# Increasing importance of regional emission controls for further reduction of PM<sub>2.5</sub> in Beijing

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#### 25 Abstract

Over the past decade, Beijing has achieved positive results in controlling fine 26 27 particulate matter (PM<sub>2.5</sub>) pollution. However, it remains a challenge to further reduce PM<sub>2.5</sub> concentrations to a lower level, such as the World Health Organization's air 28 quality guidelines (5  $\mu$ g/m<sup>3</sup>). In this study, PM<sub>2.5</sub> concentrations and emission 29 reductions over eight years covering two policy periods of air pollution abatement 30 31 (2013–2017 and 2018–2020) were compared to investigate the efficiency of emission controls in Beijing and surrounding areas. An approach based on observational data, 32 particularly including data from a regional atmospheric background station, was 33 34 employed to calculate the relative contributions of the local emissions and regional 35 transport. Results show that local emission reductions play a more important role in decreasing PM<sub>2.5</sub> in Beijing. However, following a substantial decrease in local 36 emissions over the first period, the relative contribution of regionally transported PM2.5 37 reached more than 50% during the second period. The results indicate that joint regional 38 39 prevention and control of air pollution are needed for Beijing in the future. In addition, 40 the background PM<sub>2.5</sub> concentrations over the North China Plain show an increasing 41 trend in recent years, which may be attributed to the increased atmospheric oxidation 42 capacity, thereby posing a challenge for further regional air quality management.

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44 Keywords:

45 Regional transport, local emission, background level, relative contribution, air quality
46 improvement, PM<sub>2.5</sub>

47

48 Highlights:

49 1. The concentration and emission of PM<sub>2.5</sub> in Beijing showing a distinctive variation.

50 2. The regional joint control has great efficiency in reducing pollution.

51 3. After strictly limited, the PM<sub>2.5</sub> in Beijing became dominated by regional transport.

52 4. The PM<sub>2.5</sub> background level of North China Plain shown a significantly increasing.

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# 53 **1 Introduction**

54 Fine particles with an aerodynamic equivalent diameter of 2.5 µm or less, known as PM<sub>2.5</sub>, have been linked to numerous negative human health, including cardiovascular 55 and pulmonary disease, as well as increased morbidity and mortality from respiratory 56 57 and cardiovascular disease (Du et al., 2022; Liu et al., 2023; Sang et al., 2022; Seaton et al., 1995). Over the past two decades, China has experienced severe haze pollution, 58 59 primarily caused by PM<sub>2.5</sub>. This pollution is a result of rapid urbanization and 60 industrialization (Xu et al., 2022; Zhu et al., 2023). The North China Plain (NCP) region, home to many of China's most polluted cities, experiences exacerbated pollution due to 61 its topography and meteorological conditions (Li et al., 2023; Sun et al., 2017). To 62 63 mitigate pollution, the state council of the People's Republic of China implemented a series of policies aimed at reducing air pollution. These include the Air Pollution 64 Prevention and Control Action Plan (APPCAP) from 2013 to 2017 (Zhang et al., 2019b) 65 and the Three-year Action Plan to Fight Air Pollution (TAPFAP) from 2018 to 2020 66 67 (Shao et al., 2023). As a result of these strict emission controls, Beijing has seen a significant decrease in annual average PM<sub>2.5</sub> concentrations, from 85  $\mu$ g/m<sup>3</sup> in 2013 to 68  $37 \ \mu g/m^3$  in 2020. However, the concentration still far exceeds the World Health 69 Organization's air quality guidelines (5  $\mu$ g/m<sup>3</sup>). Therefore, further efforts to mitigate 70 71 PM<sub>2.5</sub> pollution remain a critical challenge for the future (Shu et al., 2023).

To address PM<sub>2.5</sub> pollution in the Beijing region, a crucial step is to quantify its 72 sources. Anthropogenic emissions and meteorological conditions are widely recognized 73 74 as the primary drivers of severe PM<sub>2.5</sub> pollution (Chen et al., 2020; Chen et al., 2025; Liu et al., 2021; Seinfeld et al., 1998; Wen et al., 2018). Regional transport also plays a 75 76 significant role, particularly due to the surrounding industrial cities and unfavorable meteorological conditions (Feng et al., 2019; Li et al., 2021; Xu et al., 2023; Lin et al., 77 2008; Sun et al., 2023; Zhang et al., 2021b). Quantifying the influence in the 78 neighboring region is necessary (Hernández-Moreno et al., 2023; Onishi et al., 2025). 79 Distinguishing the local emission contribution (LEC) and regional transport 80

