. Particularly, the virtual potential temperature gradient in Node 9 remains a high level (> 189 190 0.7°C/100m) from surface to approximately 800 m, indicating a strong and deep surface temperature inversion 191 developed in this type. In addition, due to the strong surface inversion, vertical mixing is suppressed, resulting in a 192 strong decreasing gradient in humidity profiles. Overall, the SOM classification scheme reveals a significant 193 coupling between dynamic and thermal effects in the ABL, which is expected to considerably impact the 194 near-surface air quality.



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197Figure 3. Profiles of average wind speed (WS), relative humidity (RH) and virtual potential temperature198gradient ($\Delta \theta_v / \Delta z$) corresponding to individual ABL types (i.e., SOM nodes) at the Beijing Observatory. The199black, green and red labels of the horizontal axis correspond to $\Delta \theta_v / \Delta z$, WS and RH, respectively.

To detect the boundary layer development after sunrise, daytime boundary layer height (BLH) is estimated with 201 202 parcel method (Holzworth, 1964, 1967), i.e., intersecting each day's 08:00 radiosonde potential temperature (θ) profile at Beijing Observatory with each hour's (from 09:00 to 15:00) surface θ values, which are calculated from 203 204 surface air temperature observations. As shown in Fig. 4, the BLH on the days following a strong stable night (i.e., Node 1) is relatively flat, reflecting an inadequate development of daytime boundary layer. Similarity, Node 3 is 205 206 also followed by a flat daytime BLH variation. The maximum BLH in these two types are lower than 900 m, 207 indicating a limited space for vertical mixing in the day. In contrast, the afternoon BLH in Node 7 can reach up to 1100 m; this mixing depth is conducive to dilute the pollutants accumulated in the previous night. In Node 1, the 208 209 convective boundary layer develops well, and its maximum height on average exceeds 1500 m, far higher than the 210 values in other types.



Figure 4. Daytime boundary layer height (BLH) estimated for the four typical ABL types (i.e., Nodes 1, 3, 7 and 9).

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215 3.2 Implementing the SOM-based ABL classification scheme for urban air quality assessment

216 In the previous section, it was seen that the SOM classification scheme is an effective tool for delineation between various dynamic and thermodynamic structures within the ABL. As a further evaluation, we implement 217 the new classification scheme to quantify changes in various urban pollutant concentrations as a function of ABL 218 219 types. Since the pollutant emissions have a strong seasonality over Beijing and its surroundings, the analyses are 220 performed for winter (December to February), spring (March to May), summer (June to August), and autumn 221 (September to November), respectively. Figure 5 shows the statistical distributions of daily pollutant 222 concentrations according to the nine ABL types, along with the results of Kruskal-Wallis test and the coefficients 223 of variation of pollutant means across the various types. Figure 6 displays the type-average pollutant diurnal 224 patterns composited for the four typical ABL types (i.e., Nodes 1, 3, 7 and 9).

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226 The Kruskal-Wallis test demonstrates that the self-organized ABL types are able to distinguish between high and low loadings of air pollutants, with KW < 1% in all seasons (except for summertime SO₂ with a KW value of 227 1.5%). Furthermore, it is found that the SOM technique has a stronger discriminative power for SO_2 , $PM_{2.5}$ and 228 CO assessments, which is supported by relatively higher CV values (CV>0.30). According to the seasonal CV 229 values, this discriminative power shows a following seasonal ordering: winter > autumn > spring > summer. 230 231 Particularly, the wintertime CV value in $PM_{2.5}$ assessment reaches the maxima (0.56), indicating an extremely strong dependence of $PM_{2.5}$ air quality on the changing ABL meteorology in winter. In summer, the stable 232 233 nocturnal ABL develops later due to the longer day (Li et al., 2012), and hence avoids the larger daytime pollutant emissions, particularly the traffic peak emissions. In the absence of larger sources, the nocturnal stable layers exert 234

a limited influence on near-surface air quality; therefore, the classified ABL types have relatively weakened
discriminative power for summertime pollution assessments. In addition, wet depositions (more precipitation in
summer) play an important role in modulating summertime air quality, and to some degree disrupt the linkages
between ABL type and air quality.

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Figure 5. Daily concentrations of (a) $PM_{2.5}$, (b) O_3 , (c) NO_2 , (d) SO_2 , and (e) CO in Beijing for all nine ABL types separately in (1) winter, (2) spring, (3) summer, and (4) autumn. The solid dots denote the mean. The box and whisker plot presents the median, the first and third quartiles, and the 5th and 95th percentiles, respectively. The upper numbers denote the results of Kruskal-Wallis test (*KW*) and the coefficients of variation (*CV*) of pollutant means across the various types.

