

1 **Spatial-temporal variations and attribution analysis of SO₂**
2 **concentration in Beijing-Tianjin-Hebei regions from 2013 to 2022**

3 **Bingfen Cheng^{1,2}, Rui Zhang^{1,2}, Bo Yang^{1,2}, Guiqiang Zheng^{*1,2}, Quanjie Zhu^{1,2}, Huiqing**
4 **Lian^{1,2}**

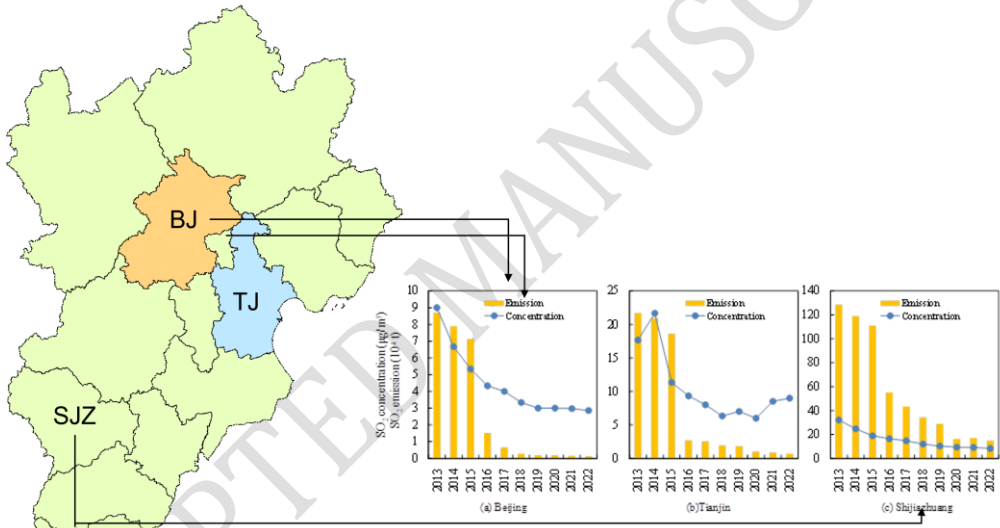
5 1. College of Emergency Technology and Management, North China Institute of Science & Technology, Langfang, 065201,
6 China.

7 2. Multi-scene water chain accident wisdom emergency technology innovation center of Hebei, Langfang, 065201, China.

8 * Corresponding authors:

9 E-mail addresses: 76382119@qq.com

10 **GRAPHICAL ABSTRACT**



The annual averaged concentration and emissions of SO₂ in three cities from 2013 to 2022

11

12 **Abstract**

13 Based on the monitoring data of air pollutants in Beijing-Tianjin-Hebei regions
14 from 2013 to 2022, the spatial-temporal distribution of SO₂ concentration and main
15 influencing factors were analyzed through multi-methods of Daniel trend test, KZ
16 filtering and WRF/CMAQ model simulations. From 2013 to 2022, the annual
17 averaged concentrations of SO₂ in Beijing, Tianjin and Hebei regions were in
18 significant downward trends, all passing the Daniel trend test ($\alpha=0.01$). The annual
19 averaged concentration of SO₂ in Beijing, Tianjin and Hebei regions decreased from
20 26.6~114.3 $\mu\text{g}\cdot\text{m}^{-3}$ in 2013 to 3.0~11.0 $\mu\text{g}\cdot\text{m}^{-3}$ in 2022, with the decline rates

21 fluctuating from 2.62~11.81 $\mu\text{g}/(\text{m}^3\cdot\text{yr})$. The average annual concentration of SO₂ in
22 Tangshan city decreased the most, reaching 92.3% while it changed to be the smallest,
23 about 78.2% in Chengde City. The reduction of coal consumption in the
24 Beijing-Tianjin-Hebei regions significantly reduced the atmospheric SO₂ emissions
25 and directly reduced the regional averaged concentrations of SO₂. Meteorological
26 factors were generally conducive to the diffusion of SO₂ concentration during
27 2013~2022, and meteorological factors contributed approximately 0.4% to 5.5% in
28 the 13 cities of the Jingjinji regions, while the contribution of anthropogenic emission
29 reduction ranged from 94.5% to 99.6%. The inter-annual variation of meteorological
30 factors during the summer periods from 2013 to 2022 was overall not conducive to
31 the dispersion of SO₂ concentrations, with the contribution ranging from -52.6% to
32 -1.0% in 13 cities. During the summer season from June to August, under easterly and
33 southerly winds, the SO₂ concentration in Beijing was approximately twice as high as
34 that under northerly and westerly winds. The contribution of southwest regional
35 transport channel to the SO₂ concentration in Beijing was 33.5%, while it increased to
36 38.9% in the southeast regional transport channel. Therefore, enhancing the
37 management and control of air pollutants in the surrounding areas of Beijing will be
38 beneficial for further reducing air pollutant levels in Beijing and achieving the air
39 quality improvement targets set for the 14th Five-Year Plan period.

40 **Key words:** SO₂; Temporal and spatial distribution; Beijing; Trend; WRF/CMAQ;
41 Regional transport; Cooperative control

42

43 **1 Introduction**

44 Sulfur dioxide in the atmosphere primarily originates from coal-fired emissions
45 (Wang et al.,2017). High concentration of SO₂ and the sulfuric acid mist and
46 secondary aerosols formed by the transformation of SO₂ gas particles pose great
47 hazards to ecosystems and human health (Qian et al.,2021). Among the six
48 atmospheric pollutants, SO₂ is a significant air pollutant in China to be decreased and
49 be controlled first (China's Ministry of Environmental Protection,2022). Since 2000,
50 China has implemented strict SO₂ emission control measures. Especially since 2013,
51 the implementation of the Air Pollution Prevention and Control Action Plan, the
52 Three Year Action Plan to Win the Blue Sky Defense War and Deepening the Battle

53 of Pollution Prevention and Control Plan have significantly accelerated air pollution
54 control, reduced SO₂ emissions, and achieved significant improvement in air quality
55 (China State Council,2013; China State Council,2018; China State Council,2021).
56 However, due to variations in functional positioning and industrial structure, the
57 relative intensity of air pollution reduction measures and the changes in SO₂
58 concentration differ significantly among various regions and cities (Wei et al.,2023).

59 The Beijing Tianjin Hebei Urban Agglomeration is China's "capital economic
60 circle"(National Bureau of Statistics,2022). The significant decrease in SO₂
61 concentration in the Beijing, Tianjin, and Hebei regions has been the most notable
62 achievement in air pollution control in China in recent years; Furthermore, it has also
63 been the primary driving force behind the substantial reduction in regional particulate
64 matter concentration (Guo et al.,2014; Zhang et al.,2021). Taking Beijing, the capital
65 city of China, as an example, with the continuous implementation of local coal-fired
66 boiler elimination, coal-fired and gas-fired boiler renovation, "coal to gas" and "coal
67 to electricity" projects in Beijing, the SO₂ concentration in Beijing has decreased to
68 single digit levels in recent years (Miao et al.,2020; National Bureau of
69 Statistics,2022).

