1 Measurement report: Nocturnal subsidence behind the cold

2 front enhances surface particulate matter in the plain regions:

3 observation from the mobile multi-lidar system

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13 Abstract. A multi-lidar system, mounted in a vehicle to monitor the profiles of temperature, wind, and 14 particle optical properties, was utilized to investigate the winter fine particulate matter ($PM_{2.5}$) pollution 15 for from a vertical perspective, in four cities in China in winter 2018. We observed the enhancement of 16 surface nocturnal PM_{2.5} in two typical plain cities (Changzhou and Wangdu), which was attributed to the 17 subsidence of PM_{2.5} transported from upstream polluted areas, with the wind turning north and 18 downdrafts dominating. Combining with the observed surface PM_{2.5}, the reanalysis meteorological data, 19 and the GEOS-Chem model simulation, we revealed the Transport-Nocturnal $PM_{2.5}$ Enhancement by 20 Subsidence (T-NPES) events occurred frequently in the two cities, with percentages of 12.2 % and 21 18.0 %, respectively during Dec. 2018 - Feb. 2019. Furthermore, the GEOS-Chem model simulation 22 further confirmed that the ubiquity of winter T-NPES events in-on a large scale including North China 23 Plain and the Yangtze River Delta. Process analysis revealed that the subsidence was closely correlated 24 with the southeasterly movement of the high-pressure system and the passage of the cold front, resulting 25 in the increase of temperature aloft, a stronger inversion layer, and further PM_{2.5} accumulation in the 26 atmospheric boundary layer. Thus, a conceptual model of the T-NPES events was proposed to highlight 27 this surface PM_{2.5} enhancement mechanism in these plain regions. However, it was not applicable did not 28 apply to the two cities in the basin region (Xi'an and Chengdu), due to the obstruction of the weather 29 system movement by the mountains surrounding the basin.

30 1 Introduction

31 The severe fine particulate matter ($PM_{2.5}$, particles with an aerodynamic diameter smaller than 2.5 µm) 32 pollution, caused by the rapid industrialization and urbanization in China (Guo et al., 2014; Huang et al., 33 2014), has essential impacts on visibility, ecosystem, regional and global climates, and human health 34 (Yue et al., 2017; An et al., 2019; De Marco et al., 2019; Li et al., 2019b; Hao et al., 2021). To mitigate 35 the PM_{2.5} pollution, the government of China has implemented the Air Pollution Prevention and Control 36 Action Plan in 2013 by strict emission controls (Gao et al., 2020). Despite Despite the fact that the annual 37 average concentration of PM_{2.5} has been significantly decreased (Ding et al., 2019; Li et al., 2019a; Zhang 38 et al., 2019b; Silver et al., 2020; Geng et al., 2021b), the PM_{2.5} levels in the majority of Chinese cities 39 are still above the World Health Organization target (WHO. 2021). Particularly, the issue of PM_{2.5} pollution remained critical in the North China Plain (NCP) and Yangtze River Delta (YRD) in winter 40 41 time (Peng et al., 2021; Qin et al., 2021).

42 The formation mechanisms of PM_{2.5} pollution were complex especially in China (Guo et al., 2014; Xiao 43 et al., 2021b), such such as the high emission intensity (Zhang et al., 2019b), the rapid chemical 44 formation of secondary particles owing to the gas phase gas phase and heterogeneous reactions (Wang et 45 al., 2017; Lu et al., 2019; Chen et al., 2020), and the interactions within the atmospheric boundary layer (ABL) (Ding et al., 2013; Gao et al., 2016; Dong et al., 2017; Li et al., 2017). While the long-range 46 47 transport also had significant impacts on the PM₂₅ pollution (Guo et al., 2014; Zhang et al., 2015; Huang 48 et al., 2018). Cold fronts, as a common synoptic circulation in winter, were usually favourable favorable 49 for the quick removal of the locally accumulated pollutants in the NCP (Zhao et al., 2013; Gao et al., 50 2017), but conversely transport the pollutants to the YRD through a long distance (Kang et al., 2019; 51 Huang et al., 2020; Kang et al., 2021). Zhou et al. (2023) indicated that the cold fronts could transport 52 the precursors to the residual layer, where the secondary pollution was rapidly driven to be generated and 53 then exacerbate near-surface air pollution as a result of the development of the daytime convective ABL. 54 However, the above studies have focused on the impact of the horizontally transported pollutants on the 55 downstream regions after the passage of the cold front. While In comparison, few studies have been conducted on the variation in the vertical direction of particulate matter in the ABL during the passage 56 57 of the cold front.

The vertical mixing exchange process between layer has great <u>impacts_impact_on</u> local air quality and the subsidence motion is associated with the evolution of <u>the</u> inversion layer (Gramsch et al., 2014; Xu et al., 2018; He et al., 2022). Zhang et al. (2022) reported that the PM_{2.5} concentration behind the cold front increased due to the subsidence motion and inversion layer. Zhao et al. (2023) suggested that the frontal downdrafts were an additional transport pathway in the nighttime to make <u>a</u> higher contribution to the ground nitrate. Both of their studies were based on the model simulations, the observational evidence of the subsidence behind the cold front and its impact on the nocturnal $PM_{2.5}$ enhancement events is still lacking. Shi et al. (2022) reported one subsidence case of particulate matter during the passage of the cold front over Wangdu, China in winter, which revealed that the subsidence was closely connectwas closely connected to the enhancement of nocturnal $PM_{2.5}$.

