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# 1 Rapid improvement in air quality due to aerosol- 2 pollution control during 2012-2018: an evidence 3 observed in Kunshan in the Yangtze River Delta, China

4

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12

## 13 **Abstract**

14 China's severe air pollution was widely concerned in the past decade. A rigorous emission  
15 control has been implemented by the Chinese government since 2013. It is essential to  
16 evaluate changes of air pollutants to understand effectiveness of present air-pollution  
17 control on improving air quality. Here, we investigate temporal trends of air pollutants,  
18 including PM<sub>2.5</sub>, SO<sub>2</sub>, NO<sub>2</sub> and O<sub>3</sub>, observed at 12 sites over the Kunshan area in the Yangtze  
19 River Delta region during 2012-2018 using Mann-Kendall statistical test. Overall, significant  
20 reduction trends of monthly PM<sub>2.5</sub> (-7.4% yr<sup>-1</sup>) were observed, together with reduction in the  
21 PM<sub>2.5</sub> to CO ratio (-5.8% yr<sup>-1</sup>), an indicator of secondary aerosol production. Secondary  
22 aerosol precursors, SO<sub>2</sub> (-10.3% yr<sup>-1</sup>) and NO<sub>2</sub> (-4.4% yr<sup>-1</sup>) also presented statistically  
23 significant reduction trends. These results reflect the consequence of emission control that  
24 leads to substantial reduction in the bulk PM<sub>2.5</sub> concentration, as well as secondary aerosols  
25 likely formed from SO<sub>2</sub> and NO<sub>2</sub>. However, O<sub>3</sub> had statistically significant increase trend (+3.4%  
26 yr<sup>-1</sup>) during 2014 – 2018. Limited formation of O<sub>3</sub> under high PM<sub>2.5</sub> and NO<sub>2</sub> concentrations

27 condition was found at daytime in summer, which might reflect one of the reasons causing  
28 the increase trend of O<sub>3</sub> under the current reduction scenario of PM<sub>2.5</sub> and NO<sub>2</sub>. Potential  
29 source contribution function analysis demonstrated that the transport from the regions  
30 located to northwest of Kunshan could contribute to high concentrations of PM<sub>2.5</sub> in all  
31 seasons, while the south and southeast could be the high potential as source areas for O<sub>3</sub>.

32

33 **Keywords:** Air quality, PM<sub>2.5</sub>, O<sub>3</sub>, trend, geographic origins, Yangtze River Delta

34

## 35 1. Introduction

36 Rapid industrialization and urbanization have been widely known as a key reason  
37 leading to air pollution, especially in developing countries. Air pollutants compose of various  
38 gases and particulate matter. Globally, fine aerosol particles (PM<sub>2.5</sub>, with the aerodynamic  
39 diameter  $\leq 2.5 \mu\text{m}$ ) and ozone (O<sub>3</sub>) are the major air pollutants in the present atmosphere.  
40 Both of them, PM<sub>2.5</sub> and O<sub>3</sub>, have adverse health risks that can also result in human  
41 premature death (Shiraiwa et al., 2017; Dockery et al., 1996; Knowlton et al., 2004). As one of  
42 the largest developing countries in the world, China has experienced fast-developing  
43 economy and urbanization during the past decades, especially in the eastern China regions,  
44 like the Yangtze River Delta. However, this obviously brings a serious consequence being air  
45 pollution issues, such as extreme haze (Huang et al., 2014) and O<sub>3</sub> pollution (Li et al., 2019).

46 During the past decade, characterization of Chinese severe haze pollution and its  
47 formation mechanism have been widely studied (Sun et al., 2014; Huang et al., 2014; Guo et  
48 al., 2014; Ding et al., 2016; Zhang et al., 2017a; Sun et al., 2016a; Wang et al., 2016b; Cheng et  
49 al., 2016). As indicated by their findings, rapid formation of anthropogenic secondary fine  
50 aerosols over a regional scale is a dominated factor for the haze event formation under  
51 stagnant meteorological conditions. In the atmosphere, secondary fine aerosols are formed  
52 via the oxidation of their gaseous precursors, e.g., sulfur dioxide (SO<sub>2</sub>), nitrogen oxides (NO<sub>x</sub>),  
53 ammonia (NH<sub>3</sub>) and volatile organic compounds (VOCs). To control haze formation and to  
54 further improve air quality, it is therefore necessary to reduce the budget of those aerosol  
55 precursors in the atmosphere (Cheng et al., 2019; Zhai et al., 2019). Several mankind  
56 emission source sectors, such as power plants, industry, transportation, agriculture, solvent  
57 use, residential activities, contribute to those aerosol precursors in China (Zheng et al., 2018).  
58 Since 2013, the China's Clean Air Action has been implemented to reduce emissions of air  
59 pollutants from these source sectors. As reported by Zheng et al. (2018), such emission  
60 control measure has successfully reduced some air pollutants, including SO<sub>2</sub> and NO<sub>x</sub> by 59%  
61 and 21%, respectively, during 2013–2017. However, VOCs and NH<sub>3</sub> emissions were not  
62 expected to be reduced, and therefore remained stable for NH<sub>3</sub> and even increased for the  
63 VOCs during 2010–2017 (Zheng et al., 2018). Although the anthropogenic source emissions  
64 during recent years have been characterized in China, understanding of temporal trends of  
65 surface concentrations of those air pollutants and their geographic source distributions

66 remains limited. Investigation of long-term trends of those pollutants by ground-based  
67 measurements can help to further understand effectiveness and/or feedbacks of the air-  
68 pollution control progress on the improvement of air quality.