70 Research on changes in SO₂ concentration in the Beijing, Tianjin and Hebei
71 regions is of great significance for scientifically and effectively exploring the next
72 stage of air pollution control measures in China. Additionally, SO₂ can be utilized as a
73 tracer to analyze the impact of regional pollution transport on air quality in Beijing.
74 Especially during the heating season, the coal consumption in Beijing and
75 surrounding areas increases exponentially compared to the non-heating periods, and
76 sudden increase in SO₂ concentration and regional air pollution transport belt under
77 easterly and southerly winds poses a significant challenge to the air quality standards
78 in Beijing (Hao et al.,2007; Huang et al.,2009; Yang et al.,2017). In order to further
79 improve the air quality in the Beijing, Tianjin and Hebei regions, explore the ideas for
80 air pollution control during the 14th Five Year Plan period, and coordinate the air
81 pollution control work such as "treating winter diseases in summer" and "combining
82 peace and war", this study comprehensively analyzed the spatio-temporal variations
83 and main influencing factors of SO₂ concentration in Beijing and surrounding cities
84 from 2013 to 2022 by multiple methods such as statistical analysis, meteorological
85 filtering, and numerical simulation.

86

87 **2 Materials and methods**

88 **2.1 Instruments and observations**

89 The Beijing, Tianjin and Hebei regions includes 13 cities such as Beijing, Tianjin,
90 and Shijiazhuang city, with a total regional area of 183400 km² and a population of
91 110 million, accounting for approximately 10% of the country's GDP (National
92 Bureau of Statistics,2022). The SO₂ monitoring data of various cities in the region is
93 downloaded from the Air Quality Release Platform of the China Environmental
94 Monitoring Station (<https://air.cnemc.cn:18007/>) and the historical dataset of air
95 quality in China (<https://quotsoft.net/air/>). There are 11 monitoring stations in Beijing,
96 15 monitoring stations in Tianjin, and 53 monitoring stations in Hebei Province. The
97 arithmetic mean of all national control assessment points in each city represents the
98 pollution level of the city. Based on the pulsed fluorescence technology, the 43i SO₂
99 analyzer (Thermo Scientific™ 43i) were applied to monitor the SO₂ concentration.
100 The meteorological data applied in the study was downloaded from the China
101 Meteorological Data Sharing Network (<http://cdc.cma.gov.cn/>) and the statistical data
102 on coal consumption in various regions was sourced from the National Statistical
103 Yearbook (<http://www.stats.gov.cn/tjsj/ndsj/>).

104 **1.2 Daniel Trend Test**

105 In order to analyze the trends of air pollutant concentration, the Daniel trend test
106 was applied in this study. It is also called Spearman's rank correlation coefficient test.
107 Spearman's rank correlation coefficient, named after Charles Spearman, is usually
108 represented by Charles Spearman and applied to test and evaluate the correlation
109 between two groups of variables (Daniel et al.1990). Spearman coefficient is
110 applicable to continuous and discrete ordinal variables, which is defined as Pearson
111 correlation coefficient between sorting variables. The Daniel trend test is a commonly
112 method for quantitative analysis of specific time series data, which is mainly applied
113 for single factor small sample test (Spearman,1904).

114 The spearman's rank correlation coefficient is a non parametric (independent of
115 distribution) rank statistical parameter. The main principle states that if there are two
116 variables X and Y, and R_x and R_y are their respective ranks, then the correlation
coefficient between the two variables is calculated by equation 2.1.

$$r_R = \frac{\sum R_x R_y - \frac{\sum R_x \sum R_y}{N}}{\sqrt{\sum R_x^2 - \frac{(\sum R_x)^2}{N}} \times \sqrt{\sum R_y^2 - \frac{(\sum R_y)^2}{N}}} \quad (2.1)$$

117

118 When R_x and R_y do not have the same level, the formula is simplified as follows:

$$r_s = 1 - \left(6 \sum_{i=1}^n (x_i - y_i)^2 \right) / (n^3 - n) \quad (2.2)$$

119

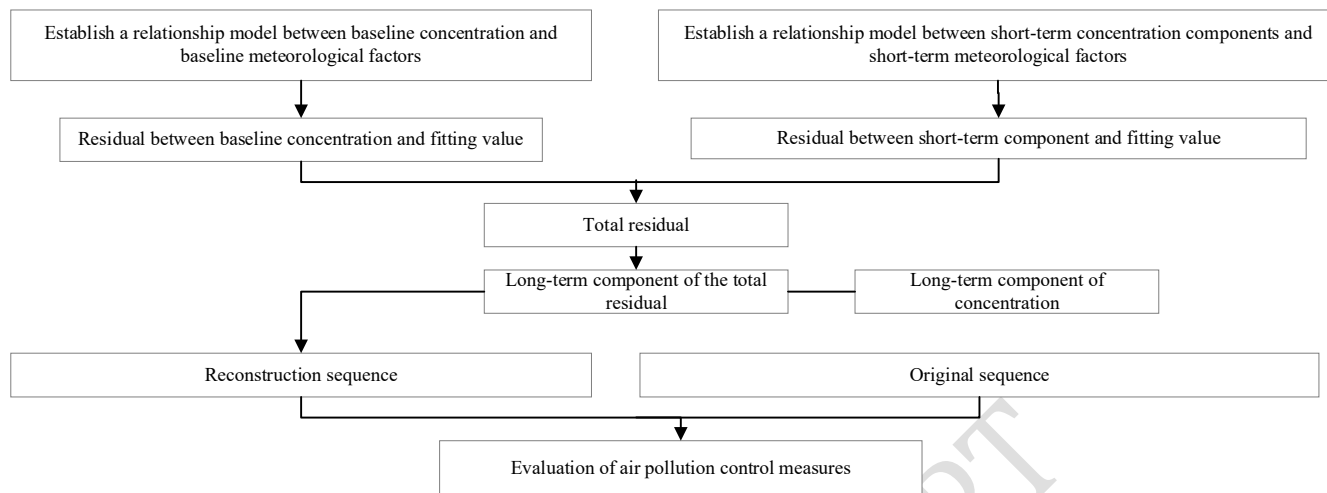
120 In the formula 2.1 and 2.2, r_s presents Spearman rank correlation coefficient; X_i
 121 presents sequence number of concentrations arranged from small to large; Y_i presents
 122 sequence number of concentration value arranged in chronological order; N presents
 123 the study period. According to the calculated rank correlation coefficient r_s , take its
 124 absolute value, and compare it with the critical value of r_s to determine the trends, that
 125 is, if $|r_s| \geq WP$ and R_s is positive, the data shows a significant upward trend; when $|r_s|$
 126 $\geq WP$ and r_s is negative, the data shows a significant downward trend.