68 To investigate the mechanisms of nocturnal $PM_{2.5}$ enhancement triggered by subsidence, the three-69 dimensional spatial and temporal distribution is crucial. Many field observations of the vertical 70 distribution of particulate matter have been performed employing various methods such as tethered 71 balloons (Wang et al., 2021; Ran et al., 2022), airplane-airplanes (Wang et al., 2018; Fast et al., 2022), 72 unmanned aerial vehicles (Song et al., 2021; Dubey et al., 2022) and the meteorological towers (Li et al., 73 2022; Yin et al., 2023). Lidar, as an active remote sensing device with high temporal and spatial 74 resolution, has been extensively employed in atmospheric detection to obtain the profile of particulate 75 matter, wind, and temperature. The ground-based and satellite-based lidar have been widely used to 76 detect the vertical distribution of aerosol. In recent years, the mobile multi-lidar system has been 77 gradually developed and has become a powerful tool to observe for observing the development of target 78 species detection in a vertical perspective. Compared with the traditional ground-lidar system, the mobile 79 multi-lidar system enables continuous mobile observations and provides information on the distribution 80 of specific factors along its path and can be used as an effective supplement to other fixed lidars. 81 Additionally, the mobile multi-lidar system can reach different cities by its portable setting in a short 82 time to carry out the fixed-point observations. The mobile lidar system had has been used to carry out 83 several observations in the past few years (Lv et al., 2020; He et al., 2021; Xu et al., 2022). He et al. 84 (2021) investigated the vertical distribution characteristics of particulate matter in the Guanzhong Plain 85 by using the mobile multi-lidar system. Xu et al. (2022) conducted an observational study on the three-86 dimensional structure of particulate matter distribution in the Guangdong-Hong Kong-Macao Greater 87 Bay Area by using the mobile multi-lidar system and proposed a conceptual model to elucidate the 88 vertical distribution of particulate matter under different wind and temperature conditions.

Here, we conducted the first nationwide field measurements in winter 2018 using the mobile multi-lidar
system during winter 2018 in China, to investigate the vertical distribution characteristics of particulate

91 matter in different cities. We focus on the observed nocturnal PM_{2.5} enhancement events and seek insights

into their characteristics and the causes, by combining with the GEOS-Chem model simulation, the surface $PM_{2.5}$ observation, and meteorological reanalysis dataset. Finally, we examine the ubiquity of this phenomenon in plain regions in China and propose a conceptual model, providing detailed vertical insights into the enhancement of nocturnal surface $PM_{2.5}$.

96 2 Data and methods

97 2.1 Multi-lidar system

A multi-lidar system was installed on the mobile observation vehicle. The vehicle, a modified 7-seater Mercedes-Benz sport utility vehicle, was equipped with three lidar instruments mounted on steel bars at the rear for stability. The mobile observation routes were primarily on flat highways, and the speed was controlled to remain around 80 km/h to minimize the impact of frequent changes in speed and vehicle bumps on the measurement results.

103 The multi-lidar system (Everise Technology Ltd., Beijing) consisted of a 3D visual scanning micro pulse 104 lidar (EV-Lidar-CAM), a twirling Raman temperature profile lidar (TRL20), a Doppler wind profile lidar 105 (WINDVIEW10), a global positioning system (GPS). The 3D visual scanning micro-pulse micropulse 106 lidar had a detection range of up to 30 km, a temporal resolution of 1 minute, and a vertical resolution of 107 15 m. The 3D lidar emitted a 532 nm laser beam vertically, which is scattered by aerosol particles in the 108 atmosphere The 3D lidar used an Nd: YAG laser to emit a 532 nm laser beam at a repetition frequency 109 of 2500 Hz, which is scattered by aerosol particles in the atmosphere. The backscattered signal is utilized 110 to calculate the aerosol extinction coefficient and depolarization ratio profile. The extinction coefficient 111 increases with higher particle pollution concentrations, while the depolarization ratio can distinguish 112 between spherical and non-spherical particles based on their size and shape. The Doppler wind profile 113 lidar provides a temporal resolution of 1 minute and a vertical resolution of 50 m. It emits a rotating 1545 114 nm laser beam using a 10 kHz repetition rate fibre pulse fiber-pulse laser and measures the Doppler shift 115 produced by the laser's backscattered signal as it passes through airborne particles such as dust, water 116 droplets in clouds and fog, polluted aerosols, salt crystals, and biomass-burning aerosols to derive the 117 horizontal and vertical wind speeds at any height. The Raman temperature profile lidar, based on Raman 118 scattering theory, calculates atmospheric temperature by detecting the rotational Raman scattering signal 119 of nitrogen or oxygen molecules in the atmosphere. Operating at a 532 nm wavelength with an Nd: YAG 120 laser at a repetition frequency of 20 Hz, it has a temporal resolution of 5 minutes and a vertical resolution 121 of 60 m. The quality of the data obtained by the lidar system was checked by the Integrated 122 Environmental Meteorological Observation Vehicle before deployment. The results showed a percentage 123 difference of less than 15% between the lidar system data and the data provided by the Shenzhen 124 Meteorological Tower, demonstrating the high accuracy of the lidar instrument (Xu et al., 2022). Data 125 during the instrument malfunction, below the blind zone, and in rainy weather had been excluded. 126 Previous studies had-have utilized this lidar system and demonstrated its reliability (Xu et al., 2018; He 127 et al., 2021). The mobile observation vehicle and multi-lidar system are shown in Figure 1(a). The more

128 details of the multi-lidar system are shown in the Table S1.



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131 Figure 1. (a) The mobile observation vehicle and multi-lidar system. (b) The For the mobile observation route and 132 stopover cities, the blue dotted line shows the sections of missing data.

133 2.2 The route of nationwide mobile observation

134 To investigate the distribution characteristics of particulate matter during winter in different regions in 135 China, the Integrated Environmental Meteorological Observation Vehicle of Sun Yat-sen University was 136 utilized to conduct the first nationwide mobile observation campaign. The campaign, which lasted 43 137 days and covered approximately 11,000 km, started in Shenzhen on 30 November, 2018 and ended in 138 Guangzhou on 11 January, 2019. This campaign surveyed the PM_{2.5} vertical profiles across 15 cities, 139 including Shenzhen, Xiamen, Wenzhou, Changzhou, Zhengzhou, Wangdu, Beijing, Shijiazhuang, 140 Linfen, Xi'an, Baoji, Chengdu, Enshi, Changsha and Guangzhou. The observation route and stopover 141 cities are shown in Figure 1(b). Due to the precipitation, there were no observations between Shenzhen-142 Xiamen and Wenzhou-Changzhou, while some GPS data were missing between Beijing-Chengdu and 143 Enshi-Changsha.