69         As one of the central cities in one of the core economic zones in eastern China,  
70 Kunshan is located in the region closed (around 100 km) to the megacity of Shanghai in the  
71 Yangtze River Delta region. Regarding to economic growth, Kunshan, with total area of  
72 approximately 900 square kilometers and around 1 million population, has ranked first  
73 among the 100 counties and cities for 13 years in China (as reported by Kunshan local  
74 chronicles Compilation Committee Office). By this rapid economic growth and the associated  
75 intensified urbanization process, the air pollution and other ecological and environmental  
76 problems have been increasingly prominent in the Kunshan region, and it has become one of  
77 four severe haze pollution zones in China. This fact poses a serious threat to urban  
78 development and the mutual restraint between economy and environment, causing wide  
79 social concern. In the present work, we quantified the temporal trends of air pollutants using  
80 Mann-Kendall test analysis, including PM<sub>2.5</sub>, the PM<sub>2.5</sub> to CO ratio, SO<sub>2</sub>, NO<sub>2</sub>, and O<sub>3</sub>, based on  
81 long-term ground-based measurements during 2012 – 2018 under different environments  
82 (including urban and near rural areas). Possible influence of regional and local pollution  
83 sources on the trends of those air pollutants was discussed. The potential influence of PM<sub>2.5</sub>  
84 changes on O<sub>3</sub> trends was explored. Geographic sources of those air pollutants were  
85 investigated using a potential source contribution function method.

86

## 87 2. Data and methods

### 88 2.1 Air pollutants dataset

89 Hourly data of air pollutants, including PM<sub>2.5</sub>, O<sub>3</sub>, NO<sub>2</sub>, SO<sub>2</sub>, and CO, were obtained from the  
90 online air quality monitoring network of Kunshan. There are 12 stations involved in this  
91 database, including 2 urban, 5 near rural, 3 town and 2 industrial sites, which has been  
92 summarized in Table S1. A meteorological parameter (i.e., wind speed) was obtained from a  
93 meteorological observation station in Kunshan.

94

### 95 2.2 Trend analysis

96 The Mann-Kendall statistical test (Mann, 1945), a non-parametric test, has been  
97 commonly used in time series analysis to identify the trends of observational datasets. In the  
98 Mann-Kendall test, the data don't have to meet the normality distribution, because non-  
99 parametric test works for all data distributions. This is a major advantage compared to  
100 parametric test, such as t test. The seasonal Mann-Kendall test is aiming to test the trend of  
101 the data with significant seasonality. In the present work, the trend analysis was performed  
102 on monthly mean data sets calculated from all, urban, and near rural sites, during the two  
103 periods, i.e., 2012 – 2018 and 2014 -2018, respectively. Before performing the Mann-Kendall  
104 test, the seasonality of the monthly data was firstly examined using the Kruskal-Wallis test  
105 (Kruskal and Wallis, 1952). If seasonality of the given data is significant (Kruskal-Wallis's  $p <$   
106  $0.05$ ), the seasonal Mann-Kendall test is applied, while the Mann-Kendall test will be applied  
107 for the data without significant seasonality (Kruskal-Wallis's  $p > 0.05$ ). The median slope of  
108 the trend was calculated using Sen's slope method (Sen, 1968). The magnitude of the trends  
109 was presented only for the data with statistically significant trend at 5% significance level  
110 (see section 3.1). To examine the response of the China's rigorous air-pollution control (an Air  
111 Pollution Action Plan released in September 2013) to the improvement of air quality over a  
112 specific region, we compared these trends during the two periods of 2012-2018 and 2014-  
113 2018. The Mann-Kendal trend analysis was performed using a R trend package (Pohlert,  
114 2018). Finally, the least-squares fitting method was also applied here to compare the Mann-  
115 Kendall statistical trend analysis results (see Tables S2-S4).