81 contribution (RTC) to pollution in Beijing is imperative for effective mitigation 82 strategies. While short-term RTC during pollution events has been extensively studied using chemical transport models (Chen et al., 2019; Jiang et al., 2015; Li and Han, 2016; 83 Lv et al., 2022; Wu et al., 2017; Zhang et al., 2019a; Zhang et al., 2021a; Zhang et al., 84 2021b), there remains a notable lack of research on the long-term impact of RTC across 85 several years (Chen et al., 2021; Zhao et al., 2020). A key challenge in this area is the 86 87 absence of accurate observational data to validate model predictions. Therefore, future 88 research should focus on developing reliable measurement tools and long-term observational studies to better understand and quantify the RTC to PM<sub>2.5</sub> pollution in 89 90 Beijing. This will be crucial for implementing effective regional prevention and control measures to reduce severe air pollution over the NCP (Lv et al., 2020; Xing et 91 92 al., 2017).

Direct observations of the local and regional contributions to PM<sub>2.5</sub> are often 93 challenging. To address this, Ge et al. (2018) developed an approach, based on 94 95 observational data, to estimate the maximal regional transport contribution (MaxRTC) 96 and the minimal local emission contribution (MinLEC) to air pollutants in Beijing. Subsequently, Tan et al. (2022) applied this method to analyze the trend in regional 97 98 transport's contribution to PM<sub>2.5</sub> in Beijing during the APPCAP period, revealing a 99 significant increase. In 2018, China introduced a new policy, TAPFAP, which imposed 100 stricter emission limits on industries, motor vehicles, heating system, and other sources. 101 Consequently, it has become essential to review and update our understanding of how regional transport contributes to PM<sub>2.5</sub> pollution in Beijing under these varying policies, 102 103 in order to plan and implement effective pollution control measures in the future.

In this study, we employ the approach developed by Ge et al. (2018), incorporating an updated method for calculating the  $PM_{2.5}$  background levels, to estimate the changes in the contributions of local emissions and regional transport to  $PM_{2.5}$  in Beijing from 2013 to 2020. Our objective is to assess the effectiveness of the emission control measures implemented during the different policy periods. To achieve this, we have defined two phases: Phase I, spanning from 2013 to 2017and encompassing the APPCAP period; and Phase II, extending from 2015 to 2020 and covering the TAPFAP period, with targets referenced to 2015. The overall motivation was to provide new insights into future air pollution control strategies, aimed at achieving lower  $PM_{2.5}$ concentrations.

114 **2 Data and methods** 

#### 115 **2.1 Study area and measurement site descriptions**

The NCP is the most populous plain in China, extending from the Yanshan Mountains 116 117 in the north and Taihang Mountains in the west, and reaching Bohai Bay in the east (Figure 1(a)). The geography of the NCP is favorable for the formation of pollution 118 episodes as the region is an air pollutant convergence zone. The westerly synoptic flow 119 across the Taihang Mountains sinks and warms adiabatically, creating a low-pressure 120 trough on the lee side (Jin et al., 2021b). During winter, the prevailing winds typically 121 exhibit a northeasterly pattern, while during summer they shift to a southwesterly 122 pattern, given the distinct influence of the subtropical high-pressure system (Fu et al., 123 2014; l., 2012; Jin et al., 2021a). 124

125 As Figure 2(a) shows, cities with high primary  $PM_{2.5}$  emissions are located on the edge of the plain along the foothills of the mountains, and are distributed from 126 southwest to northeast. Southwesterly transport occurs frequently during regional 127 128 pollution episodes and serves as a trigger (Hu et al., 2022; Li et al., 2021). One of the 129 most polluted cities is Shijiazhuang, in the southwest of the region, which is the capital 130 of Hebei Province and often ranked as one of the most polluted cities in China. Beijing 131 has relocated much of its industry to nearby regions, but still suffers from heavy pollution. Shangdianzi (SDZ) Station, situated northeast of Beijing within the Beijing 132 133 municipality, is one of the regional Global Atmosphere Watch (GAW) stations in China, 134 and it has only a few small villages and low anthropogenic emissions in its vicinity (Lin et al., 2008). Shijiazhuang was selected in this study as a typical polluted city to 135 136 represent source regions on the NCP. Beijing was selected as the receptor area, while 137 SDZ was selected as the background site to represent the background concentration of