127

128 1.3 Kolmogorov-Zurbenko filter method

129 To filter out the influence of meteorological factors, it is necessary to further
 130 adjust and reconstruct the observation sequence of atmospheric pollutants'
 131 concentration. The KZ filter is a statistical method based on iterative moving average
 132 algorithm to remove high-frequency changes in original data proposed by RAO et al.
 133 (1994). The KZ filtering method can further decompose the original series data of air
 134 pollutants into long-term components, seasonal components, and short-term
 135 components, which are independent of each other (Zurbenko et al., 1996). The study
 136 first established a multiple linear relationship between the short-term and baseline
 137 components of SO_2 concentration and meteorological factors, and then obtained the
 138 total residual. Subsequently, the authors applied the KZ filtering to the total residual
 139 to obtain the long-term component and added it to the long-term component of the
 140 original SO_2 time-series data to obtain a reconstructed sequence data after filtering out
 141 meteorological factors. The specific method and operation were shown in **Fig.1**.

142



143

144

Fig.1 The KZ filtering technology method in this study

145

1.4 WRF/CMAQ Model settings

146

147

148

149

150

151

152

153

154

155

156

157

158

159

160

161

162

163

164

165

166

167

The research selected the WRF/CMAQ Air Quality Simulation System to study the spatio-temporal variations and source apportionment of SO₂ in the Jingjinji regions. This model is a commonly and widely applied for the air quality numerical simulations (CEMPD, 2014; MMMD, 2014). The spatial resolution of the CMAQ simulation area in the study was 9km×9km, covering the Jingjinji regions and its surrounding areas. Due to the fact that the publicly available China regional scale list on the MEIC website (<http://www.meicmodel.org/>) is from 2017, the study used the year 2017 as the baseline scenario. The study employed CMAQ-ISAM (Integrated Source Apportionment Method) to calculate the local and regional contributions of atmospheric pollutant emissions in various regions within the simulated grids.

Taking the WRF-CMAQ benchmark scenario simulation verification results in 2017 as an example (**Table 1**), the ratio of the simulated PM_{2.5} and SO₂ values to the measured values was between 0.5 and 2.0, indicating a consistent fluctuation pattern on all time scales. The range of correlation coefficients between simulated and measured values of PM_{2.5} and SO₂ was 0.62~0.79, 0.64~0.71, respectively. The range of standardized average deviation was 0.1~1.0, 1.3~8.7, and the range of root mean square error was 24.9~70.2, 25.8~87.4, separately. Overall, the simulated SO₂ concentration in various cities was relatively higher, while the PM_{2.5} concentration was slightly lower. Much of the variation and discrepancy was primarily attributed to the uncertainty of regional emission inventories; overall, the simulated PM_{2.5} and SO₂ values in Beijing, Tianjin, and Shijiazhuang cities aligned well with the measured values.

Table 1 Comparisons between measured and simulated SO₂ and PM_{2.5} concentrations in 2017

Contaminant	City	Observed	Simulated	R	Standardized mean deviation	Root mean square error
PM _{2.5}	Beijing	58.0	57.2	0.73	0.1	24.9
	Tianjin	62.0	60.8	0.62	0.9	60.6
	Shijiazhuang	65.0	64.9	0.67	1.0	70.2
SO ₂	Beijing	4.0	5.8	0.71	1.3	25.8
	Tianjin	8.0	9.7	0.64	7.2	67.8
	Shijiazhuang	19.0	21.3	0.65	8.7	87.4

169

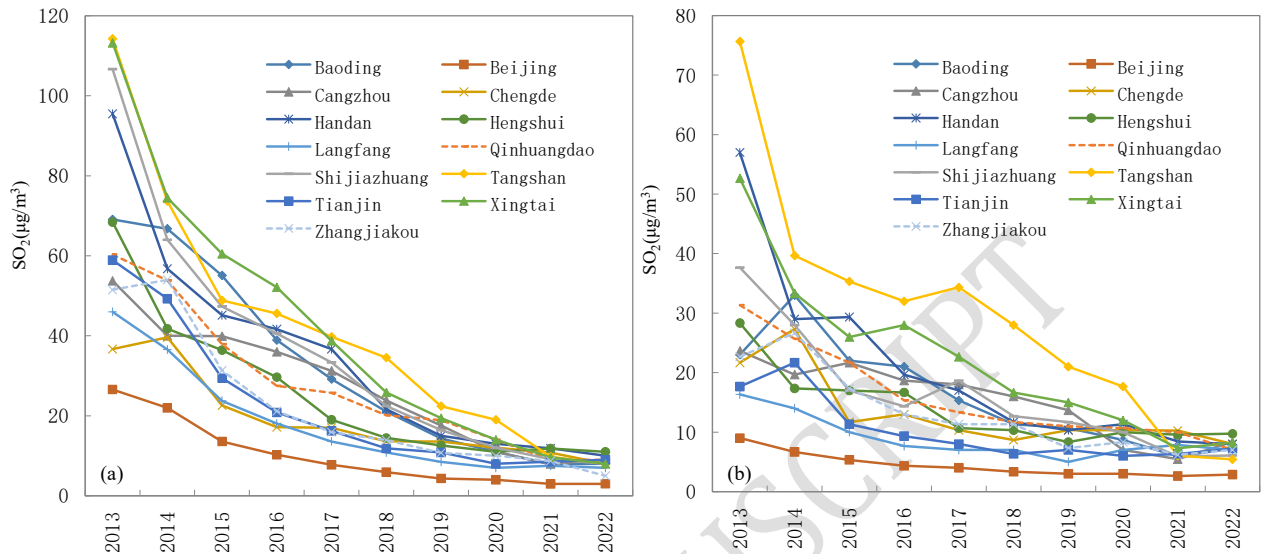
170 3 Results and Discussions

171 3.1 Spatio-temporal distribution of SO₂ concentration

172 As shown in **Fig.2**, from 2013 to 2022, the annual averaged concentration of SO₂
 173 in all cities in the Jingjinji regions presented significant downward trends, from
 174 26.6~114.3 $\mu\text{g}\cdot\text{m}^{-3}$ in 2013 decreases to 3.0~11.0 $\mu\text{g}\cdot\text{m}^{-3}$ in 2022. The cumulative
 175 decrease in SO₂ concentration in Tangshan was the largest, reaching 92.3% while it
 176 changed to be the smallest in Chengde, reaching 78.2% during 2013~2022. Except for
 177 Handan and Hengshui cities, the SO₂ concentrations in other 11 cities were all at the
 178 single digit levels, with the lowest annual averaged SO₂ concentration at 3.0 $\mu\text{g}\cdot\text{m}^{-3}$ in
 179 Beijing in 2022. The decrease trends of SO₂ annual averaged concentration in 13
 180 cities from 2013 to 2022 all passed the Daniel trend tests ($\alpha=0.01$) with the highest
 181 declining rate of 11.81 $\mu\text{g}/(\text{m}^3\cdot\text{yr})$ in Tangshan city and the smallest descent rate of
 182 2.62 $\mu\text{g}/(\text{m}^3\cdot\text{yr})$ in Beijing city.