116

### 117 **2.3 Potential geographic origins analysis**

118 Potential source contribution function (PSCF) (Polissar, 1999) – a statistical approach  
119 – has been widely used to determine possible geographic origins of observed air pollutants.  
120 The PSCF describes the relationship between the coordinates of air mass back-trajectories  
121 and the ambient concentrations of air pollutants (Fleming et al., 2012). The magnitude of  
122 PSCF is defined as the grid-based probability linked to the pollutant origins, which is  
123 calculated by the ratio of  $m_{ij}$  to  $n_{ij}$ .  $m_{ij}$  represents the number of trajectory endpoints in a grid  
124 cell  $(i,j)$  associated with a given pollutant threshold value, and  $n_{ij}$  is the total count of  
125 trajectory endpoints. Therefore, the PSCF analysis allows us to understand the high potential  
126 source region that contributes the high concentration of air pollutants at the receptor site. To  
127 minimize a limitation of PSCF caused by a limited number of  $n_{ij}$  points in a grid cell, a  
128 downweighing function ( $w_{ij}$ ) was performed here, as described in details by previous studies  
129 (Zhang et al., 2019b;Waked et al., 2014). In the present work, the PSCF analysis was  
130 performed based on 72-h back trajectory data that was calculated every 1 h using the  
131 HYSPLIT model (Draxler and Rolph, 2013) at an ending height of 100 m above ground level  
132 (a.g.l.). Global Data Assimilation System (GDAS) files with a  $0.2^\circ \times 0.2^\circ$  horizontal resolution  
133 were used here for the back-trajectory analysis. The receptor site was setting up at the near  
134 center (longitude:  $120^\circ 8'$ , latitude:  $31^\circ 2'$ ) of Kunshan. The average concentrations of the air  
135 pollutants observed from all sampling sites were applied for the PSCF analysis. The threshold  
136 value of the ambient concentrations was set up at 75<sup>th</sup> percentile for the PSCF calculation,  
137 which is consistent with many previous studies (e.g., (Sun et al., 2015;Zhang et al., 2017b).  
138 All PSCF-related analysis was done here using an Igor-Pro user-friendly toolkit (ZeFir) (Petit et  
139 al., 2017).

140

## 141 **3. Results and discussion**

### 142 **3.1 Trends and variations of major air pollutants**

143 In China, the air-pollution control policies have been implemented by the Chinese  
144 government since 2010 (Zheng et al., 2018), while a new cleaner air action – so-called  
145 national “Ten Measures” – has been implemented to further improve the air quality since  
146 end of 2013 (Zhang et al., 2019a). Within such new clean air policies, the national emission  
147 standards have been applied for emission control linking to different source sectors (Zheng et  
148 al., 2018; Zhang et al., 2019a). For instance, the national standards GB 29620-2013, GB 4915-  
149 2013, and GB 13271-2014, were implemented to reduce some industrial sectors, i.e., Brick,  
150 Cement, and industrial boiler, respectively (Zheng et al., 2018). Instead of the Euro 3  
151 standard, the Euro 4 standard was applied for diesel vehicle of transportation sector (Zheng  
152 et al., 2018). As a consequence of the 2013 cleaner air action, better air quality is logically  
153 expected, however ground-based measurements are needed to directly evaluate those  
154 pollution control policies. Figure 1 presents the trends in surface concentrations of PM<sub>2.5</sub>, SO<sub>2</sub>,  
155 NO<sub>2</sub>, and O<sub>3</sub>, as well as the ratio of PM<sub>2.5</sub> to CO measured at all sampling sites. As a result of  
156 Fig. 1a and Fig. S1a, PM<sub>2.5</sub> observed at all sampling sites (including urban, near rural, town,  
157 and industry areas), have an excellent consistency, suggesting the comparable surface mass  
158 concentrations of fine aerosols over the Kunshan region. During the both periods, i.e., 2012-  
159 2018 and 2014-2018, PM<sub>2.5</sub> exhibits statistically significant ( $p < 0.05$ ) decreasing trends within  
160 90 % confidence interval. For the average values calculated from all sites’ observations, it can  
161 be noted that the change ratio ( $-8.2 \text{ \% yr}^{-1}$ ) during 2014 – 2018 is higher than that of  $-7.4 \text{ \%}$   
162  $\text{yr}^{-1}$  during the whole period. Reduction in primary emissions of PM<sub>2.5</sub>, by those mitigation  
163 measures (especially strength industrial emission standards) (Zheng et al., 2018; Zhang et al.,  
164 2019a), can partly explain the reduction trend of primary fractions of PM<sub>2.5</sub> observed in  
165 ambient air (Figure 1a). In fact, the reduction in total PM<sub>2.5</sub> can be overall due to reductions  
166 in both, primary and secondary aerosol fractions. To further explore the trends associated  
167 with secondary production of PM<sub>2.5</sub>, an inert combustion tracer (CO) was applied to  
168 normalize the PM<sub>2.5</sub> mass concentrations (i.e., the PM<sub>2.5</sub>-to-CO ratio) to minimize influence of  
169 primary combustion emissions (e.g., gasoline vehicles, coal combustion and biofuel burning)  
170 and meteorological dilution effect (de Gouw, 2005; Zhang and Cao, 2015; Hu et al., 2013; Sun

171 et al., 2016b). As Fig. 1b shows, the PM<sub>2.5</sub>-to-CO ratios calculated with all sites datasets  
172 present significant decreasing trends for the both periods, which is roughly consistent with  
173 the PM<sub>2.5</sub> concentrations trends (Fig. 1a). These results could confirm that the significant  
174 reduction in PM<sub>2.5</sub> observed at a city level can be partly explained by the reduction of  
175 secondary production in PM<sub>2.5</sub>. Those results reflect a fact of the enhanced role of the more  
176 rigorous air-pollution control in the reduction of fine aerosols in the atmosphere during the  
177 recent past years (Zheng et al., 2018), especially in the Yangtze River Delta region (Zhai et al.,  
178 2019).