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In the case of $PM_{2.5}$, NO_2 and CO, the most stable atmospheric conditions (i.e., Node 9) are associated with

248 dramatically increased near-surface pollutant concentrations in all seasons except summer. The wintertime average concentrations of PM_{2.5}, NO₂ and CO in Node 9 reach up to 197.2 µg/m³, 100.2 µg/m³ and 3.6 mg/m³, 249 respectively. These values are 3-8 times higher than that in Node 1 (i.e., near neutral condition), with the highest 250 251 increasing amplitude (a factor of 7.3) related to PM2.5. As have known, Node 9 corresponds to the strongest 252 nighttime stability in the lower ABL and the lowest daytime BLH. All of these ABL characteristics are extremely conducive to the accumulation of air pollutants emitted near the ground. For Node 3, the concentrations of PM_{2.5}, 253 NO₂ and CO are the second-highest compared to those of the other types. This ABL type features the strongest 254 255 stability in the upper ABL, suggesting that processes operating at the different heights throughout the ABL may 256 have a significant impact on near-surface pollutant concentrations.

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The diurnal cycles of PM_{2.5}, NO₂ and CO are extremely pronounced under the strong stable conditions, 258 259 although very reduced on the days with near neutral night. On average, the wintertime diurnal range of PM_{2.5} 260 increases from 18.2 μ g/m³ in Node 1 to 95.4 μ g/m³ in Node 9. The corresponding diurnal range increase for NO₂ is 18.9 to 33.6 μ g/m³, and for CO 0.2 to 1.7 mg/m³. In Node 1, the diurnal variations are characterized by a weak 261 two-peak pattern following the traffic rush hours, suggesting that traffic is the primary driver of these pollutants' 262 263 diurnal cycles in Beijing (Liu et al., 2012). However, the diurnal effects of traffic emissions are significantly 264 amplified by the stable ABL dynamics. It is clear that the more stable conditions near the ground, the higher peak 265 concentrations are observed. In winter, the stable ABL conditions exert a more important influence on the evening traffic emissions, resulting in a broad evening peak. In contrast, the morning peak signature is much lower since 266 267 the morning emission is counteracted by the destabilization of the ABL. However, as human activities begin earlier during the warm season, maximum concentrations in spring and summer are typically observed during the 268 morning rush hours. 269

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However, increasing atmospheric stability has the opposite effect on near-surface O₃ concentrations. Since 271 272 aerosols can absorb and reflect solar radiation and thereby inhibit the photochemical production of O_3 (Gao et al., 2016;Kaufman et al., 2002), the lowest average O₃ concentration is observed in Node 9. In addition, considering 273 that ozone is mainly produced in the upper ABL, near-surface O₃ should be strongly modulated by down-mixing 274 275 processes (Tang et al., 2017b; Tang et al., 2017a). In this light, the varying daytime O₃ peaks across the ABL types can be partly attributed to the various magnitudes of vertical mixing. This is supported by daytime BLH. As have 276 shown in Fig. 4, the daytime BLH is highest in Node 1, followed by Node 7, Node 3 and the lowest in Node 9. 277 278 Such ordering is generally consistent with the daytime O₃ peaks in these types. Due to the persistent down-mixing

caused by strong wind shears, the near-surface O_3 remains a relatively high nocturnal concentration (e.g. about 45 $\mu g/m^3$ in winter) in Node 1. In contrast, the stable nocturnal conditions (e.g., Nodes 9, 7 and 3) are commonly associated with low O_3 concentration (e.g. about 16 $\mu g/m^3$ in winter) due to the lack of vertical mixing, as well as the strong chemical titration by NO emitted from vehicles.





Figure 6. Mean hourly composites of (a) PM_{2.5}, (b) O₃, (c) NO₂, (d) SO₂, and (e) CO in Beijing for the four typical ABL types separately in (1) winter, (2) spring, (3) summer, and (4) autumn

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The highest average SO_2 concentrations are typically observed in Node 3, but occasionally in Node 9. Over the North China Plain high stacks are emitting a significantly larger amount of SO_2 compared to small stacks (Zhao et al., 2012). The frequent surface temperature inversions together with the large SO_2 emissions from higher stacks favor the formation of elevated SO_2 pollution layers over Beijing (Chen et al., 2009). If sufficiently strong, the 292 surface temperature inversion can even isolate near-surface observations from the influence of elevated pollution 293 layers (Salmond and McKendry, 2005). This explains the commonly lower near-surface SO₂ concentration in 294 Node 9 than that in Node 3. However, after a stable night, the burst of turbulent activity in the morning coincides 295 with a rapid increase in near-surface SO₂ concentration, resulting in a pre-noon peak. Since there is no significant 296 increase in SO_2 emission at the surface at that time, the result strongly suggests that the SO_2 peaks are due to the 297 downward mixing from the elevated SO₂ pollution layers. Regarding the physical mechanism of the noontime-peak SO₂ pattern, Xu et al. (2014) have made a detail explanation in a previous study. Nevertheless, the 298 299 wintertime SO₂ concentration signature does not always show a distinct pre-noon peak (e.g., Node 7). This may 300 attribute to the increased SO₂ emissions from household heating in winter (Liao et al., 2017). Like other primary 301 pollutants, the local SO₂ emissions become trapped close to the surface under the stable nocturnal condition, 302 resulting in a much higher nighttime peak compared to the pre-noon peak.