183 Compared to the annual averaged concentrations, the overall SO₂ concentrations
 184 during the summer periods were at a lower level throughout the year. As shown in
 185 **Table 2**, from 2013 to 2022, the averaged concentration of SO₂ during summer
 186 seasons in 13 cities also presented significant downward trends, from 9.0~75.7 $\mu\text{g}\cdot\text{m}^{-3}$
 187 in 2013 decreases to 3.0~9.7 $\mu\text{g}\cdot\text{m}^{-3}$ in 2022. The cumulative decrease in SO₂
 188 concentration in Tangshan was the largest, reaching 91.3% while it changed to be the
 189 smallest in Tianjin, reaching 55.9% during 2013~2022. Similarly, the decrease trends
 190 of SO₂ concentration during summer seasons in 13 cities from 2013 to 2022 all passed
 191 the Daniel trend tests ($\alpha=0.01$) with the highest declining rate of 7.80 $\mu\text{g}/(\text{m}^3\cdot\text{yr})$ in
 192 Tangshan and the smallest speed of 0.68 $\mu\text{g}/(\text{m}^3\cdot\text{yr})$ in Beijing. Compared to the whole

193 year, the decrease rate of SO₂ concentrations in summer was relatively lower, which
 194 was mainly related to the low coal consumption in summer.
 195



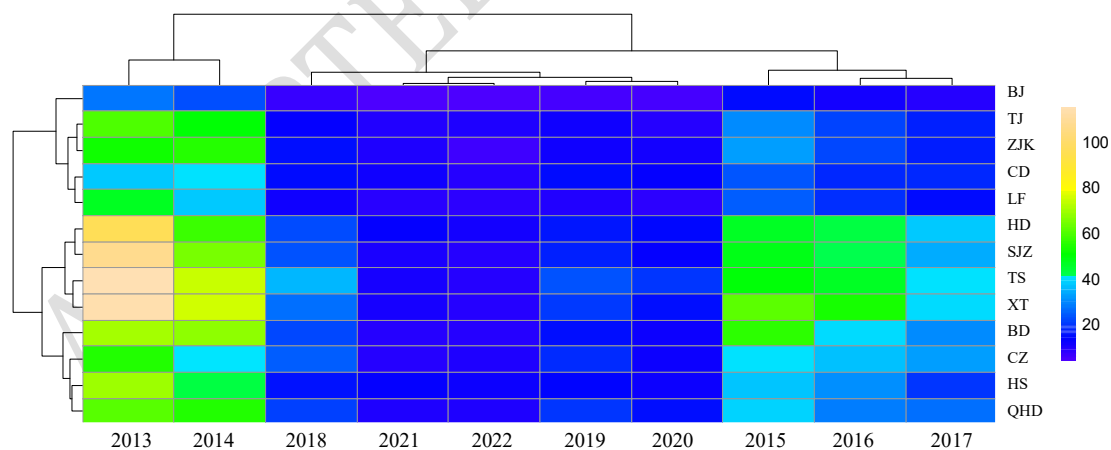
196
 197 **Fig. 2 Variation of SO₂ concentration in Beijing, Tianjin and Hebei regions from 2013 to 2022**
 198 **(a. annual averaged concentration; b. averaged concentration during Summer)**

199 **Table 2 Daniel trend tests of SO₂ concentration in 13 cities from 2013 to 2022**

Cities	Annual comparison				Summer comparison			
	rs	Wp($\alpha=0.01$)	k	Trend	rs	Wp($\alpha=0.01$)	k	Trend
Baoding	-1.000	0.834	-6.79	Significant decrease	-0.976	0.834	-1.75	Significant decrease
Beijing	-1.000	0.834	-2.62	Significant decrease	-1.131	0.834	-0.68	Significant decrease
Cangzhou	-1.000	0.834	-4.97	Significant decrease	-0.976	0.834	-1.70	Significant decrease
Chengde	-0.976	0.834	-3.39	Significant decrease	-1.083	0.834	-1.84	Significant decrease
Handan	-1.000	0.834	-9.50	Significant decrease	-0.952	0.834	-5.45	Significant decrease
Hengshui	-1.000	0.834	-6.38	Significant decrease	-0.976	0.834	-2.07	Significant decrease
Langfang	-1.000	0.834	-4.33	Significant decrease	-1.155	0.834	-1.04	Significant decrease
Qinhuangdao	-1.000	0.834	-5.92	Significant decrease	-1.000	0.834	-2.84	Significant decrease
Shijiazhuang	-1.000	0.834	-10.96	Significant decrease	-0.929	0.834	-3.51	Significant decrease
Tangshan	-1.000	0.834	-11.81	Significant decrease	-0.976	0.834	-7.80	Significant decrease
Tianjin	-1.000	0.834	-5.55	Significant decrease	-0.952	0.834	-1.16	Significant decrease
Xingtai	-1.000	0.834	-11.69	Significant decrease	-0.976	0.834	-4.94	Significant decrease
Zhangjiakou	-0.976	0.834	-5.50	Significant decrease	-0.988	0.834	-2.54	Significant decrease

200 For the spatial distribution of SO₂ concentrations based on the Euclid Distance
 201 with weights, the 13 cities in the Jingjinji regions can be grouped into three categories.

202 The concentrations of SO₂ in Beijing, Zhangjiakou, Chengde, Langfang, and Tianjin
 203 were relatively low and clustered into the first category. The northern cities of the
 204 Jingjinji regions is in a relatively complete natural landscape, a good ecological
 205 environment, and low intensity of industrial activities. The impact of human and
 206 natural factors on SO₂ concentration in these areas was relatively minimal, with
 207 relatively low inter-annual fluctuations. The four cities of Handan, Tangshan, Xingtai,
 208 and Shijiazhuang were in relatively heavy SO₂ pollution, and their SO₂ concentrations
 209 were clustered into the second category; furthermore, Tangshan and Xingtai were the
 210 two high-value centers in the Beijing, Tianjin and Hebei regions, with the highest SO₂
 211 concentration levels. These two cities are mainly concentrated in the southern and
 212 eastern parts of the Beijing, Tianjin and Hebei regions, which have a significant
 213 proportion of heavy industry and a large amount of coal consumption. The left four
 214 cities of Baoding, Cangzhou, Hengshui, and Qinhuangdao were clustered into the
 215 third category. In addition, the temporal distribution of SO₂ concentrations could also
 216 be divided into three categories: 2013~2014, 2015~2017, and 2018~2022. The
 217 gradient of annual averaged SO₂ concentration in 13 cities in 2013 was 87.7 $\mu\text{g}\cdot\text{m}^{-3}$
 218 and it decreased to 8.0 $\mu\text{g}\cdot\text{m}^{-3}$ in 2022 while it was 66.7 $\mu\text{g}\cdot\text{m}^{-3}$ and 6.8 $\mu\text{g}\cdot\text{m}^{-3}$ during
 219 summer seasons in 2013 and 2022 respectively, which revealed the decreasing
 220 gradient difference in north to south spatial direction year by year.