179 Resulting in the reduction in secondary PM<sub>2.5</sub> fractions, another evidence of  
180 significant decreasing trends of SO<sub>2</sub> and NO<sub>2</sub> – gas-phase precursors of secondary inorganic  
181 aerosols – have been observed accordingly, as indicated by the results of Figs 1 c and d. It  
182 should be noted that the reduction ratios in the average SO<sub>2</sub> concentration (10.3 and 11.0 %  
183 yr<sup>-1</sup> during 2012-2018 and 2014-2018, respectively) over all sites are approximately two  
184 times higher than those of the NO<sub>2</sub> (-4.4 and -5.8 % yr<sup>-1</sup>). The features of SO<sub>2</sub> and NO<sub>2</sub> trends  
185 observed here are comparable to their emission trends for larger spatial scales on average,  
186 especially over mid-eastern China regions (Zheng et al., 2018;Zhang et al., 2019a). This may  
187 suggest that reduction in ambient SO<sub>2</sub> and NO<sub>2</sub> in a smaller region can be associated with  
188 those emission control policies over larger-regional scales. As reported by Zhang et al.  
189 (2019a), strength industrial emission standards are the largest contributor among some  
190 mitigation measures, including phase out small and polluting factories, phase out outdated  
191 industrial capacities, upgrades on industrial boilers, Promote clean fuels in the residential  
192 sector, and strengthen vehicle emission standards, to the reduction of air pollutants (i.e., SO<sub>2</sub>  
193 and NO<sub>2</sub>, as well as PM<sub>2.5</sub>) by the pollution control measures during the 2013-2017. Thus,  
194 reduction in SO<sub>2</sub> and NO<sub>2</sub> in Kunshan region can be made associated with those pollution  
195 control policies. Figures 2 a1 and b1 describe the relationship between the PM<sub>2.5</sub> to CO ratio  
196 and NO<sub>2</sub> and SO<sub>2</sub> observed in urban and near rural sites, which is characterized by higher  
197 ratio values associated with higher concentrations of both gaseous precursors. This implies  
198 that the high secondary PM<sub>2.5</sub> production in Kunshan could be partly explained by enhanced  
199 secondary inorganic aerosol formation from SO<sub>2</sub> and NO<sub>2</sub> chemistry in both urban and near-  
200 rural environments (Xie et al., 2015;Cheng et al., 2016;Wang et al., 2016a;Sun et al., 2016b).  
201 As shown in Figs. 2 a2 and b2, the ratios of NO<sub>2</sub> to SO<sub>2</sub> have increasing trends at both urban

202 and near-rural sites over recent years, suggesting an implication that NO<sub>2</sub>-related aerosol  
203 chemistry might play a more important role in PM<sub>2.5</sub> pollution in the present and even future  
204 atmosphere. Interestingly, a higher ratio of NO<sub>2</sub> to SO<sub>2</sub> is observed at urban sites compared  
205 to that observed at near-rural sites (Figure 2), which is likely due to larger influence of  
206 transportation sectors on ambient NO<sub>2</sub> concentrations in urban environment. As presented  
207 in Figure 1d, there is no significant trend at 5% significance level for the mass concentration  
208 of NO<sub>2</sub> in urban environments, while significant decreasing trends are observed in near-rural  
209 environments during both periods, 2012-2018 and 2014-2018. This reflects that a more  
210 substantial influence of local emissions (e.g., traffic) on the ambient NO<sub>2</sub> burden in urban  
211 environments rather than in near-rural environments. Moreover, the similar reduction rates  
212 (-7.8 % yr<sup>-1</sup> versus -7.2 yr<sup>-1</sup>) for near-rural sites during the two periods suggest a less effective  
213 emission control strategy by the new mitigation measures for the ambient NO<sub>2</sub> pollution  
214 control. As illustrated by Fig. S1d, the average mass concentration of NO<sub>2</sub> observed at urban  
215 sites is higher than that at near rural sites through the whole year, highlighting larger  
216 emissions of NO<sub>2</sub> in urban environments due to more intensive traffic emissions (i.e.,  
217 transportation sector). Overall, these results above highlight the more effective control in  
218 SO<sub>2</sub> in the ambient air – mainly from industrial combustion sources – rather than NO<sub>2</sub> that is  
219 from transportation sources under the recent emission control policies (Zheng et al., 2018).  
220 Such feedback of the lack effectiveness of reduction in NO<sub>2</sub> to its corresponding secondary  
221 particulate production budget has been also observed by a very recent study, indicating a  
222 less change ratio of nitrate compared to sulfate during recent years in Nanjing (Ding et al.,  
223 2019).