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304 **3.3** Quantifying the contribution of ABL anomaly to typical-month PM_{2.5} air quality

To improve air quality, the Chinese government promulgated "Air Pollution Prevention and Control Action Plan" 305 in 2013. As a consequence, observed annual mean PM2.5 concentrations decrease by about 37% over Beijing 306 307 during 2013-2017. However, severe wintertime PM_{2.5} pollution events still frequently wreaked havoc across 308 Beijing and its surroundings, which resulted in severe damages to the environment and human health (Gao et al., 309 2017;Gao et al., 2015). It is therefore pressing to understand the factors affecting the occurrence of such serious PM_{2.5} pollution. Previous studies highlighted potential importance of atmospheric conditions to the wintertime 310 PM_{2.5} air quality (Cai et al., 2017). Since the fraction of time for which the different atmospheric conditions 311 dominate can vary from year to year, elucidation of the meteorological roles in those serious pollution periods has 312 a significant importance. In this section, we evaluate the contribution of ABL anomaly to elevated PM_{2.5} 313 314 concentration in three typical pollution months, i.e., January of 2013, December of 2015 and December of 2016.

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Heavy PM_{2.5} pollution episodes occurred frequently in January of 2013, December of 2015 and December of 2016, resulting in anomalously high month-averaged PM_{2.5} concentrations in the Beijing urban area (180.1 μ g/m³, 151.8 μ g/m³ and 151.2 μ g/m³, respectively). Figure 7 shows the hourly PM_{2.5} variations in the three selected months, along with daily ABL types. In general, the pollution episodes were associated with Nodes 3 and 9, and the clean episodes corresponded to Node 1. For example, the severe pollution episode that occurred from 9–14 January 2013 was due to the alternate control of Nodes 3 and 9, and the pollution episode from 15–21 December 2016 was related to the persistency of Node 9. Conversely, multiday control of Node 1 caused a clean episode

from 14–16 December 2015. The linkages between $PM_{2.5}$ air quality and ABL type are consistent with the previous long-term analyses, indicating that the changing ABL type is one of the primary drivers of day-to-day variations in wintertime $PM_{2.5}$ air quality over Beijing.





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Figure 7. Time series of hourly PM_{2.5} concentrations in (a) January of 2013, (b) December of 2015, and (c) December of 2016. The daily ABL types (i.e., SOM nodes) are shown at the top of each plot (red numbers).

Figure 8 illustrates a comparison of the occurrence frequency of the nine ABL types between the three selected months and the whole 5-year winter period. Compared with the winter-averaged frequency, notable differences are that the stable ABL conditions increased and the near neutral nights decreased during the three polluted months. For example, Node 9 occurrence was nearly trebled in December of 2016, and total occurrence of Nodes 3 and 9 doubled in January of 2013. These results highlighted potential contribution of ABL anomaly to the elevated PM_{2.5} concentrations in these pollution months.



Figure 8. Occurrence frequency of the ABL types (i.e., SOM nodes) during (a) January 2013, (b) December 2015, and (c) December 2016. The winter-averaged frequency during the 5-year (2013–2017) period is repeated on each plot for comparison.

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343 Assuming the linkages between ABL type and PM_{2.5} loading are constant in different years, the contribution of 344 the anomalous ABL meteorology to PM_{2.5} air quality can be estimated through a meteorology-to-environment 345 method, which is revised from the circulation-to-environment method proposed by Zhang et al. (2012). For each selected month, we define the deviation in $PM_{2.5}$ from the 5-year winter-averaged concentration (C_{WIN}) as the total 346 347 anomaly (C), which is due to the combined effects of emission and meteorology. The anomaly calculated from the mean PM_{2.5} loadings for nine ABL types and their occurrence frequencies during each month can be 348 considered as the "ABL-driven" anomaly, which represents the PM2.5 change caused by the anomalous ABL 349 meteorology. The ABL-driven anomaly (C_{ABL}) is calculated through $\sum_{i} F_i \cdot C_i - C_{WIN}$, where F_i is the 350 351 occurrence frequency of type-i ABL during a specific month and C_i is the corresponding PM_{2.5} loading featuring that type. The ratio of C_{ABL} to C' is then used to assess the relative contribution of the ABL anomaly to the total 352 anomaly. The calculated results (Table 1) shows that the ABL-driven $PM_{2.5}$ changes are 44.4 μ g/m³ in January 353 2013, 22.2 μ g/m³ in December 2015, and 34.6 μ g/m³ in December 2016, which explain 58.3%, 46.4% and 73.3% 354 355 of total anomaly in respective months. These quantitative estimations demonstrate that the elevated PM_{25} 356 concentrations during the three polluted months can be largely attributed to anomalous ABL conditions.