221
 222 **Fig.3** Spatial clustering of SO₂ annual average concentration in the Beijing-Tianjin-Hebei regions (BJ:
 223 Beijing, TJ: Tianjin, ZJK: Zhangjiakou, CD: Chengde, LF: Langfang, Hd: Handan, SJZ: Shijiazhuang,
 224 TS: Tangshan, XT: Xingtai, BD: Baoding, CZ: Cangzhou, HS: Hengshui, QHD: Qinhuangdao)

225 3.2 Impact of meteorological and emission reduction measures

226 After filtering out meteorological factors through KZ filtering method (as shown
 227 in **Table 3**), the decrease rate of annual averaged SO₂ concentration in 13 cities in

228 Jingjinji regions changed from 78.2~92.3% in the original observation sequence to
229 72.2~92.0% in the reconstructed sequence from 2013 to 2022. Meteorological factors
230 decreased 0.3~8.5% to the accumulated drop of SO₂ concentration in 13 cities while
231 the human efforts to reduce emissions contributed about 94.5~99.6% to the annual
232 decrease in SO₂ concentration during 2013~2022. During summer seasons, the
233 decrease rate of annual averaged SO₂ concentration in 13 cities changed from
234 55.9~91.3% in the original observation sequence to 72.2~92.2% in the reconstructed
235 sequence. Accordingly, the meteorological factors increased 0.9~28.2% to
236 accumulated drop of SO₂ concentration in 13 cities while the human efforts to reduce
237 emissions contributed about 100% to the decrease in SO₂ concentration. Compared
238 with the whole year, there was no coal-fired heating consumption in the Beijing,
239 Tianjin and Hebei regions during summer seasons, and the diffusion of SO₂
240 concentration was more susceptible to meteorological conditions such as the East
241 Asian monsoon. Under the dominant southerly wind direction affected by the East
242 Asian monsoon, cities in the northern part of the region were significantly affected by
243 the regional transport which was the unfavorable meteorological conditions for the
244 SO₂ concentration (Bai et al., 2022; Huang et al., 2020; Shen et al.,2021).

245 From 1998 to 2012, Beijing completed sixteen stages of air pollution prevention
246 and control measures (BMBS,2022). Starting from 2013, the Beijing Clean Air Action
247 Plan for 2013~2017 was implemented, systematically carrying out air pollution
248 prevention and control work, with a focus on reducing coal consumption, controlling
249 vehicles and oil consumption, pollution control and emission reduction, and clean
250 dust reduction. Starting from 2016, Beijing implemented the "Comprehensive Action
251 Plan for Autumn and Winter Air Pollution Control in the Beijing Tianjin Hebei and
252 Surrounding Areas". In 2018, the "Three Year Action Plan for Winning the Blue Sky
253 Defense War in Beijing (2018~2020)" was implemented. From 2021 to 2023, Beijing
254 continued to implement the three-year action plan to deepen the battle against air
255 pollution. The total coal consumption in Beijing decreased from 2019 million tons in
256 2013 to approximately 1 million tons in 2022, with a cumulative reduction of 95.0%.
257 In 2015, the core area was basically free of coal, and the sixth urban area had no
258 coal-fired boilers. In 2017, the plain region achieved coal free transformation,
259 becoming the first region in the country to basically solve the problem of coal
260 pollution. According to National Statistical Yearbook Data, the total coal consumption

261 in Tianjin decreased from 52.78 million tons in 2013 to approximately 37.23 million
 262 tons in 2020, with a cumulative decrease of 29.5%. Since 2018, all scattered coal in
 263 urban and rural areas in Tianjin was cleared to be zero. The total coal consumption in
 264 Hebei Province decreased from 317 million tons in 2013 to approximately 260
 265 million tons in 2020, with a cumulative reduction of 18%. By implementing measures
 266 such as coal to gas and coal to electricity projects, the government in Hebei
 267 significantly promoted the transformation of new energy, and by the end of 2020, the
 268 scattered coal in rural areas of this province's plains was similarly cleared to zero. As
 269 a result, the atmospheric SO₂ emissions in Beijing, Tianjin, and Hebei regions
 270 decreased significantly from 8.70, 21.68, and 128.5 million tons in 2013 to 0.11, 0.65,
 271 and 14.6 million tons in 2022 (**Fig.4**). From the correlation between coal consumption
 272 and annual averaged SO₂ concentration in the three regions (**Table 4**), the Pearson
 273 correlation coefficients in Beijing, Tianjin, and Hebei provinces in recent years were
 274 0.974**, 0.926**, and 0.980**, respectively. The Pearson correlation coefficients
 275 between SO₂ emissions and annual averaged SO₂ concentrations were 0.924**,
 276 0.891**, and 0.939**, respectively. These statistical data indicated that the significant
 277 reduction in coal consumption in the Beijing, Tianjin and Hebei regions had led to a
 278 significant reduction in atmospheric SO₂ emissions, and then directly reduced the
 279 average concentration of SO₂ in the region.