224 For O<sub>3</sub> (see Fig. 1e and Table S2-S4), there is no statistically significant trend ( $p > 0.05$ )  
225 for the period 2012-2018. Interestingly, there is a tendency for increasing O<sub>3</sub> concentrations  
226 (+3.4 % yr<sup>-1</sup>,  $p < 0.05$ ) during 2014-2018. This indicates that the improvement in air quality in  
227 Kunshan is driven by reduction of the fine particles levels, while O<sub>3</sub> pollution becomes a  
228 more serious role in air pollution under the emission control strategies of China's clean air  
229 actions since 2013 (Zheng et al., 2018). O<sub>3</sub> is a secondary pollutant produced from  
230 photochemical oxidation of its precursors, i.e., volatile organic compounds (VOCs) and  
231 nitrogen oxides (NO<sub>x</sub> = NO + NO<sub>2</sub>). Thus, the increasing trend in O<sub>3</sub> could be probably linked  
232 to feedbacks of the current air-pollution emission control, which will be further discussed in

233 section 3.2.

234 Since different sampling sites may present different environment features with, for  
235 instance, different source distributions. More intense local emissions (e.g., traffic and  
236 cooking) occur in urban region rather than in rural area. To further understand influence  
237 urban emissions on air quality, we compared the trends of air pollutants between urban and  
238 rural. Overall, the reduction ratios of PM<sub>2.5</sub> observed at near rural areas are generally higher  
239 than at urban areas for both periods (Fig. 1a). There is no statistically significant trend in the  
240 ratio of PM<sub>2.5</sub> to CO (Fig. 1b) and NO<sub>2</sub>(Fig. 1d) for urban areas during the two periods.  
241 However, significant trends of NO<sub>2</sub> calculated from rural sites are observed during the two  
242 periods, the reduction ratios of which are higher than them for all sites. These results  
243 confirm the strong influence of local combustion source emissions on the trend variation at  
244 urban sites. Therefore, the less change in PM<sub>2.5</sub> in urban region could be linked to the more  
245 important impact of local primary emissions (i.e., traffic source) at urban sites rather than in  
246 the rural areas, which is actually comparable to limited change in PM<sub>2.5</sub> emitted from  
247 transportation sector during recent years (Zheng et al., 2018). The ratio of PM<sub>2.5</sub> to CO has  
248 significant decreasing trends for the both periods (Fig. 1b), which is consistent with the  
249 trends of O<sub>3</sub> with a higher change ratio at rural sites than in urban areas during the period of  
250 2014-2018. This reflects that the ambient PM<sub>2.5</sub> composition in rural areas in Kunshan could  
251 be dominated by regional productions of secondary aerosols that are more sensitive to the  
252 regional-scale emission reduction control. SO<sub>2</sub> also presents decreasing concentrations with  
253 significant trend for both urban and rural areas, along with similar reduction ratios,  
254 suggesting a regional pollution characterization of SO<sub>2</sub> in Kunshan but with the lack influence  
255 of local sources emission.

256 Figure S2 illustrates comparison of diurnal cycles of air pollutants at different wind  
257 speed conditions (i.e., high and low WS, respectively) during the periods representing the  
258 beginning and end of the sampling years (i.e., 2012-2014 and 2016-2018, respectively). The  
259 thresholds to classify the high and low WS conditions are the average values of wind speed  
260 during nighttime in each season. The hourly data of air pollutants observed at all sampling  
261 sites were applied in this analysis. The objective of such data analysis is to roughly explore  
262 influence of regional- and local-dominated sources on air pollutants, which is similar as a  
263 previous study (Chambers et al., 2019). Regional and local pollution sources may be generally

264 associated with the high and low WS conditions, respectively. As presented in Fig. 3, the  
265 mass concentrations of air pollutants (including SO<sub>2</sub>, NO<sub>2</sub> and PM<sub>2.5</sub>) during 2012-2014 at  
266 both high and low WS conditions are higher than those during 2016-2018. Annual mean  
267 concentration of those pollutants at the two WS conditions have decreasing trends in each  
268 season, as illustrated by the results of Figure S3. Those results suggest that reduction in  
269 those pollutants in ambient air can be associated with both, local and regional emission  
270 control measures (Zheng et al., 2018;Zhang et al., 2019a). The diurnal cycles of SO<sub>2</sub> is  
271 characterized by a high peak in early morning in each season during 2012-2014 and 2016-  
272 2018 (Fig. S2a), which could be partly explained by enhanced anthropogenic emissions. NO<sub>2</sub>  
273 exhibits typical diurnal variations with two peaks at rush hours in early morning and evening  
274 (Fig. S2b) due to major influence of local traffic emissions. O<sub>3</sub> shows distinct diurnal cycles  
275 with maximum loadings at around noon in each season (Fig. S2c), because of strong solar  
276 radiation and significant photochemical production of O<sub>3</sub> during daytime.