To compare the vertical distribution characteristics of particulate matter in different regions, we conducted fixed-point observations for several pollution days in four representative cities in the East China region (Changzhou), North China Plain (Wangdu), Guanzhong Basin (Xi'an), and Sichuan Basin (Chengdu). The dates and duration of the fixed-point observations are presented in Table 1. In the following analysis, only the data obtained in the four fixed-point measurements are used since it has an enough time duration to show the vertical variation of PM_{2.5}.

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Table 1. Date and cities of fixed-point observations

Date	Cities	Coordinate	Landform
2018.12.11-2018.12.14	Changzhou	119.97°E, 31.83°N	Plain area
2018.12.18-2018.12.22	Wangdu	115.25°E, 38.67°N	Plain area
2018.12.31-2019.01.02	Xi'an	109.01°E, 34.22°N	Basin area
2019.01.04-2019.01.09	Chengdu	103.92°E, 30.58°N	Basin area

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152 2.3 Surface PM_{2.5} data and ERA5 reanalysis data

153 The nationwide hourly observations of surface PM_{2.5} in China are obtained from the China National

154 Environmental Monitoring Center (CNEMC) network (https://quotsoft.net/air, last accessed: March 2nd,

155 2023). Here, we used the hourly $PM_{2.5}$ concentration data from the whole winter of 2018 (Dec. 2018 – 156 Feb. 2019) and selected data from the closest monitoring station to show the change in surface $PM_{2.5}$ 157 concentration at the four observation sites.

158 The spatial distribution of daily average surface PM_{2.5} concentration is obtained from the TAP team 159 (http://tapdata.org.cn), with a spatial resolution of 10 km. Based on machine learning algorithms and 160 multi-source data information, the TAP team has built a multi-source data fusion system that integrates 161 ground observation data, satellite remote sensing information, high-resolution emission inventories, air 162 quality model simulations, and other multi-source information (Geng et al., 2021a; Xiao et al., 2021a). 163 In addition to the observation data, we also apply the three-dimensional meteorological data from the ERA5 dataset for the winter of 2018 (https://quotsoft.net/air, last accessed: March 2nd, 2023) (Munoz-164 165 Sabater et al., 2021). This dataset contained temperature, horizontal and vertical wind speed, and 166 direction at pressure levels, as well as two-dimensional data including sea-level pressure and 2-m 167 temperature. The ERA5 dataset is the fifth generation of the European Centre for Medium-Range 168 Weather Forecasts (ECMWF) atmospheric reanalysis of the global climate. The ERA5 dataset has a 169 horizontal resolution of $0.25^{\circ} \times 0.25^{\circ}$, a vertical resolution of 25 hPa, and a temporal resolution of 1 h.

170 2.4 HYSPLIT backward trajectory model

The Hybrid Single Particle Lagrangian Integrated Trajectory Model (HYSPLIT) (Stein et al., 2015), developed by NOAA Air Resources Laboratory, which is a valuable tool for simulating the movement of air mass and the transport of pollutants in the atmosphere, is used in our study to obtain the sources of particulate matter at different heights. Altitudes of 100, 500, and 1000 m were set as the end pointsendpoints of the trajectories, the meteorological input for the trajectory model was the FNL dataset, and each trajectory was calculated for 24 h duration.

177 2.5 GEOS-Chem model description

178 Given the short-term (less than one week) fixed-point observation duration of the mobile observation 179 vehicle in each city, we employ the global three-dimensional chemical transport model GEOS-Chem 180 version 13.3.1 to interpret the vertical observations (available at 181 https://github.com/geoschem/GCClassic/tree/13.3.1, last assessed: March 2nd, 2023, (Bey et al., 2001)) 182 and to simulate the distribution of particulate matter concentrations during winter 2018 in China. We 183 perform the nested-grid version of the GEOS-Chem simulation at a spatial of 0.5° (latitude) $\times 0.625^{\circ}$

184 (longitude) resolution over East Asia (60-150°E, 11°S-55°N), The model has 47 vertical layers with 18 185 layers in the below 3 km. Boundary chemical conditions for the nested models are archived from a consistent global simulation run at 4° latitude $\times 5^{\circ}$ longitude resolution. Meteorological input is from the 186 187 Modern-Era Retrospective analysis for Research and Application version 2 (MERRA-2) (Gelaro et al., 188 2017). We conduct the model simulation from 2018/11-2019/02 with the first one-month as a spin-up. 189 The model mechanisms and emissions mostly follow our previous study (Wang et al., 2022). In short, 190 the GEOS-Chem model describes a comprehensive stratospheric and tropospheric ozone-NO_x-VOCs-191 aerosol-halogen chemical mechanism (Wang et al., 1998; Park et al., 2004; Parrella et al., 2012; Mao et 192 al., 2013). Photolysis rates are computed using the Fast-JX scheme (Bian and Prather. 2002). Advection 193 of tracers in GEOS-Chem is accomplished through the TPCORE advection algorithm. Boundary The 194 boundary layer mixing process is described in (Lin and McElroy. 2010). Dry and wet deposition of both 195 gas and aerosols is considered (Wesely, 1989; Zhang et al., 2001). We apply the latest version of the 196 Community Emissions Data System (CEDSv2) anthropogenic emissions inventory (O'Rourke et al., 197 2021), in which the emissions over China have been adjusted to align with the Multi-resolution Emission 198 Inventory for China (MEIC) inventory (Zheng et al., 2018). 199 Figure S1 showed the comparison of model results with observations for monthly mean $PM_{2.5}$, and the

200 correlation coefficients between model and observation were about 0.6, which meant that the model

201 results provided a relatively good reproduction of the observations.