280 **Table3 Cumulative decrease of SO₂ original and filtered concentrations at different cities in this**
 281 **study**

Period	Parameter/%	BJ	TJ	SJZ	TS	HD	XT	BD	CZ	LF	HS	CD	QHD	ZJK
Annual	Decrease of original sequence	89.5	84.8	92.1	92.3	89.7	87.3	87.7	83.4	84.4	83.9	78.2	81.0	90.3
	Decrease of reconstructed series	89.0	84.0	91.4	92.0	88.2	85.6	87.4	82.8	83.9	81.9	72.2	79.4	81.8
	Decrease of meteorological factors (favorable)	0.5	0.8	0.7	0.3	1.5	1.7	0.3	0.6	0.5	2	6	1.6	8.5
	Contribution of anthropogenic emission reduction measures	96.2	95.7	94.5	97.0	94.8	94.7	95.9	96.1	95.3	94.6	99.6	96.3	97.2
Summer	Decrease of original sequence	66.9	55.9	83.6	91.3	86.1	79.3	71.9	67.7	57.5	65.5	63.1	75.8	74.4
	Decrease of reconstructed series	89.2	84.1	91.5	92.2	88.4	85.8	87.3	82.6	83.9	82.1	72.2	79.9	82.3
	Increase of meteorological factors (unfavorable)	22.3	28.2	7.9	0.9	2.3	6.5	15.4	14.9	26.4	16.6	9.1	4.1	7.9

Contribution of anthropogenic emission reduction measures	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0
---	-------	-------	-------	-------	-------	-------	-------	-------	-------	-------	-------	-------	-------	-------

282 (BJ: Beijing, TJ: Tianjin, ZJK: Zhangjiakou, CD: Chengde, LF: Langfang, Hd: Hangdan, SJZ:
 283 Shijiazhuang, TS: Tangshan, XT: Xingtai, BD: Baoding, CZ: Cangzhou, HS: Hengshui, QHD:
 284 Qinhuangdao)

285 **Table4 Correlation statistics between main air pollutants and meteorological, social factors from**
 286 **2013 to 2022**

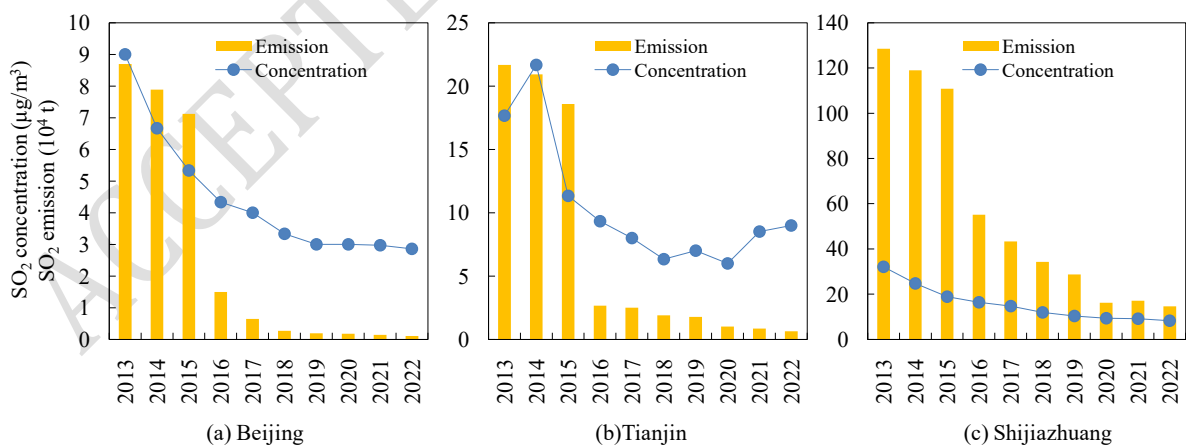
Correlation coefficient	Wind speed	T	Surface pressure	Relative humidity	Precipitation	SO ₂ emissions #	Coal consumption
Beijing	0.460	-0.599	-0.866*	0.808*	0.121	0.924**	0.974**
Tianjin	0.124	-0.380	-0.267	0.661*	-0.117	0.891**	0.926**
Shijiazhuang	0.827*	-0.727	-0.675	0.514	0.206	0.939**	0.980**

287 Note: * * and * indicated significant correlation at the 0.01 and 0.05 levels (bilateral) respectively.

288 #SO₂ emissions and coal consumption data were downloaded from the website

289 (<https://data.stats.gov.cn/easyquery.htm?cn>).

290 The coal consumption data was the statistical data up to 2020.



291

292 **Fig.4 The annual averaged concentration and emissions of SO₂ in three cities from 2013 to 2022**

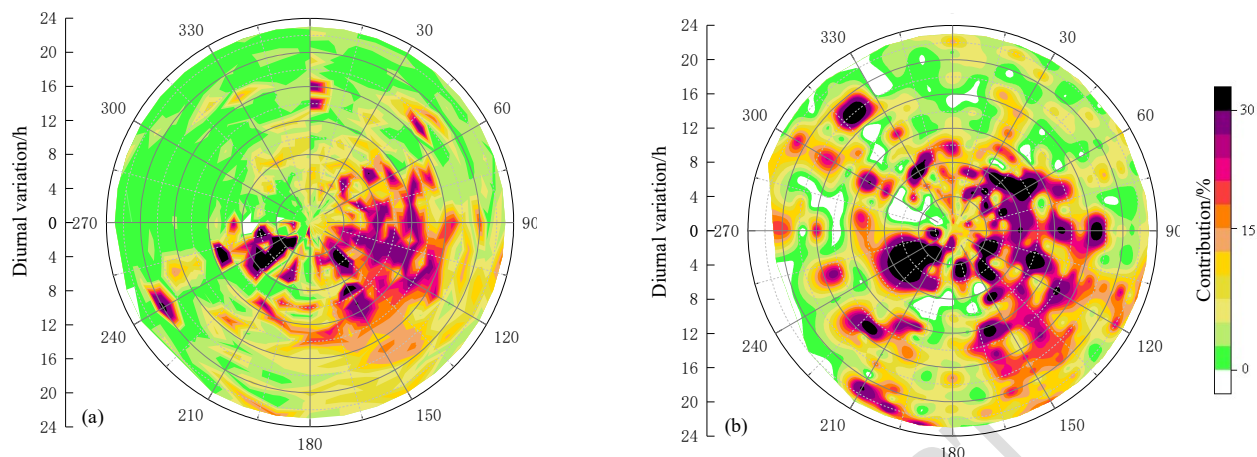
293

294 **3.3 Regional transport of SO₂ concentration**

295 In addition to anthropogenic emissions reduction and meteorological impacts,
 296 regional transport is also an important factor affecting the distribution of SO₂

297 concentration in the Beijing, Tianjin and Hebei regions. The simulation results of the
298 WRF-CMAQ benchmark scenario showed that in 2017, the local contributions of SO₂
299 in Beijing, Tianjin, and Shijiazhuang were 69.0±8.8%, 70.9±8.9% and 81.5±4.1%,
300 respectively. In summer, the local contributions of SO₂ in Beijing, Tianjin, and
301 Shijiazhuang were 55.9±8.5%, 62.6±6.1%, and 72.2±3.8%, respectively. Among the
302 three cities, the local contribution of SO₂ was largest in Shijiazhuang city while it was
303 smallest in Beijing city. Compared with the whole year, the contribution of SO₂
304 regional transport in summer in three cities significantly increased by 10~20%. The
305 concentration of SO₂ in Beijing was mainly affected by the transport of neighboring
306 areas such as Baoding, Tangshan, Langfang, and Tianjin cities. The concentration of
307 SO₂ in Tianjin was mainly affected by the transport of neighboring areas such as
308 Tangshan, Langfang, and Shandong Tianjin cities. The concentration of SO₂ in
309 Shijiazhuang was mainly affected by the transport of neighboring areas such as Henan,
310 Shanxi, Xingtai, and Handan cities in Beijing, Tianjin and Hebei regions.