277 As shown in Fig. S2d, PM<sub>2.5</sub> presents more complex diurnal patterns under different  
278 wind speed conditions in different seasons, since these variations are driven by very different  
279 compositions, sources, and evolution processes of PM<sub>2.5</sub> in different seasons, as well as  
280 meteorological conditions (Sun et al., 2015;Zhang et al., 2015b;Cheng et al., 2019). The  
281 concentration of PM<sub>2.5</sub> at the low WS condition is higher than that at the high WS condition  
282 (Figs. S2d and S3c), which could be explained by local primary emissions and enhanced  
283 secondary aerosol productions, as well as less impact of atmospheric dilution at near-  
284 stagnant meteorological conditions on loadings of atmospheric aerosols. The ratio of PM<sub>2.5</sub> to  
285 CO at the low WS condition is also higher than that at the high WS condition in different  
286 periods and seasons (Fig. S3d), which may further support enhanced secondary fine aerosol  
287 formation at the low WS conditions. This can be also supported by higher SO<sub>2</sub> and NO<sub>2</sub>– gas-  
288 phase precursors of secondary inorganic PM<sub>2.5</sub> that accounts for a large fraction of PM<sub>2.5</sub>  
289 mass in the Yangtze River delta (Huang et al., 2012;Zhang et al., 2015c;Zhang et al.,  
290 2015b;Zhang et al., 2017a) – at the low WS conditions (Fig. S3a and S3b). Those results are  
291 also consistent with previous findings that formation of haze pollution events occurs more  
292 frequently under stagnant meteorological conditions in the eastern China regions (Sun et al.,  
293 2016a;Sun et al., 2014;Ding et al., 2016;Zhang et al., 2017a). As shown in Fig. S3, the mass  
294 concentration of PM<sub>2.5</sub> and the ratio of PM<sub>2.5</sub> to CO display decreasing trends at both low and

295 high WS conditions in each season, suggesting positive influence of regional and local  
296 pollution controls on reduction in ambient PM<sub>2.5</sub> in the Kunshan region. The detailed reasons  
297 leading to these variations can be further investigated with aerosol chemical speciation  
298 measurements and source apportionment, which however were not achieved by the present  
299 study.

300

### 301 **3.3 Relationship between PM<sub>2.5</sub> and O<sub>3</sub> in summer**

302 The increasing trends of O<sub>3</sub> have been discussed in the section above. As shown in Fig.  
303 1e, the significant enhancement of O<sub>3</sub> concentrations are observed during summertime in  
304 each year, indicative of more health risk of O<sub>3</sub> occurring in summer in the recent years. We  
305 therefore further investigated the possible reasons increasing O<sub>3</sub> in summer and whether  
306 this increase is linked to the PM<sub>2.5</sub> concentration variations and its precursors reductions  
307 during the recent years.

308 Figure 3 shows the relation between O<sub>3</sub> and PM<sub>2.5</sub> at different NO<sub>2</sub> concentrations  
309 conditions at daytime in summer. It can be clearly seen that there is a positive relationship  
310 between O<sub>3</sub> against PM<sub>2.5</sub> at lower NO<sub>2</sub> levels (< around 20 – 30 µg m<sup>-3</sup>). This suggests that  
311 the photochemical chemistry can account for increases in the summertime PM<sub>2.5</sub> production  
312 to a certain extent. However, O<sub>3</sub> presents very limited change in concentrations as increase  
313 of PM<sub>2.5</sub> at higher NO<sub>2</sub> levels (> around 30 µg m<sup>-3</sup>) (see Figs. 3 and S4). This describes a  
314 phenomenon of limited formation of O<sub>3</sub> under higher NO<sub>2</sub> conditions, which might suggest  
315 that reduction in NO<sub>x</sub> concentrations could be one of the reasons leading to increase of O<sub>3</sub>  
316 production. In fact, several recent previous studies have also found such a possible reason  
317 based on field measurements (Xu et al., 2019) and modeling simulations (Li et al., 2019).  
318 Furthermore, Xu et al. (2019) suggested that O<sub>3</sub> production was in a VOC-limited regime in  
319 the downtown and sub-urban areas in Shanghai –near our study area. Modeling results  
320 achieved by a chemical transport model (GEOS-Chem) simulation also suggested that O<sub>3</sub>  
321 production in megacity cluster in the Yangtze River Delta was VOC-limited (Li et al., 2019).

322 The presence of aerosol particles can influence photochemical reactions in the  
323 atmosphere (Li et al., 2019), since complex aerosol chemistry could act as a sink of