202 3 Results and discussions

203 **3.1 The observation of nocturnal PM2.5 enhancement in plain areas**

204 During the fixed-point observation in Changzhou, we observed a typical surface PM_{2.5} concentration 205 enhancement event starting at 4:00 and lasting until 10:00 on 13 December. As shown in Figure 2(a), the 206 concentration of $PM_{2.5}$ increased from 69 to 151 µg/m³. Figure 2(b-c) showed shows the spatiotemporal 207 distribution of the extinction coefficient and depolarization ratio. There was a clear layer with a low 208 extinction coefficient below 500 m from 16:00 on 12 December to 4:00 on 13 December, indicating low 209 $PM_{2.5}$ concentration near the surface. Meanwhile, an aerosol layer with a high extinction coefficient of 210 about 0.7 km⁻¹ appeared at 500-1,000 m. Figure 2(d-e) depicted the west winds prevailed prevailing in 211 the layer of 500-1,000 m with a wind speed (WS) of about 7 m/s. Based on the daily averaged-average 212 concentration of PM_{2.5} on 12 December shown in Figure <u>S2S1</u>, the western area of the observation

site in Changzhou suffered from severe air pollution with the concentration of $PM_{2.5}$ exceeding 150 μ g/m³.

214 Under the strong forcing of the west winds, the regional transport of aerosol from the west of Changzhou

215 was detected, leading to a high extinction coefficient layer at 500-1,000 m. The spatiotemporal

216 distribution of the vertical velocity in Figure 2(e) indicated the dominant updraft winds in the ABL,

217 which was conducive to the suspension of pollutants at 500-1,000 m.

However, the prevailing winds at 500-1,000 m shifted to the northwest/north after 4:00 on 13 December. By 8:00, the north wind dominated in the ABL. The change in wind direction affected the transport process of pollutants at 500-1,000 m, after which the transport basically disappeared. Meanwhile, the downdraft winds dominated above 500 m (Figure 2(e)) and the aerosol layer suspended at the-500-1,000 m began to gradually transport and diffuse downward into the lower layer of ABL, which enhanced the nocturnal surface $PM_{2.5}$ concentration. Noteworthy, after 4:00 on 13 December, the surface temperature was close to the temperature at 950 hPa, suggesting that the structure of the ABL was stable and was

225 conducive to the accumulation of the $PM_{2.5}$.

226 The sea surface field showed the cold high-pressure system moved southeast with increasing strength 227 from 20:00, 12 December to 8:00, 13 December (Figure \$3\$2). The change in the synoptic weather 228 system was accompanied by a cold frontal passage. The cold frontal passage was inferred to start at about 229 4:00, 13 December and last about 4 hours, which was further illustrated by the clockwise rotation of the 230horizontal wind from the ground to the upper layer (Shi et al., 2022) and the transition from updrafts to 231 downdrafts, the observation site was located behind the cold front after 4:00 where the descending 232 movements dominated. Under the influence of the subsidence, the pollutants transported by the west 233 advection diffused downward to the low layer and further aggravated the local air quality.





Figure 2. (a) Surface PM_{2.5} concentration, surface temperature and 950 hPa temperature, (b) Extinction coefficient,
(c) Depolarization ratio, (d) Horizontal wind, and (e) Vertical wind, during the observation in Changzhou from 12
December to 14 December. The black box indicated the nocturnal surface PM_{2.5} enhanced event.

238 After 8:00, the concentration of surface PM_{2.5} increased rapidly and peaked at around 10:00, the extinction coefficient below 1,000 m also reached a high level with 1.0 km⁻¹ at the same time and the 239 240 depolarization remained at about 0.01. The surface temperature began to rise and the convective ABL 241 developed rapidly, which enhanced the vertical mixing and resulted in the rapid increase in surface PM_{2.5} 242 (Zhou et al., 2023). And the north winds following the passage of the cold front dominated in the ABL 243 after 8:00, which could bring pollution from the NCP to the YRD (Kang et al., 2019; Huang et al., 2020). 244 Therefore, we attribute the increase in the concentration of surface $PM_{2.5}$ from 4:00 to 10:00 to the 245 combination of the subsidence behind the cold front before 8:00, vertical mixing caused by the 246 development of the convective ABL, and the transport by the north winds.

247 We also found similar nocturnal surface $PM_{2.5}$ enhancement events during the fixed-point observation in 248 Wangdu, on 19 December and 21 December respectively (Figure 3(a)). The concentration of PM_{2.5} 249 started to enhance at 1:00, 19 December, and meanwhile, the layer of pollutants above 1,000 m started 250 to transport and diffuse to the lower layer of ABL which was reflected by the change of the extinction 251 coefficient shown in Figure 3(b). Unfortunately, due to the instrument malfunction, the wind profile data 252 was unavailable and we used the ERA5 data instead, which previously showed good consistent 253 consistency with the observation of with Doppler wind lidar (Shi et al., 2022). As shown in Figure 3(d), 254 from 10:00, 18 December to 0:00, 19 December, southwest winds prevailed above 1,000 m and the WS 255 exceeded 8 m/s, a persistent southerly wind could result in severe air pollution in the NCP (Cai et al., 256 2017; Callahan et al., 2019; Zhang et al., 2019a). The wind forced the regional advection of pollutants 257 from the south region suffered suffering from serious air pollution (Figure S4S3) to the observation site. 258 Meanwhile, the updrafts dominated in the ABL which facilitated the suspension of pollutants in the upper 259 layer. After 0:00, 19 December, as the cold high-pressure system moved southwest accompanied by a 260 cold front (Figure <u>\$5</u>\$4), the prevailing winds above 1,000 m shifted to the northwest gradually and 261downdrafts dominated behind the cold frontal passage. The changes in the horizontal and vertical wind 262 fields caused the advection of pollutants to disappear basically and the pollutants layer suspended above 263 1,000 m began to transport and diffuse downward to the low layer of ABL. The passage of the cold front 264 at 0:00, 19 December, lasted for 4 hours, and the subsidence behind the cold front caused the pollutants 265 to diffuse downward, enhancing the concentration of nocturnal PM_{2.5}.





Figure 3. (a) Surface PM_{2.5} concentration, surface temperature, and 950 hPa temperature, (b) Extinction coefficient,
(c) Depolarization ratio, (d) Horizontal wind, and (e) Vertical wind, during the observation in Wangdu from 18
December to 21 December. The black boxes indicated the Nocturnal PM_{2.5} enhancement events.