#### 138 $PM_{2.5}$ in the NCP region.

#### 139 **2.2 Observational data**

The hourly PM<sub>2.5</sub>, SO<sub>2</sub>, NO<sub>2</sub> and CO concentration data from 2013 to 2020 at 87 sites 140 in the NCP region were collected from the China National Environmental Monitoring 141 Centre (CNEMC) (http://www.cnemc.cn, last access: 17 October 2024). Specifically, 142 there are 12, 17 and 54 observation stations in Beijing, Tianjin and Hebei Province, 143 respectively, including 9 stations in Shijiazhuang. To reduce errors from individual sites, 144 145 the averaged PM<sub>2.5</sub> as well as the related pollutants (e.g., SO<sub>2</sub> and NO<sub>x</sub>) concentrations 146 from all stations within each city were calculated. The CNEMC observation network does not cover the background station. Therefore, data from the SDZ atmospheric 147 background station in 2013–2020 operated by the China Meteorological Administration 148 were used to represent the concentrations of pollutants in the BG region. Commercial 149 150 instruments from Thermo Electron Corporation, USA, have been deployed to measure O<sub>3</sub> (TE 49C), NO/NO<sub>2</sub>/NO<sub>x</sub> (TE 42ITL), CO (TE48C) and SO<sub>2</sub> (TE 43CTL) at SDZ. 151 For PM<sub>2.5</sub>, an R&P model TEOM 1400A instrument with a 2.5-µm cyclone inlet and a 152 153 humidity control system was used. More details can be found in Zhao et al. (2009). Emissions data for primary PM<sub>2.5</sub>, SO<sub>2</sub> and NO<sub>x</sub> in the NCP region during the study 154 period were obtained from the Multi-resolution Emissions Inventory for China 155 (http://www.meicmodel.org, last access: 17 October 2024) (Li et al., 2017; Zheng et al., 156 157 2018), with a spatial resolution of  $0.25^{\circ} \times 0.25^{\circ}$ . The yearly averaged emissions of the three pollutants over the Beijing and Shijiazhuang areas during 2010-2020 were 158 geographically averaged calculated, and these are listed in Table S1. Meteorological 159 160 variables at Beijing and the SDZ background station during 2013–2020 were obtained 161 from the China Meteorological Administration, and these were recorded at hourly resolution. The nearest site of meteorological measurement stations to the CNEMC 162 stations were selected, with the distances between the meteorological and 163 164 environmental stations not exceeding 500 meters.

# 165 2.3 Approach for distinguishing local emissions from regional 166 transport

167 In this study, the relative contribution from local emissions and regional transport were 168 calculated following the method of Ge et al. (2018). This approach has been widely 169 used in other studies for the comparison with the model results in NCP (Yao et al., 2022; Khuzestani et al., 2022; Song et al., 2023; Qu et al., 2024), Shandong (Zhao et al., 2018) 170 171 and Wuhan (Ma et al., 2021). The PM<sub>2.5</sub> concentrations in the background (B), receptor (R), and source (S) regions were divided into several parts: 172 173  $C_S = C_{S \text{ local}} + BK$ (1) $C_R = C_R |_{local} + C_{Regional} + BK$ 174 (2) $C_B = \beta \cdot C_{R \text{ local}} + C_{R \text{ egional}} + BK$ (3)175 Here, Clocal represents the concentration contributed by local emissions, CRegional is 176 177 the regional transport influences from polluted areas outside Beijing, which are mostly located to the south and east of Beijing. Figure 1 shows them at their respective sites. 178 179 BK is the background concentration over the NCP, which is determined from clean air masses.  $\beta$  reflect the relationship between local emissions (C<sub>R local</sub>) in urban Beijing 180

and the impacts on the SDZ background site, with the  $\beta$  value always physically 181 182 higher than zero. We assume that C<sub>Regional</sub> in receptor region R is the same as that in background region B based on the similar distance between the source region and the 183 184 receptor and background regions. Indeed, the study by Ge et al. (2018) demonstrates that the uncertainty associated with this assumption can be ignored. Thus, the  $C_{Regional}$ 185 effects can be eliminated by subtracting Eq. (3) from Eq. (2), and then the MinLEC of 186 urban Beijing can be calculated in Eq (4), and the MaxRTC can be deduced from Eq 187 (5): 188

189 
$$\operatorname{Local} = \frac{C_{R \text{ local}}}{C_{R}} = \frac{C_{R} - C_{B}}{(1 - \beta)C_{R}} \ge \frac{C_{R} - C_{B}}{C_{R}}$$
(4)

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$$\operatorname{Transport} = \frac{C_{\text{Regional}}}{C_{\text{R}}} = \frac{C_{\text{R}} \cdot BK \cdot C_{\text{R local}}}{C_{\text{R}}} \leqslant \frac{C_{\text{B}} \cdot BK}{C_{\text{R}}}$$
(5)

191 The difference between  $C_R$  and  $C_B$  is partly from the impacts of local emissions on 192 the air pollutants in Beijing. A larger difference between  $C_R$  and  $C_B$  therefore 193 corresponds to a larger MinLEC (Ge et al., 2018). Subtracting the BK value from  $C_B$ 194 represents the MaxRTC, since there are very limited local emissions in SDZ.