311 In 2017, the average frequency of wind directions at ground level in Beijing,
312 from highest to lowest, was southerly, northerly, easterly, and westerly, with values of
313 32.5%, 30.4%, 29%, and 7.3%, respectively (Fig.5). This study further classified
314 Baoding, southern Hebei, and Henan province as southwest regional transport
315 channels, and Langfang, Tianjin, Tangshan, Cangzhou, and Shandong districts as
316 southeast regional transport channels. The numerical simulation results demonstrated
317 that the contribution of the southwest transport channel to the concentration of SO₂ in
318 Beijing during summer was about 33.5%, and the contribution of the southeast
319 transport channel to the concentration of SO₂ in Beijing was about 38.9% in 2017.
320 The higher regional transport contribution was mainly concentrated in the wind
321 direction of 60~150 degrees. Due to the high position of the elevated source exhaust
322 pipe in the Beijing, Tianjin and Hebei regions, the atmospheric pollutants were
323 transported further and led to a larger impact under the influence of higher wind
324 speeds in summer. The results of this study were comparable and reliable compared
325 with other relevant literature on SO₂ regional transport (Lin et al., 2019; Tan et
326 al., 2020).



327

328 **Fig.5 Regional transport contribution of SO₂ to Beijing under different wind directions in 2017**

329 **(a. the whole year; b. the summer period)**

330 **4 Conclusions**

331 China is the third largest emitter of SO₂ in the world, contributing approximately
 332 8% of global anthropogenic SO₂ emissions, second only to India and Russia
 333 (Greenpeace Environment Trust,2019). The Beijing, Tianjin and Hebei region is a
 334 world-class urban agglomeration centered around the capital city of Beijing. It is a
 335 new engine for innovation driven economic growth and a demonstration area for
 336 ecological restoration and environmental improvement in China. In 2022, the
 337 proportion of Beijing, Tianjin and Hebei regions to the national GDP was 8.3%, and
 338 the SO₂ emissions in the Beijing, Tianjin and Hebei regions accounted for 6.3% of the
 339 country (National Bureau of Statistics, 2022).

340 This study analyzed the spatial-temporal distribution of SO₂ concentration and
 341 main influencing factors in the Beijing, Tianjin and Hebei regions through
 342 multi-methods of statistics, KZ filtering and model simulations. From 2013 to 2022,
 343 the annual averaged concentrations of SO₂ in Beijing, Tianjin and Hebei regions were
 344 all in obvious downward trends, and all passed the Daniel trend test ($\alpha=0.01$). The
 345 annual averaged concentration of SO₂ in Beijing, Tianjin and Hebei regions decreased
 346 from 26.6~114.3 $\mu\text{g}\cdot\text{m}^{-3}$ in 2013 to 3.0~11.0 $\mu\text{g}\cdot\text{m}^{-3}$, with the decline rates fluctuating
 347 from 2.62~11.81 $\mu\text{g}/(\text{m}^3\cdot\text{yr})$. The reduction of coal consumption in the
 348 Beijing-Tianjin-Hebei regions significantly reduced atmospheric SO₂ emissions and
 349 directly reduced the regional average concentration of SO₂. Meteorological factors
 350 were generally conducive to the diffusion of SO₂ concentration during 2013~2022,

351 and the meteorological factors contributed about 0.4~5.5% in 13 cities while the
352 contribution of anthropogenic emission reduction changed to 94.5~99.6%. Regional
353 transport is an important factor affecting the distribution of SO₂ concentration in the
354 region. During the summer months from June to August, under easterly and southerly
355 winds, the SO₂ concentration in Beijing was approximately twice as high as that
356 under northerly and westerly winds. The contribution of the southwest transport
357 channel to the SO₂ concentration in Beijing was 33.5%, which increased to 38.9% in
358 the southeast transport channel.

359 To further enhance regional air quality, it is recommended to continue reducing
360 coal consumption in the Beijing, Tianjin, and Hebei regions, particularly in rural areas,
361 while ensuring energy security. Additionally, it is essential to solidify the progress
362 made in transitioning to cleaner energy sources in flat areas and prevent a resurgence
363 of uncontrolled coal combustion. The government should take emergency measures to
364 reduce heavy air pollution episodes during autumn and winter seasons. During the
365 autumn and winter months, a significant amount of SO₂ emitted from coal combustion
366 in key areas is converted into sulfate under suitable conditions, which can contribute
367 to an increase in particle concentration. Early prediction and forecasting, coupled with
368 the activation of emergency emission reduction measures in advance, can help
369 mitigate the accumulation rate of sulfate. Moreover, enhancing the management and
370 control of air pollutants in the surrounding areas of Beijing can aid in further reducing
371 pollutant levels and achieving air quality improvement targets in the region.

372 This research has observed that SO₂ concentrations at many sites are below the
373 national environmental air quality standard limits, with some even reaching
374 single-digit levels in the Beijing, Tianjin, and Hebei regions. It is recommended to
375 further optimize the placement of monitoring stations, with a focus on locating SO₂
376 monitoring stations in urban-rural fringe areas or rural areas, while gradually reducing
377 SO₂ monitoring instruments in urban areas. Against the backdrop of national policies
378 promoting carbon peak and carbon neutrality, reducing coal consumption can not only
379 lower carbon emission intensity but also decrease SO₂ emissions, therefore, strategic
380 planning should prioritize coordinated and regional control of SO₂ and CO₂
381 concentrations during the 14th Five-Year Plan period.

382

383 **Acknowledgements:** This work was supported by the “Fundamental Research Funds

384 for the Central Universities” (No. 3142023017, 3142021002, 3142023021), the
385 “Hebei Province Higher Education Science and Technology Research Project”
386 (ZC2024016), and the supported by “the Central Government Guides Local Funds for
387 Science and Technology Development” (No. 236Z0307G).

388 **Author Contributions**

389 Bingfen Cheng and Guiqiang Zheng designed the study, performed data analysis and
390 wrote the manuscript. Other coauthors contributed to the research design,
391 proofreading and revision.