270 The pattern of the nocturnal surface $PM_{2.5}$ enhancement event on 21 December was highly similar to that 271 on 19 December. However, the pollutant advection process lasted a longer duration which started at 272 16:00, 20 December and ended at 0:00, 21 December (Figure 3(b)), and the WS of the southwest wind above 1,000 m exceeded 12 m/s meeting the standard of the low-level jet (LLJ) (Kraus et al., 1985; Hu 273 274 et al., 2013). The area south of the observation site in Wangdu suffered from more severe air pollution 275 with the concentration of PM_{2.5} exceeding 300 μ g/m³ (Figure S685). Under the strong forcing of the 276 southwester-southwestern LLJ and the updrafts depicted in Figure 3(d-e), an aerosol layer with high 277 extinction coefficient exceeding 2 km⁻¹ formed and was suspended at 1,000-1,500 m from 16:00, 20 278 December to 0:00, 21 December. Meanwhile, Figure 3(c) showed that the layer with low depolarization was consistent with the layer with <u>a high extinction coefficient</u>, further <u>confirmed confirming</u> the role of
 transportation.

281 After 0:00, the wind direction of LLJ began to change due to the southeasterly movement of the high-282 pressure system accompanied by a cold front (Figure S7S6). The passage of the cold front started at 0:00, 283 21 December, and lasted for 4 hours, after which the downdrafts dominated below 1,500 m (Figure 3(e)), 284 and the northwester northwestern LLJ no longer transported pollutants from the southern area but greatly 285 enhanced the turbulent mixing (Shi et al., 2022). Under the influence of the turbulence generated by LLJ 286 and subsidence behind the cold front, the aerosol-rich layer suspended at 1,000-1,500 m was gradually 287 transported and diffused downward into the lower layer of ABL, ultimately enhancing the concentration 288 of surface $PM_{2.5}$, which was consistent with the result reported by Shi et al. (2022), with the secondary 289 inorganic aerosol increasing simultaneously during the subsidence process as observed by the tethered 290 balloon.

Noteworthy, when both nocturnal surface $PM_{2.5}$ enhancement events in Wangdu occurred, the temperature at 950 hPa showed an increasing trend as a result of the heating of the air by compression as it descended, while the surface temperature continuously declined (Figure 3(a)). The opposite variation of surface temperature and temperature at 950 hPa stabilized the lower atmosphere. The stronger inversion layer was probably induced by subsidence (Carlson and Stull. 1986). With the more stably stable atmospheric layer and inversion during subsidence, the concentration of surface PM_{2.5} enhanced (Gramsch et al., 2014; Largeron and Staquet. 2016).

3.2 Transport-Nocturnal PM_{2.5} Enhancement by Subsidence events

299 During the fixed-point observation, we found the causes of three nocturnal PM_{2.5} enhancement events in 300 different cities were similar. The processes included can be summarized in the following as three steps: 301 First, the horizontal winds with high wind speed forced the transport of pollutants from the upstream 302 region, while the updrafts dominated, both resulting in the formation and suspension of an aerosol layer 303 with high extinction coefficient at the high layer of the ABL. Then, under the influence of the 304 southeasterly movement of the high-pressure system and the passage of the cold front, the horizontal 305 wind direction shifted to the north or northwest and the downdrafts became dominant. Finally, the 306 transport of pollutants disappeared due to the change of wind direction, and under the subsidence behind 307 the cold front, the aerosol-rich layer suspended at the high layer was gradually transported and diffused 308 downward into the lower layer of the ABL, ultimately enhancing the concentration of nocturnal PM_{2.5}.

309 Here, we defined this pollution pattern as T-NPES (Transport-Nocturnal $PM_{2.5}$ Enhancement by 310 Subsidence) events.

311 To investigate the occurrence frequency of T-NPES events, we employed the GEOS-Chem model to 312 simulate the distribution of particulate matter concentrations in China during the whole winter of 2018 313 (Dec. 2018 – Feb. 2019). We utilized the simulated PM_{2.5} at 950 hPa and 900 hPa to represent the high-314 altitude PM_{2.5} concentration. We selected the closest grid data of the wind field data, 950 hPa₂ and 2-m 315 temperature data from the ERA5 dataset to the observation site in Changzhou and Wangdu to show the 316 meteorological condition. By analysing analyzing the hourly concentration variation of $PM_{2.5}$ and the 317 distribution of the wind field during the three months of winter 2018 in Changzhou and Wangdu, we 318 found 11 typical T-NPES events in Changzhou accounting for 12.2% and 18 T-NPES events in Wangdu 319 accounting for 18%, which indicated that the T-NPES events were a relatively common phenomenon in 320 the two cities.

321 Figure 4 showed shows the average pattern of all T-NPES events in Changzhou, the trend of the 322 simulated $PM_{2.5}$ was consistent with the observation, confirming the credibility of the simulations. As 323 shown in Figure 4(a), the enhancement of nocturnal surface $PM_{2.5}$ started at 21:00, when there was no 324 significant enhancement in anthropogenic $PM_{2.5}$ emissions, while the high-altitude $PM_{2.5}$ represented by 325 $PM_{2.5}$ at 900 hPa and 950 hPa started to decrease, which was consistent with the observed event in 326 Changzhou described in Section 3.1. According to the distribution of the wind field (Figure 4(b)), west 327 winds with high wind speed prevailed in the layer above 1,000 m from 0:00 to about 18:00, which was 328 conducive to the transport of pollutants. And the The updrafts dominated from 0:00 to 12:00, forcing the 329 pollutants suspending in the upper layer, which was reflected by the enhancing $PM_{2.5}$ concentration at 330 high altitude high altitude (Figure 4(a)). Despite the downdrafts dominated dominating after 12:00, there 331 was no immediate reduction in $PM_{2.5}$ concentration at high altitude high altitudes, which might be related 332 to the fact that the horizontal wind direction had not changed, and the transport of pollutants continued. 333 A brief updraft before 21:00 suspended the pollutants at high altitude high altitudes. After 21:00, 334 northwester winds and downdrafts dominated in the ABL and the high-altitude PM_{2.5} began to gradually 335 transport and diffuse downward causing the enhancement of surface concentration of PM_{2.5}, and this 336 process continued until 4:00 in the next day. The surface temperature and the temperature at 950 hPa 337 gradually approached, which is consistent with the observed case in Changzhou, indicating that the 338 structure of the ABL was stable and was conducive to the accumulation of the PM_{2.5}. As shown in Figure 339 \underline{S} , the average sea level pressure indicated that the southeasterly movement of the high-pressure

340 system and the passage of <u>the cold front</u>, which resulted in the shift in wind direction and subsidence
341 behind the cold front, were the main causes of the T-NPES events.