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362 Table 1. Estimated contribution of ABL anomaly to elevated PM_{2.5} concentration in January of 2013,

363 December of 2015 and December of 2016.

Pollution month	Month-averaged	Total anomaly C'	ABL-driven	Contribution ratio of
	PM _{2.5} concentration	$(\mu g/m^3)$	anomaly C_{ABL}	ABL-driven
	$(\mu g/m^3)$		$(\mu g/m^3)$	anomaly (%)
January 2013	180.1	76.1	44.4	58.3
December 2015	151.8	47.8	22.2	46.4
December 2016	151.2	47.2	34.6	73.3

365 **4. Summary**

The influence of ABL meteorology on Beijing's air quality is relatively unclear due to the lack of long-term observations. Meanwhile, the long years of routine radiosondes remain underutilized as a tool for urban pollution studies. In this study, the SOM was applied to 5-year (2013–2017) radiosonde-based θ_v profiles to classify the state of ABL over Beijing. The classified ABL types were then evaluated in relation to near-surface air quality, with an attempt to understand the roles of the changing ABL structure in air quality variation in Beijing. The main findings are as follows:

1) The SOM provides a continuum of nine ABL types (i.e., SOM nodes), and each is characterized with distinct dynamic and thermodynamic conditions within the ABL. Node 1 represent a near neutral layer with the lowest θ_v gradient and the highest wind speed. Node 3 features a strong static stability in the upper ABL. In contrast, Node 9 and Node 7 respectively correspond to the moderate and strong static stability in the lower ABL.

377 2) The self-organized ABL types are capable of characterizing the influence of nocturnal mixing on near-surface pollutant loadings. From the near neutral (i.e., Node 1) to strong stable conditions (i.e., Node 9), the average 378 379 concentrations of PM_{2.5}, NO₂ and CO increased dramatically during all seasons except summer. Meanwhile, 380 the diurnal cycles of these pollutant species are strongly modulated by ABL dynamics. Although the 381 modulation effect varies from season to season, the higher peak concentrations commonly occur under the 382 more stable conditions. However, increasing stability has opposite effect on O_3 , resulting in the lowest O_3 level in Node 9. For SO₂, the highest average concentrations are typically observed in Node 3. The pre-noon 383 SO₂ peaks are more significant after a strong stable night. 384

385 3) Analysis of three typical wintertime pollution months (i.e., January 2013, December 2015 and December
 386 2016) suggests that the ABL types are one of the primary drivers of day-to-day PM_{2.5} variations in Beijing.

387 During the three pollution months, the frequency of the stable ABL types (i.e., Nodes 9 and 3) increases 388 significantly compared to the 5-year winter mean. Using a meteorology-to-environment method, the relative 389 (absolute) contributions of the ABL anomaly to elevated $PM_{2.5}$ concentrations are estimated to be 58.3 % 390 (44.4 µg/m³) in January 2013, 46.4 % (22.2 µg/m³) in December 2015, and 73.3 % (34.6 µg/m³) in December 391 2016.

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This work revealed the common pattern of the ABL influences on Beijing' air quality. The established linkages 393 394 between ABL type and air quality could be useful for developing an operational forecast and warning system. In 395 addition, this work demonstrated that the SOM-based ABL classification scheme is a helpful tool for 396 understanding urban air pollution. Since the SOM technique is good at feature extraction, the coarse-resolution radiosondes can be taken as input to classify the state of the ABL. Therefore, the SOM-based ABL classification 397 scheme can take advantage of the long-term available radiosondes, making it a simple and economical alternative 398 to other approaches to stability classification. We believe that the pollution-related ABL research and the 399 formulation of pollution control measures could benefit from application of the SOM analytical tool. 400

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402 Data availability

The datasets used in this study are publicly available at the University of Wyoming (http://weather.uwyo.edu/), the
Ministry of Environmental Protection of the People's Republic of China (http://datacenter.mep.gov.cn/), the U.S.
Department of State Air Quality Monitoring Program (http://www.stateair.net/).

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407 **Competing interests**

408 The authors declare no conflict of interest.

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