392 **Additional Information**

393 Competing Interests: The authors declare no competing interests.

394 **References:**

- 395 Bai Y, Zhao T, Hu W, et al. 2022. Meteorological mechanism of regional PM_{2.5}
396 transport building a receptor region for heavy air pollution over Central
397 China. *Science of the Total Environment*,808,151951.
- 398 BMBS (Beijing Municipal Bureau of Statistics). 2022. Beijing Statistical Yearbo
399 ok, <http://www.bjstats.gov.cn>.
- 400 CEMPD, UNC. 2014.Operational guidance for the Community Multiscale Air Q
401 uality (CMAQ) modeling system. [http://www.cmascenter.org/help/model_do](http://www.cmascenter.org/help/model_docs/cmaq/4.7.1/CMAQ_4.7.1_OGD_28june10.pdf)
402 [cs/cmaq/4.7.1/CMAQ_4.7.1_OGD_28june10.pdf](http://www.cmascenter.org/help/model_docs/cmaq/4.7.1/CMAQ_4.7.1_OGD_28june10.pdf), 2014-09-01.
- 403 China's Ministry of Environmental Protection. 2022. China Environment Yearbo
404 ok. The China Environment Yearbook Press, Beijing.
- 405 China State Council. 2013.Air Pollution Prevention and Control Action Plan. [ht](https://www.gov.cn/zhengce/content/2013-09/13/content_4561.htm)
406 [tps://www.gov.cn/zhengce/content/2013-09/13/content_4561.htm](https://www.gov.cn/zhengce/content/2013-09/13/content_4561.htm) (accessed Jan
407 uary 13, 2013).
- 408 China State Council. 2018.Triennial Action Plan to Win the Battle of Blue Sky
409 Defense. [https://www.gov.cn/zhengce/zhengceku/2018-07/03/content_5303158.](https://www.gov.cn/zhengce/zhengceku/2018-07/03/content_5303158.htm)
410 [htm](https://www.gov.cn/zhengce/zhengceku/2018-07/03/content_5303158.htm) (accessed July 3, 2018).
- 411 China State Council. 2021. Opinions of the Central Committee of the Commun
412 ist Party of China and the State Council on Deepening the Battle of Pollu
413 tion Prevention and Control. [https://www.gov.cn/gongbao/content/2021/conten](https://www.gov.cn/gongbao/content/2021/content_5651723.htm)
414 [t_5651723.htm](https://www.gov.cn/gongbao/content/2021/content_5651723.htm) (accessed November 2, 2021).

415 Daniel, Wayne W.1990."Spearman rank correlation coefficient".Applied Nonpara
416 metric Statistics (2nd ed.) Boston: PWS-Kent, 358–365. ISBN 0-534-91976
417 -6.

418 Greenpeace Environment Trust (GET). 2019. Global SO₂ emission hotspot data
419 base: Ranking the world's worst sources of SO₂ pollution.Greenpeace Envir
420 onment Trust. [https://www.greenpeace.org.au/wp/wp-content/uploads/2019/08/
421 Global-Hotspot-and-Emission-Sources-for-SO₂_August-2019_AU_final.pdf](https://www.greenpeace.org.au/wp/wp-content/uploads/2019/08/Global-Hotspot-and-Emission-Sources-for-SO2_August-2019_AU_final.pdf).

422 Guo S,Hu M,Zamora ML,*et al.* 2014. Elucidating severe urban haze formation
423 in China[J].Proceedings of the National Academy of Sciences of the Unite
424 d States of America,111:17373-17378.

425 Hao J M, Wang L T, Shen M J, *et al.* 2007.Air quality impacts of power pla
426 nt emissions in Beijing. Environmental Pollution,147:401-408.

427 Huang Q,Cheng S Y,Chen D S,*et al.* 2009.Beijing SO₂ Source Apportionment f
428 rom Regional Power Plants During Heating Season.Reserrch of Environme
429 ntal Science, 22(5):567-573.

430 Huang X, Ding A, Wang Z, *et al.* 2020. Amplified transboundary transport of
431 haze by aerosol–boundary layer interaction in China.Nature Geoscience ,13,
432 428-434.

433 Lin C A, Chen Y C, Liu C Y, *et al.*, 2019. Satellite-derived correlation of SO
434 2, NO₂, and aerosol optical depth with meteorological conditions over Eas
435 t Asia from 2005 to 2015. Remote Sensing,11 (15):1–21.

436 Miao Z, Liu S C, Chen X D *et al.* 2020. Driving factors and spatio-temporal
437 features underlying industrial SO₂ emissions in “2+26” in North China and
438 extended cities. Chinese Journal of Population, Resources and Environmen
439 t, 2020,18(4):1-9.

440 MMMD, NCAR. 2014.Weather Research and Forecast ARW Version 3 Modelin
441 g Systems User's Guide. [http://www.mmm.ucar.edu/wrf/users/docs/user-gui
442 de-V3/ARWUsers-GuideV3.pdf](http://www.mmm.ucar.edu/wrf/users/docs/user-guide-V3/ARWUsers-GuideV3.pdf), 2014-09-01.

443 National Bureau of Statistics. 2022. China City Statistical Yearbook. ChinaStatis
444 tics Press, Beijing.

445 Qian H Q, Xu S D, Cao J, *et al.* 2021.Air pollution reduction and climate co-
446 benefits in China's industries. Nature Sustainability,4(5):1-6.

447 Rao S T, Zurbenko I G.1994.Detecting and Tracking Changes in Ozone Air
448 Quality.Air Waste, 44:1089-1092.

449 Shen L, Zhao T, Wang H,et al. 2021.Importance of meteorology in air pollutio
450 n events during the city lockdown for COVID-19 in Hubei Province, Cent
451 ral China.Science of the Total Environment,754, 142227.

452 Spearman C. 1904. The proof and measurement of association between two thi
453 ngs. Journal of Applied Psychology, 15(1): 72-101.

454 Tan W,Liu C,Wang S S,et al.,2020. Long-distance mobile MAX-DOAS observ
455 ations of NO₂ and SO₂ over the North China Plain and identification of r
456 egional transport and power plant emissions.Atmospheric Research,245:1050
457 37.

458 Wang J, Zhao B, Wang S, *et al.* 2017.Particulate matter pollution over China a
459 nd the effects of control policies. Science of the Total Environment, 584-5
460 85: 426.

461 Wei Y X, Zhang X Y, Zhang H ,*et al.* 2023.Spatial and temporal distribution
462 of sulfur dioxide and main emission sources in China. China Environment
463 al Science,43(11):5678-5686.

464 Yang X,Wang S J, Zhang W Z, *et al.*2017.Are the temporal variation and spati
465 al variation of ambient SO₂ concentrations determined by different factors?
466 Journal of Cleaner Production,167:824-836.

467 Zhang X Y, Wang Z, Cheng M M, *et al.* 2021. Long-term ambient SO₂ conce
468 ntration and its exposure risk across China inferred from OMI observation
469 s from 2005 to 2018. Atmospheric Research,247:20-30.

470 Zurbenko I, Chen J, Rao S T,*et al.* 1996.Detecting discontinuities in time serie
471 s of upper air data: Demonstration of an adaptive filter technique. Journal
472 of Climate,9:3548-3560.