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Figure 4. The average for all T-NPES events in Changzhou. (a) The concentration of PM_{2.5} at different levels,
surface PM_{2.5} of observation (red line), surface PM_{2.5} of simulation (blue line), and PM_{2.5} at 900 hPa and 950 hPa.
(b) The horizontal winds (arrows), the vertical winds (shaded), the temperature at 2 m₂ and temperature at 950 hPa.

346 Figure 5 showed shows the average pattern of all T-NPES events in Wangdu, which was similar to that 347 in Changzhou. Figure 5(a) demonstrated that the trend of simulated PM_{2.5} was consistent with the 348 observation before 22:00 but was different thereafter. The trend of high-altitude PM_{2.5} was increasing 349 before 15:00 due to the transport of pollutants by prevailing southwester horizontal winds and the 350 dominant dominance of updrafts which suspended the aerosol shown in the Figure 5(b). After 18:00, the 351 prevailing winds began to turn to northwest and ultimately turn to north at 0:00 in the next day, while a 352 brief updraft between 18:00 and 20:00 suspended the pollutants at high altitude high altitude. The ABL 353 was dominated by the northwesternorthwestern winds and downdrafts after 21:00. Simultaneously, the 354 high-altitude PM_{2.5} began to gradually transport and diffuse downward causing the enhancement of 355 surface concentration of PM_{2.5}. The temperature at 950 hPa increased and the surface temperature 356 declined (Figure 5(b)), which agreed with the two observation examples in Wangdu. The opposite 357 variation of temperature at different heights stabilized the ABL and further enhanced the 358 concentration of PM_{2.5}. By analysing analyzing the weather circulation patterns, the causes of the T-

NPES events were the same with as those in Changzhou and were attributed to the southeasterly
movement of the high-pressure system and the passage of the cold front (Figure <u>S9</u>S8).

Overall, the average patterns of T-NPES events in Changzhou and Wangdu were essentially in good agreement with the three cases of T-NPES in the two cities. But there were still slight differences, such as the change of Wangdu caused by the movement of high-pressure lasted a longer time in the average situation and the start time of subsidence behind the cold front was also not consistent, which were due to each T-NPES event was not exactly the same.



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Figure 5. The average for all T-NPES events in Wangdu. (a) The concentration of PM_{2.5} at different levels, surface
 PM_{2.5} of observation (red line), surface PM_{2.5} of simulation (blue line), and PM_{2.5} at 900 hPa and 950 hPa. (b) The
 horizontal winds (arrows), the vertical winds (shaded), the temperature at 2 m₁ and temperature at 950 hPa

370 3.3 The universality of T-NPES events in eastern China

371 Despite the mobile observation vehicle had no observations in other cities of the NCP, the YRD_a and the 372 Loess Plateau, we could still utilize the simulated data and the ERA5 data to investigate the universality 373 of T-NPES events occurrence in other cities. We selected Shijiazhuang, Beijing_a and Tianjin as 374 represented cities of the NCP, Shanghai_a and Nanjing as represented cities of the YRD_a and Taiyuan, 375 Linfen as represented cities of the Loess Plateau. We found the <u>a</u> similar pattern of T-NPES events in all 376 these cities. However, these T-NPES events in different cities had some differences in detail. Here we 377 divided the T-NPES events into four types based on the status of PM_{2.5} after T-NPES events. More information on the types, frequency of the T-NPES events, and their percentage of the winter 2018 was
shown in Table 2.

380 The typical representation of Type 1 was is shown in Figure S10S9, the characteristic of Type 1 was that 381 the southwester-southwestern winds transported the pollutants in the high-altitude of the ABL, then the 382 wind direction shifted to north and downdrafts dominated, finally, pollutants in high-altitude diffused 383 into lower layer causing the surface PM_{2.5} enhanced. However, after the T-NPES event, the north wind 384 near the ground was not strong enough to remove the pollutants, causing the a high level of $PM_{2.5}$ lasting 385 the next day morning and may resulting result in aggravation of the air pollution in the following day. 386 The characteristic of the T-NPES event of Type 2 was basically consistent with Type 1. However, after 387 the T-NPES event, as north winds became stronger, pollutants were rapidly removed, resulting in a clean 388 boundary layer throughout (Figure S11S10). Even when the pollutants were removed more quickly by 389 stronger north winds, the subsidence process might not be observed. Type 1 and Type 2 were both 390 observed in the NCP cities, while Type 1 predominated in Wangdu and Shijiazhuang, and Type 2 in 391 Beijing and Tianjin.

Tuble 2. Statistics of the T TATES events in cities during Dec. 2010 Teb. 2017					
Area	Туре	City	Frequency (days)	Percentages	
	Type 1 and 2	Wangdu	18	20.0%	
NCP		Shijiazhuang	18	20.0%	
		Beijing	13	14.4%	
		Tianjin	14	15.6%	
YRD	Type 3	Changzhou	11	12.2%	
		Shanghai	7	7.8%	
		Nanjing	8	8.9%	
Loess Plateau	Type 4	Linfen	18	20.0%	
		Taiyuan	13	14.4%	

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Table 2. Statistics of the T-NPES events in cities during Dec. 2018 – Feb. 2019

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Figure <u>S12</u><u>S11</u> showed_shows_the typical representation of Type 3. The prevailing wind transporting pollutants was not southwest but west and the start and end of the T-NPES event were later than for Type 1 and 2. After the T-NPES event, the increase of 2-m temperature and the development of convective ABL led to the vertical mixing and the increase of surface PM_{2.5}. Additionally, the stronger north wind

might transport the pollutants from the NCP to the YRD. The Type 3 was similar to the example in
Changzhou in Section 3.1 and indicative of a typical pattern in the YRD cities.