324 atmospheric oxidants, e.g., OH radical, hydrogen peroxide ( $H_2O_2$ ), and  $O_3$  (Fu et al., 2019). In  
325 the atmosphere, these oxidants can react with aerosol precursors (e.g.,  $SO_2$ ,  $NO_2$  and VOCs)  
326 to form secondary aerosol particles (Zhang et al., 2015a), and thus these oxidants are  
327 depleted via such aerosol formation processes. As Fig. 3 shows, the higher ratio of  $PM_{2.5}$  to  
328 CO – an indicator of secondary aerosol production – is observed corresponding to the high  
329  $PM_{2.5}$  concentrations for the scenario of the  $O_3$  versus  $PM_{2.5}$  under high  $NO_2$  concentrations  
330 levels. This result could theoretically suggest that changes in ambient concentrations of  
331  $PM_{2.5}$  and its precursors have considerable influence on  $O_3$  concentrations levels (Li et al.,  
332 2019;Fu et al., 2019). As indicated by Li et al. (2019), rapid reduction in surface  $PM_{2.5}$   
333 concentrations could result in a slowing down sink of hydroperoxy ( $HO_2$ ) radicals and thereby  
334 increase in  $O_3$  production. Hence, our observational results could be partly explained by such  
335 a mechanism leading to the increasing trend of  $O_3$  observed during the recent years (Li et al.,  
336 2019).

337

### 338 **3.4 Geographic sources**

339 Figure 4 shows the seasonal characterization of the potential source areas for  $PM_{2.5}$ ,  
340 the  $PM_{2.5}$  to CO ratio,  $SO_2$  and  $NO_2$ . As shown in Fig. 4a,  $PM_{2.5}$  shows a similar source region  
341 characterized by a high PSCF band along Hengshui-Xuzhou-Hefei-Nanjing-Shanghai in all  
342 seasons. These results are generally consistent with the high potential source areas of the  
343  $PM_{2.5}$  to CO ratio (Fig. 4b), suggesting that regional transport of secondary aerosol  
344 productions from the northwest may play a dominant role in formation of severe winter haze  
345 pollution in Kunshan. Besides of that, the contribution from local areas near Kunshan to the  
346  $PM_{2.5}$  pollution cannot be ignored in each season (Fig. 4a). Meanwhile, a similar potential  
347 source area for  $SO_2$  is also observed in winter, spring and fall, as indicated by Fig. 4c, which is  
348 characterized by a broader band and even longer regional transport pathways up to Hebei  
349 Province, one of the most polluted regions in China caused by intense emissions of coal  
350 combustion (Wang et al., 2018;Koukouli et al., 2018;Tian et al., 2010). In summer, high PSCF  
351 region of  $SO_2$  is mainly dominated by local scale (Fig. 4c), which is similar as the PSCF  
352 distributions for  $NO_2$  (Fig. 4d), as well as the ratio of  $PM_{2.5}$  to CO with higher PSCF values over  
353 the Yangtze River Delta region (Fig. 4a). These results indicate the importance of local

354 sources emissions for PM<sub>2.5</sub> pollution in summer. Compared to SO<sub>2</sub> in other seasons (i.e.,  
355 winter, spring, and fall), a relative narrow source area located at northwest and southwest  
356 and is observed for NO<sub>2</sub> (Fig. 4d), which is indicative of a regional transport contribution to  
357 NO<sub>2</sub> in addition to local emissions. Those results highlight again the importance of controlling  
358 NO<sub>2</sub> and SO<sub>2</sub> emissions over a region scale in reduction of ambient PM<sub>2.5</sub> burden. According  
359 to Fig. S5, showing the year-round PSCF of PM<sub>2.5</sub>, SO<sub>2</sub>, and NO<sub>2</sub>, as well as the ratio of PM<sub>2.5</sub>  
360 to CO, all air pollutants in each year show similar PSCF spatial distributions, suggesting a  
361 common characteristic of the potential sources in winter every year. Thus, regional transport  
362 from these areas (i.e., northwest) and emissions over local-scale regions play an important  
363 role in the formation of haze pollution in the region near Shanghai during the whole  
364 investigation period 2011 - 2018.

365 Figure 5 presents the spatial distributions of PSCF values for O<sub>3</sub> in all seasons, which  
366 are overall characterized by high potential source region located in southwestern, southern,  
367 and southeastern areas, e.g., Hangzhou-Shanghai. This suggests that air masses origins from  
368 these regions could have potentially important impact on O<sub>3</sub> formation observed in the  
369 Kunshan region. In the warm months, higher concentrations of O<sub>3</sub> are frequently observed in  
370 each year (Fig. 1e). These distinct PSCF regions located in southeastern region could then  
371 explain the possible source origins of O<sub>3</sub> episodes occurred in summer and spring. Moreover,  
372 the PSCF characteristics of O<sub>3</sub> are comparable among each year, as indicated by the  
373 summertime results of Fig. S6. Given that many cities are located in the high potential PSCF  
374 of O<sub>3</sub>, the transport of air masses from these areas would have a potentially high impact on  
375 the formation of O<sub>3</sub> pollution episodes in the region near Shanghai in summer. Therefore, the  
376 combination of emissions control of NO<sub>x</sub> and VOCs in a regional scale, such as over megacity  
377 cluster area, could help to improve the summertime air pollution caused by O<sub>3</sub>.