The background concentration BK in Eq. (5) is important in calculating MaxRTC 195 196 and has usually been estimated based on the lowest concentration of air pollutants at the background station. Lin et al. (2009) picked the first peak in the frequency 197 198 distribution of the concentration of air pollutants decomposed by two Lorentzian curve fittings as the background value. Ge et al. (2018) used this method to calculate the PM<sub>2.5</sub> 199 in Beijing and SDZ, and took their average as BK value. However, the regional 200 background is not constant over the long term. Changes can cause biases in calculating 201 202 the contribution from regional transport using Eq. (5). Ruckstuhl et al. (2012) developed the Robust Extraction of Baseline Signal (REBS) technique to estimate the background 203 level of trace gases and well-mixed air pollutants. This method is based on separation 204 of pollutant concentration into three parts-the background value, regional contribution, 205 206 and measurement errors from instruments-similar to the assumptions we use here, and has been applied at SDZ background station in other studies (Yao et al., 2012; Yao et 207 al., 2014). However, Liu et al. (2019) argued that REBS was not suitable for estimating 208 the background of carbon monoxide at SDZ, and instead recommended the 209 210 meteorological method (MET) (Fang et al., 2014), which is based on the mean concentrations under specified meteorological conditions every year. To make our 211 212 results more robust, both REBS and MET methods were employed in this study to 213 estimate long-term PM<sub>2.5</sub> background concentrations over the NCP. More detailed 214 descriptions of the mathematical functions and criteria used can be found in the 215 supplementary materials.

To assess the effect of transport from the source region, the regional transport efficiency ( $\alpha$ ) was defined as

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$$\alpha = \frac{C_{\text{Regional}}}{C_{\text{S local}}} \leqslant \frac{C_{\text{B}} - \text{BK}}{C_{\text{S}} - \text{BK}}$$
(6)

Following Eq. (1),  $C_{S \text{ local}}$  is the concentration in the source region after removal of

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background concentrations and represents the contribution that can be transported from 220 the polluted areas. The ratio between  $C_{S \text{ local}}$  and  $C_{Regional}$ , given by  $\alpha$ , represents the 221 222 efficiency of regional transport, with a higher value signifying a greater regional 223 transport effect (RTE). Furthermore, to assess the efficiency of emission control on pollution mitigation, the emission control efficiency (ECE) proposed by Tan et al. (2022) 224 was employed in this study. The ECE is expressed as the concentration change 225 226 corresponding to given emission change of a certain air pollutant, and is calculated as the concentration decreasing rate (CDR) divided by the emission decreasing rate (EDR). 227 More details can be found in supplementary materials. 228

# **3 Results and discussion**

# 230 **3.1 PM<sub>2.5</sub> changes under different policies**

# **3.1.1 Significant mitigation of air pollution in Phase I**

Figure 2 illustrates the spatiotemporal variation of observed PM2.5 and associated 232 233 primary emissions during the study period over the NCP region. In general, emissions in the NCP showed a significant decline in phase I, especially in the megacities of 234 235 Shijiazhuang, Baoding, Tangshan, Beijing, and Tianjin, where the decrease exceeded 236  $0.05 \ \mu g/m^2/s/yr$ . The decline in emissions in the surrounding rural areas is less pronounced. Table S2 lists the annual mean and maximum PM2.5 concentrations for 237 Beijing, the SDZ background station and Shijiazhuang. As expected, PM<sub>2.5</sub> 238 239 concentrations in the NCP region showed a significant reduction in Phase I, associated 240 with the implementation of emission controls. The largest reduction can be seen in Shijiazhuang, at approximately 20  $\mu$ g/m<sup>3</sup>/yr, which is twice or even three times the 241 reduction seen in Beijing. However, the SDZ background station did not experience a 242 significant change in concentration. 243



245 ownership, and GDP for Beijing and Shijiazhuang. Coal consumption has substantially reduced, while vehicle ownership and GDP have increased in both areas, particularly 246 in Beijing. This suggests that most cities in the NCP region have reduced their emissions 247 without compromising economic development. The emission control efficiencies for 248 249 Beijing and Shijiazhuang are presented in Table 1. The value in Shijiazhuang is higher than that for Beijing, indicating that the source region (i.e., Shijiazhuang) has a higher 250 control efficiency. Overemphasizing local emission control measures can make it more 251 252 difficult and less efficient to abate air pollution, especially in a downstream receptor 253 region such as Beijing (Tan et al., 2022).