400 The typical representation of Type 4, which was mainly occurred in the Loess Plateau cities, was shown 401 in Figure <u>\$13</u>\$12. During the T-NPES event, the change of wind direction was only observed above 402 1,500 m while the wind speed below was so weak that the shiftshirt in wind direction was not significant, 403 which was significantly different from the wind field of the other three types. The reason for the 404 difference between Type 4 and other types was mainly related to the topography of the Loess Plateau, 405 which has a blocking effect on the movement of the high-pressure system. Noteworthy, after the analysis 406 of these T-NPES events in different cities in China, we suggested that the T-NPES events were a common 407 pattern of the nocturnal PM_{2.5} enhancement, but did not always have an impact on the air pollution of the 408 following day. The pollution levels on the following day depended more on the strength of the cold front, 409 local pollution conditions, the structure of ABL, and regional transportation. Further quantification is 410 needed to determine the relationship between the T-NPES events and the pollution levels on the 411 following day.

412 To gain deeper look insights into the mechanism of nocturnal $PM_{2.5}$ enhancement, we systematically 413 documented instances of nocturnal PM_{2.5} enhancement during the winter of 2018 in Wangdu and Changzhou according to the surface $PM_{2,5}$ observation. We identified 48 such events in Wangdu and 27 414 in Changzhou, with proportions of T-NPES events of 37.5% and 40.7%, respectively. The results implied 415 416 that T-NPES represents merely one among multiple pathways contributing to the nocturnal PM_{2.5} 417 enhancement. We checked the nocturnal $PM_{2.5}$ enhancement events that were not caused by T-NPES in 418 Wangdu, the dominant wind field distributions within the ABL were southerly or characterized by static 419 light wind, which indicated that the nocturnal $PM_{2.5}$ enhancement might result from either horizontal 420 transport from polluted regions in the southern areas or the local accumulation of particulates in the stable 421 ABL. In the nocturnal PM_{2.5} enhancement events of non-T-NPES conditions in Changzhou, 422 higher wind speeds in the ABL and predominantly from the northern and southwestern, which indicated 423 the nocturnal PM_{2.5} enhancement might result from horizontal transport from the NCP (Huang et al., 424 2020) or caused by other reasons. For example, from the perspective of chemical formation, the nocturnal 425 atmospheric oxidation may elevate the nighttime aerosol concentration (Wang et al., 2023; Yan et al., 426 2023). In addition, we found the T-NPES event does not always cause a nocturnal $PM_{2.5}$ increase, in a 427 few cases, the strong north wind following the cold front play a role in remove removing the aerosol. In 428 summary, the T-NPES just represents one vertical transport mechanism that can collectively 429 contributes contribute to the enhancement of nocturnal PM_{2.5} with other physical and chemical processes

430 (Zhao et al., 2023). Further understanding of the coupling effect of transportation as well as the chemical

431 formation to the nocturnal PM_{2.5} enhancement is thus highly needed.

432 Based on these mentioned above, we suggested that the T-NPES events were a common phenomenon in 433 winter in plain areas such as the NCP and the YRD. A conceptual model was thus developed and shown 434 in Figure 6, there were-was the transportation of aerosol by the horizontal winds in the high altitude 435 above 1,000 m and the updrafts dominated before night, which was conducive to the formation and 436 suspension of the aerosol layer. Then, as-with the southeasterly movement of the high-pressure system 437 and the passage of the cold front at about the time of midnight, the wind direction began to turn to 438 north/northwest, causing the aerosol diluted to dilute. Finally, the downdrafts dominated in the ABL and 439 the LLJ might enhance the turbulentturbulence. Under the influence of subsidence behind the cold front 440 and turbulence, the depth of the aerosol layer suspended in the high altitude above 1,000 m began to 441 decrease and the pollutants gradually transported and diffused downwards into the lower layer of the 442 ABL, enhancing the concentration of surface PM_{2.5}.



443



445 Figure 6. Conceptual scheme of the T-NPES events

446 3.4 No T-NPES event occurred in Basin areas

We further checked the fix-point measurement in Xi'an and Chengdu, two cities with typical basin topography. The results indicated that there were essentially no T-NPES events in either city, suggesting the <u>conceptual-concept</u> did not work. Figure 7(a) indicated that the concentration of surface PM_{2.5} had no enhancement during the night from 23:00 on 31 December to 4:00 on 1 January, and from 22:00 on 1 January to 3:00 on 2 January in Xi'an. PM_{2.5} remained at a high concentration, while the extinction coefficient did not show the subsidence process, suggesting that the T-NPES events were not common here.

454 Taking the night of 31 December as an example, from 18:00 on 31 December to 4:00 on 1 January, the 455 concentration of surface $PM_{2.5}$ increased before 23:00 and then stabilized. Then it stabilized at high 456 values, while the extinction coefficient remained at a high level with about 1.0-1.2 km⁻¹ near 500 m. As 457 shown in Figure 7(d), from 18:00, 31 December to 6:00, 1 January, a light wind layer appeared below 458 1,000 m, with ~1 m/s. Such a static and stable condition was conducive to the accumulation of locally 459 generated particulate matter near the ground, causing the concentration of $PM_{2.5}$ to enhance between 460 18:00 and 23:00 on 31 December and the formation and maintenance of the aerosol layer at about 500 461 m. Noteworthy, the wind direction at the low layer was southeaster, while it was the opposite northwester 462 at about 1,000 m, which was the typical characteristic of mountain-valley breeze circulation. The 463 dominance of downdrafts below 500 m suggested that Xi'an was in the upper area of the nocturnal 464 mountain-valley breeze circulation. The mountain-valley breeze circulation could only be observed when 465 the background WS was relatively weak, which further indicated a stable structure of the ABL. The 466 example on 1 January was similar to the above one, with the extinction coefficient reaching 2 km⁻¹ and 467 <u>the</u> depolarization ratio decreasing after 21:00 due to the hygroscopic growth of aerosol by the rise in 468 relative humidity.



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Figure 7. (a) Surface PM_{2.5} concentration and relative humidity, (b) Extinction coefficient, (c) Depolarization ratio,
(d) Horizontal wind, and (e) Vertical wind, during the observation in Xi'an from 18:00, 31 December to 10:00, 2
January. The two black boxes were the time period period to be studied.

473 Due to the topography of the basin in Xi'an, the mountain-valley breeze circulation, or the horizontal 474 winds with lower WS always dominated in-the ABL, which was not conducive to the transport and 475 dispersion of particulate matter. The stable structure of the ABL resulted in the particulate matter 476 accumulated accumulating in the low layer, which was the main feature of the nocturnal particulate 477 matter distribution in Xi'an.

Figure 8 showed that the concentration of surface $PM_{2.5}$ also had no significant enhancement but remained a high value over nighttime in Chengdu. The distribution of the extinction coefficient in the two black boxes presented <u>a</u> double-layer structure, one layer near 250 m and another layer suspended at 481 about 500 m. Meanwhile, the wind field exhibited typical mountain-valley breeze circulation, as shown 482 in the two black boxes in Figure 8(d), which presented westerly wind near 250 m and southeasterly wind 483 above 500 m. The variation in wind direction due to the mountain-valley breeze circulation at different 484 layer_layers_might be responsible for the double layer double layer of particulate matter. Figure S14S13 485 illustrated-illustrates the backward trajectories when the double-layer appeared. The layer of particulate 486 matter at about 100 m might have originated from the southwest area of Chengdu, whereas the layer of 487 particulate matter at 500 m and 1,000 m might have originated from the northeast area of Chengdu. The 488 different sources of particulate matter were consistent with the mountain-valley breeze circulation in 489 Chengdu, further demonstrating the dominance of the mountain-valley breeze in the static and stable 490 ABL at night.

491 The orange box in Figure 8 indicated that the distribution of particulate matter in the ABL of Chengdu 492 under the dominance of northeasterly winds with high WS. Both the extinction coefficient and the 493 depolarization ratio showed a stratified structure, with the extinction coefficient initially higher below 494 750 m and lower above 750 m, whereas the depolarization ratio exhibited the opposite trend. The main 495 cause of this phenomenon was that the different sources of particulate matter in the two layers. Under 496 the influence of the dominant updrafts, local emissions with a high depolarization ratio were transported 497 upwards, while the lower layer was occupied by particulate matter with a lower depolarization ratio 498 transported by the northeasterly wind. As the continuous transport of the northeasterly wind, the entire 499 ABL was occupied by transported particulate matter with a high extinction coefficient and a low 500 depolarization ratio.

501 Due to the short time during the fixed-point observation period, it is difficult to make a universal 502 conclusion that no T-NEPS occurs in basin regions. Therefore, we further checked the surface and model 503 simulation data of the two basin cities for three months in winter 2018. We found that, unlike the plain 504 area, the T-NEPS events were almost never arely observed in the basin regions. It confirmed that the 505 conceptual model was indeed not applicable in the basin area. This was mainly attributed to the fact that 506 the movement of the weather system was blocked by the mountains surrounding the basin. Therefore, 507 the movement of the high-pressure system and the passage of the cold front had a weak impact on the 508 basin region. Without the downdrafts and the shift in wind direction associated with the movement of 509 the high-pressure system and the passage of the cold front, the structure of the ABL between Xi'an and 510 Chengdu was relatively stable, making it difficult for particulate matter to be transported and diffused, 511 and thus accumulate in the ABL at night. During the three months, we found that the wind field in Xi'an

512 was dominated by the light winds, while in Chengdu there were two states: one is was dominated by light 513 winds and the other by strong northeasterly windwinds. Fortunately, our fixed-point observations had 514 captured these typical processes indeed. In addition, considering the wind fields in basin cities were 515 mainly dominated by light winds, which was the main characteristic in of basin area (Bei et al., 2016; 516 Shu et al., 2021) and was similar to the wind fields below 1,500 m in Taiyuan and Linfen of the Type 4. 517 Therefore, we suggested that the Loess Plateau cities might serve as a crucial transitional zone between 518 the plains and the basin as introduced in Section 3.3. In summary, the conceptual model of T-NPES 519 events was applicable to the plain areas which were more influenced by the movement of the weather 520 system in winter, such as the NCP and YRD, but not to the basin areas.



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Figure 8. (a) Surface PM_{2.5} concentration and relative humidity, (b) Extinction coefficient, (c) Depolarization ratio,
 (d) Horizontal wind, and (e) Vertical wind, during the observation in Chengdu from 20:00, 7 January to 8:00, 10
 January. The two black boxes were the time period period of the double-layer structure, the orange box was the time
 period period to be studied.

526 4 Conclusions and outlook

527 In this study, we reveal that the T-NPES is a relatively common and important pathway that causes PM_{2.5} 528 pollution in the surface layer in the plain areas in winter China. The fixed-point observations in 529 Changzhou and Wangdu demonstrated that the T-NPES was associated with the subsidence of particulate 530 matter in the upper layer due to the movement of high pressure high pressure and the passage of the cold 531 front. Model simulations further confirmed the ubiquity of T-NPES events in plain areas, despite these 532 event types varied case by case. However, the observations in Xi'an and Chengdu indicated that the event 533 was less occurred in the basin areas, as the impact of the weather system was weakened by the obstruction 534 of mountains surrounding the basin. In further future studies, more multi-lidar measurement 535 measurements should be conducted in other cities in the plains and basin areas to look insight to the 536 detailed mechanism of T-NPES events. In addition, more works are urgently needed to uncover the 537 vertical profiles of chemical components of the particulate matter, since it may also be affected by the 538 coupling of physical and chemical processes.

539 **Code/Data availability.** The datasets used in this study are available at: 540 <u>https://doi.org/10.5281/zenodo.8368944</u> (Wang et al., 2023).

541 Author contributions. H.C.W. and S.J.F designed the study. Y.M.W. and H.C.W. analysed analyzed

the data, H.L.W. and X.L. provided the GEOS-Chem model simulation results, and Y.M.W. and H.C.W.

543 wrote the paper with input from all coauthors.

544 **Competing interests**. The authors declare that they have no conflicts of interest.

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