# **3.1.2 Improvement slowdown in Phase II**

In Policy II (TAPFAP), the government requires that the PM2.5 concentration in 2020 255 should be reduced by 18% compared to 2015. Therefore, for the analysis from a policy 256 257 perspective, 2015 should be the target year for Phase II. As Figure 2(d) shows, the rate of emission reduction over the NCP in Phase II is slower. The rate of decline in primary 258 PM<sub>2.5</sub> emissions in most industrial cities (e.g., Shijiazhuang, Baoding) is only about one 259 fifth of that in Phase I, reaching 0.01–0.02  $\mu$ g/m<sup>2</sup>/s/yr. Tangshan experiences the largest 260 reduction, at about 0.05  $\mu$ g/m<sup>2</sup>/s/yr. Similarly, the PM<sub>2.5</sub> concentration in most cities 261 also shows a slower reduction, with the rate generally being below 10  $\mu$ g/m<sup>3</sup>/yr. 262

Taking Shijiazhuang as an example, the rate of reduction in PM<sub>2.5</sub> concentration 263 during Phase II was  $-8.1 \ \mu g/m^3/yr$  ( $-10.8 \ \mu g/m^3/yr$  (p < 0.01) during 2018-2020), 264 roughly half that in Phase I  $-17.3 \ \mu g/m^3/yr$ , see (Figure 3 a). This indicates that the 265 change in PM<sub>2.5</sub> concentration aligns with the change in emissions in the source region. 266 267 However, compared to Shijiazhuang, Beijing's emission reductions in Phase II were 268 more significant, with major emission reductions rate in  $PM_{2.5}$  and  $SO_2$  (except  $NO_x$ ) at about half that of Phase I (Figure 3 d). Interestingly, the rate of decline in the PM<sub>2.5</sub> 269 concentration remains nearly constant from Phase II to Phase I ( $-8.7 \mu g/m^3/yr$  in Phase 270 II ( $-7.4 \ \mu g/m^3/yr$  (p < 0.01) during 2018-2020) vs.  $-7.2 \ \mu g/m^3/yr$  in Phase I), which 271 appears to contradict the decreasing rate of decline in precursor emissions in Beijing. 272

In contrast, the rest of the NCP region experiences a concurrent decrease in both the reduction rate of emissions and concentrations. Nevertheless, the rate of decrease in concentration in Beijing during Phase II is comparable to that in Shijiazhuang, and both are similar to other cities in the NCP region. Despite the slowdown in emissions reduction, the emission control efficiency in both Beijing and Shijiazhuang increases (Table 1), indicating more effective control of  $PM_{2.5}$  emissions over the whole of the NCP in Phase II (Kong et al., 2023).

280 The results highlight controlling regional emissions is now more crucial for 281 addressing pollution in Beijing than controlling local emissions, especially at low 282 concentrations. This finding aligns with the model results of Lv et al. (2020), who simulated the effects of COVID-19 lockdown on reducing local traffic emissions in 283 284 Beijing and their impacts on air quality. They found that although traffic emissions decreased substantially during the COVID-19 pandemic, their impacts on air quality 285 were offset by the increased atmospheric oxidization capacity in surrounding regions. 286 Consequently, they recommended synchronously controlling regional sources to 287 288 maximize the benefits of controlling local traffic emissions.

#### **3.2 Decreasing contribution of local emissions**

290 In the past 10 years, numerous studies have focused on the relative contributions to air pollution in Beijing from local emissions and regional transport. A total of 30 291 292 publications that discuss the regional transport and the local emission contributions to 293 PM<sub>2.5</sub> pollution in Beijing during 2013–2020 are reviewed here. The sources, methods 294 and key findings are summarized in Table S3. Most of these studies focused on single 295 case studies, with only five simulated the annual average contribution. None of them 296 accounted for the variation from year to year. Additionally, most previous studies 297 quantified the contribution using a chemical transport model, leaving a gap for reliable assessments based on observations. 298

In this study, an approach based on observations was employed to estimate the MinLEC and MaxRTC to PM<sub>2.5</sub> in Beijing. Figure 4 (a) presents the results for MinLEC 301 in Beijing between 2013 and 2020. The citations of these studies can be found in the 302 supplementary materials. The results of the MinLEC to PM2.5 in Beijing are within the range of previously reported literature values, with consistent temporal variation from 303 2013-2019. Specifically, during the first three years, the MinLEC displayed a slightly 304 increasing trend from 53% in 2013 to 58% in 2015. The median values of various 305 models during different periods show a significant increase from 46% in 2013 to 61% 306 in 2015. However, starting from 2015, it shows a significant decrease, dropping to 307 308 nearly 30% by 2019. An independent samples t-test was conducted to compare the differences between the periods of 2013-2015 and 2018-2020. The results revealed a 309 310 statistically significant difference between the mean value of the two groups, with a tvalue of -8.96 (p < 0.01). These findings suggest that the reduction in local emissions 311 312 is likely the dominant factor contributing to Beijing's PM<sub>2.5</sub> mitigation efforts. The decline in local emissions may have led to a reduction of 30.7  $\mu$ g/m<sup>3</sup> in PM<sub>2.5</sub> 313 concentrations over the last eight years and could be the primary reason why the 314 concentration in Beijing has declined at a rate similar to that of other cities. 315

Our values for MinLEC fall within the range of LEC values reported in the literature, but are slightly lower. This difference is likely due to our calculation providing an estimate of the lower limit of LEC. The difference between MinLEC and the LEC derived from other studies highlights the need for further validation of the impact of the rapid decrease in local emissions.

# 321 **3.3 Increasing contribution of regional transport**

As shown in Figure 4 (b), the MaxRTC in Beijing experienced a significant increase, rising from 40% in 2013 to nearly 50% in 2020. Regional transport is significantly affected by the wind direction, particularly when southerly winds dominate (Hu et al., 2022; Li et al., 2021). Yang et al. (2016) assessed the frequency of southerly winds using the sine of the vector-average wind direction and found that it contributed 22% to  $PM_{2.5}$  pollution in Beijing. Despite successful emission control measures in surrounding cities, where  $C_{s local}$  has shown a substantial decline (Figure 5 a), their

relative impacts on Beijing's air quality are intensifying. The differences between Cs 329 local and C<sub>Regional</sub> are narrowing, indicating the increased importance of regional 330 transport (Figure 5 a). It is worth noting that RTE exhibited a strong seasonal variation, 331 indicating that meteorological conditions, rather than the Cs local, are the main factor 332 333 instead driving the trend. The regional transport effect depicted in Figure 5(b) displays a significant increasing trend of 2.2% yr<sup>-1</sup>. Concurrently, the sine of the vector-average 334 wind direction showed the opposite trend, further indicating a potential relationship 335 336 between southerly winds and the regional contribution to PM<sub>2.5</sub> in Beijing.

As mentioned above, the PM2.5 concentration across the entire NCP region decreased 337 significantly due to the emission control measures. However, the yearly background 338 values, calculated using REBS method, show an increase from 7.25  $\mu$ g/m<sup>3</sup> in 2013 to 339 more than 10  $\mu$ g/m<sup>3</sup> after 2016 (Table S2). This increase is also confirmed by the results 340 obtained from the MET method, as shown in Figure 5 (c, d). In contrast to previous 341 studies (Ge et al., 2018; Tan et al., 2022), background values have been increasing year 342 by year, suggesting they may play an increasingly important role in the variation of 343 344 PM<sub>2.5</sub> pollution over the NCP region, particularly during future clean-up periods when PM<sub>2.5</sub> concentrations reach very low levels. Besides, the nighttime PM<sub>2.5</sub> and nighttime 345 O<sub>3</sub> during the high O<sub>3</sub> concentrations periods show a positive correlation with R reached 346 at 0.39 (p<0.01, two-tailed significance). On the contrary, negative correlation was 347 348 found during low  $O_3$  concentrations periods (-0.26, p < 0.01). This suggests that increases in nighttime ozone concentrations tend to coincide with increases in nighttime 349 PM2.5 from 2013 to 2020, which may be due to the dominance of secondary sources 350 for both nighttime ozone and PM2.5. Furthermore, it is also worth noting that 351 352 incorporating the background contribution into the regional effect 353 (MaxRTC + Background) increases the RTC estimate for the 2019-2020 period to 70%, which is close to the median value reported in other studies. This evidence highlights 354 the importance of considering the background as an important factor, as it can 355 substantially influence estimates of the contribution of regional transport (up to 30% in 356 this study). 357

# 358 **3.4 Enhanced background value**

359 As mentioned above, the background level of PM<sub>2.5</sub> at the SDZ station showed a clear upward trend during 2013-2020 (Figure 5 d). This trend is also evidenced by the 360 concentration heatmap at the SDZ background station (Figure S2), which demonstrates 361 362 a notable increase in low concentration values. The Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model was employed to generate 72-hour air masses 363 364 backward trajectories from 2013 to 2020. The GDAS meteorological datasets with horizontal resolution of 0.5° from the Air Resources Laboratory (ARL) were used. 365 These backward trajectories start from the SDZ background station at 100 m height 366 367 AGL (Figure 6 a-h)), with a restart every 6 h. They were calculated and clustered annually to enhance the representativeness of the trajectory ensembles. As shown in 368 Table S4 and Figure 6 (i, j), the averaged concentrations of PM<sub>2.5</sub> and O<sub>3</sub> were 369 calculated for each cluster. Although the air masses clusters varied from year to year, 370 371 three typical clusters were evident at the SDZ stations, one characterized by southbound 372 short-range and two by northbound short- and long-range. It is worth noting that both of the PM<sub>2.5</sub> and O<sub>3</sub> concentrations from north direction showed a significant increasing 373 trend with more frequency, especially in long-range clusters. This could be one of the 374 375 reasons for the increasing background level of PM<sub>2.5</sub> in NCP region.

In order to investigate in-depth, yearly variations in different percentiles (5%, 25%, 376 50%, 75%, 95%) of PM<sub>2.5</sub> and O<sub>3</sub> concentrations from 2013 to 2020 were calculated 377 and shown in Figure 7a (selected percentiles) and Figure S3 (whole dataset). The 378 significant reduction in PM<sub>2.5</sub> concentrations above the 75<sup>th</sup> percentile is attributable to 379 the pollution controls implemented in the NCP region in recent years. In contrast, a 380 slight upward trend in PM<sub>2.5</sub> concentrations below the 25<sup>th</sup> percentile is considered to 381 indicate an increase in background levels. The nighttime O3 concentrations also 382 displayed a clear upward trend, especially at the 95<sup>th</sup> percentile. In order to explore the 383 relationship between low concentration of PM2.5 and nighttime O3, wind rose were 384 plotted (Figure 7 b and c). The results show a clear similarity in wind direction and 385 wind frequency distribution, with high values originating from the northwest. In other 386

words, the increase in low PM<sub>2.5</sub> concentrations from the northwest is associated with 387 an increase in higher nighttime O<sub>3</sub> concentrations. Besides, the nighttime PM<sub>2.5</sub> and 388 nighttime O<sub>3</sub> during the high O<sub>3</sub> concentrations periods show the positive correlation 389 with coefficient reached at 0.39 (p<0.01, two-tailed significance). On the contrary, 390 391 negative correlation was found during low O<sub>3</sub> concentrations periods (-0.26, p<0.01). This suggests that elevated nighttime O3 concentrations tend to coincide with the 392 increased nighttime PM<sub>2.5</sub> from 2013 to 2020. These may be due to the fact that higher 393 394 O<sub>3</sub> concentrations at nighttime enhance the oxidative production of secondary aerosols. At night, O<sub>3</sub> oxidizes NO<sub>2</sub> to form NO<sub>3</sub> radicals, which subsequently react with NO<sub>2</sub> to 395 form N<sub>2</sub>O<sub>5</sub> (Brown and Stutz, 2012), thus leading to the formation of secondary aerosols 396 through heterogeneous chemistry (Jo et al., 2019; Qu et al., 2019). 397

# **4 Implications and conclusions**

399 In this study, we compared two phases of PM<sub>2.5</sub> pollution variations in the NCP region 400 from 2013 to 2020 and found that Phase I exhibited more substantial emission reductions across most NCP cities compared to Phase II. Figure 8 illustrates the 401 evolving contributions of local emissions and regional transport across the two policy 402 403 periods. MinLEC exhibits a decreasing trend, while MaxRTC demonstrates an upward 404 trend. A comparison of the median values between the two policy periods reveals a significant shift, with MinLEC decreasing by 18% and MaxRTC increasing by 7%. 405 Furthermore, the relative contribution of regional transport to PM2.5 pollution in Beijing 406 407 has increased notably. To effectively address future PM<sub>2.5</sub> pollution in Beijing, it is imperative to consider the influence of the surrounding areas from a broader, regional 408 409 perspective.

Despite the observation-based approach in this study, which does not account for all PM<sub>2.5</sub> dynamics, our findings highlight a notable increase in regional contributions. Additionally, PM<sub>2.5</sub> background concentrations in the NCP region have risen in recent years, surpassing the World Health Organization's air quality guidelines. Achieving further reductions in PM<sub>2.5</sub> pollution will require long-term, large-scale control measures. Policymakers should address not only primary emissions from urban and industrial areas but also broader meteorological and atmospheric oxidant factors. Such an approach is vital to meet China's pollution and carbon reduction targets over the next decade.

# 419 ASSOCIATED CONTENT

# 420 Supporting Information

- 421 Detailed Information about the REBS and MET methods, along with supporting tables and figures,
- 422 is provided in the supporting information.

#### 423 **Data availability**

- 424 The data that support the findings of this study are available on request from the corresponding
- 425 author, Prof. Ge, upon reasonable request.

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#### 429 Notes

430 The authors declare no competing financial interests.

# 431 Author statement

432 Lingkai Dong: Methodology, Formal analysis, Data Curation, Writing - Original Draft,
433 Visualization. Weili Lin: Investigation, Data Curation. Zhiqiang Ma: Investigation, Data Curation.
434 Wei Wang: Investigation, Data Curation. Lei Kong: Investigation, Data Curation. Xiaobin Xu:

Resources, Data Curation. Oliver Wild: Validation, Writing - Review & Editing. Yuanlin Wang:
Data Curation, Funding acquisition, Writing - review & editing. Baozhu Ge: Conceptualization,
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# 690 **Tables**

691	Table 1 The concentration decrease rate (CDR), emission decrease rate (EDR), and
692	emission control efficiency (ECE) of PM <sub>2.5</sub> in Beijing and Shijiazhuang during the
693	different policy periods.

Luder	Phase I		Phase II <sup>a</sup>	
Index	Beijing	Shijiazhuang	Beijing	Shijiazhuang
CDR (%)	-9.9	-13.7	-15.1	-8.2
EDR (%)	-11.1	-12.0	-11.3	-5.5
EDR <sup>b</sup> (%)	-6.9	-12.9	-2.9	-4.8
ECE	0.89	1.14	1.31	1.48
ECE <sup>b</sup>	1.44	1.06	5.22	1.71

a: To avoid the influence of COVID-19, the averages for 2015 and 2019 were chosen

695 for calculation.

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# 699 Figures



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**Figure 1.** (a) Geographic locations and terrain of the NCP and the three cities/stations

in this study— Shijiazhuang (SJZ), Beijing (BJ), and Shangdianzi (SDZ) (source:
 NASA satellite map; http://worldwind.arc.nasa.gov). (b) Assumptions regarding PM<sub>2.5</sub>
 concentrations in the three regions.



Figure 2. Spatial distribution of (a) NCP emissions in 2013, and (b–d) reductions in emissions (shading) and concentrations at observation sites (spots) during (b) 2013– 2020, (c) 2013–2017, and (d) 2015–2020. Grid points with emissions below 0.02  $\mu g/m^2/s$  are ignored.



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Figure 3. Time series of the concentration for site (a) SJZ, (b) SDZ, (c) BJ and (d) Normalized emissions of primary  $PM_{2.5}$ , SO<sub>2</sub> and NO<sub>x</sub> from 2013 to 2020. The 90 days smoothed curves (blue), and the linear regression lines during Phase I (red dashed line), and Phase II (green dashed line) were also plotted. The slope (k<sub>1</sub>, k<sub>2</sub>) units are  $\mu g/m^3/d$ in (a-c) and  $\mu g/m^2/s/m$  in (d).



Figure 4. Box plots of the (a) local emissions contribution (LEC) and (b) regional transport contribution (RTC) calculated in this study and other studies (references summarized in Table S3). The black lines mark contribution = 50%. Gray ang orange shading indicate the contribution of more than 50% and less than 50%, respectively.



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Figure 5. (a) Local  $PM_{2.5}$  concentrations in Shijiazhuang and regional pollution transport received in Beijing, and (b) trends of regional transport effeciency a and the sin(WD), and (c, d) the time series of  $PM_{2.5}$  daily mean concentration in Shangdianzi (grey) and the background values calculated by the REBS method (red) and the MET method (green), and (e) variations in MaxRTC under three different background level: background values calculated from Ge et al. (2018) method (black), using Beijing and Shangdianzi averages (blue), and using REBS (red).



732 733 Figure 6. The 72-hour backward tracectories over the Shangdianzi station for each year from 2013 to 2020 (a-h), respectively. The annual average concentration of PM<sub>2.5</sub> (i) and O<sub>3</sub> (j) in the north trajectories from 2013 to 2020.

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Figure 7. Changes in different percentiles of  $PM_{2.5}$  and  $O_3$  between 2013 and 2020 (a), Wind rose plots for the lowest 25% of  $PM_{2.5}$  (b) and the highest 50-100% of nighttime  $O_3$  (c). The abbrevations of Per5, Per25, Median, Per75 and Per95 at the vertical coordinate of figure (a) represent 5, 25, 50, 75 and 95 percentile, respectively.



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744 Figure 8. Annual changes in the MinLEC, MaxRTC and background values of PM<sub>2.5</sub>

745 levels in Beijing, along with their median changes between the two policy periods.