378

#### 379 **4. Conclusion**

380 We conducted a comprehensive analysis in long-term and multi-site dataset of air pollutants  
381 (including PM<sub>2.5</sub> and O<sub>3</sub>) observed in Kunshan during the period of 2012-2018. The trends of  
382 those air pollutants were calculated using Mann-Kendall statistical test. The monthly PM<sub>2.5</sub>  
383 averaged from all sites data presented statically significant reduction trend (-7.4% per year)

384 during 2012-2018. The ratio of PM<sub>2.5</sub> to CO also had a significant reduction trend for the  
385 averages of all sites, which is consistent with the PM<sub>2.5</sub>, suggesting the possible reduction of  
386 secondary aerosol production during the past several years. This possibility is supported by  
387 the significant reduction trend of aerosol precursors, SO<sub>2</sub> (-10.3% per year) and NO<sub>2</sub> (-4.4%  
388 per year). Such higher change ratio of SO<sub>2</sub> suggested a more effective emission control  
389 strategy for atmospheric SO<sub>2</sub> burden than for NO<sub>2</sub>. However, there was no significant trend  
390 for NO<sub>2</sub> observed at urban sites, emphasizing the importance of local traffic emissions for the  
391 temporal variations of NO<sub>2</sub>. Interestingly, O<sub>3</sub> presented significant increase trend (+3.5% per  
392 year) during the period of 2014-2018. Enhanced formation of O<sub>3</sub> was observed to be  
393 associated with lower PM<sub>2.5</sub> and NO<sub>2</sub> concentrations during daytime in summer, which  
394 implies that the current emission control might provide favorable environment conditions  
395 (e.g., low PM<sub>2.5</sub> loadings) for O<sub>3</sub> formation. The geographic origins of air pollutants were  
396 investigated using PSCF analysis, which presented different source regions between PM<sub>2.5</sub>  
397 and O<sub>3</sub> across different seasons. Higher potential source areas for PM<sub>2.5</sub> were located to the  
398 northwest of Kunshan in all seasons, while the south and southeast were the major  
399 geographic origins of O<sub>3</sub> especially in spring and summer. Overall, these results demonstrated  
400 that improvement of air quality was due to reduction in the concentration level of PM<sub>2.5</sub>,  
401 along corresponding reduction in its precursors of SO<sub>2</sub> and NO<sub>2</sub> during the past years.  
402 Effective and well balanced controls are needed to further reduce both, ambient PM<sub>2.5</sub> and  
403 O<sub>3</sub> pollution, especially over urban areas.

404

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409

410 **Conflicts of Interest**

411 The authors declare no conflict of interest.

412

413 **Figure captions**

414

415 **Figure 1.** Monthly trends in (a) PM<sub>2.5</sub>, (b) PM<sub>2.5</sub>-to-CO ratio, (c) NO<sub>2</sub>, (d) SO<sub>2</sub>, and (e) O<sub>3</sub>. Two  
416 methods (see Tables S2-S4), including Mann-Kendall test and least-squares fitting, have been  
417 applied for the time series statistical analysis aiming to quantify the trends over the two  
418 periods, i.e., A (2012-2018) and B (2014-2018), respectively. Note that the trend was not  
419 further quantified for the data without statistically significant trend according to the Mann-  
420 Kendall test.

421

422 **Figure 2.** Daily ratio between NO<sub>2</sub> and SO<sub>2</sub> as a function of (a) the measuring dates and (b)  
423 the PM<sub>2.5</sub> to CO ratio, at urban and near rural sites, respectively.

424

425 **Figure 3.** Relationship between hourly O<sub>3</sub> and PM<sub>2.5</sub> at different NO<sub>2</sub> concentrations  
426 conditions at daytime (10:00 – 17:00) in summer (June-July-August) during 2012-2018. The  
427 relationship between O<sub>3</sub> and PM<sub>2.5</sub> are averaged (square-shaped points) based on the levels  
428 of NO<sub>2</sub> concentrations, where error bars indicate standard deviations. Hourly average data  
429 points (circle points) are color-scaled by date during the entire period, and size-scaled by the  
430 PM<sub>2.5</sub> to CO ratio.

431

432 **Figure 4.** Seasonal potential source regions of (a) PM<sub>2.5</sub>, (b) the PM<sub>2.5</sub> to CO ratio, (c) SO<sub>2</sub>, and  
433 (d) NO<sub>2</sub>, which were achieved from PSCF analysis during wintertime. The cities marked in  
434 each panel are Beijing (BJ), Shijiazhuang (SJZ), Weifang (WF), Jinan (JN), Hefei(HF),  
435 Xuzhou(XZ), Nanjing(NJ), Shanghai(SH), Hangzhou(HZ), Nanchang(NC), and Wenzhou(WZ),  
436 Fuzhou(FZ). Shijiazhuang (SJZ), and Hengshui (HS). The color scales refer to the PSCF values.  
437 The point in red in each panel represents the location of this study.

438

439 **Figure 5.** Seasonal potential source regions of O<sub>3</sub>: (a) winter; (b) spring; (c) summer; and (d)  
440 fall. The cities marked in each panel are same as Figure 5. The color scales refer to the PSCF

441 values. The point in red in each panel represents the location of this study.

442

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Figure 1.

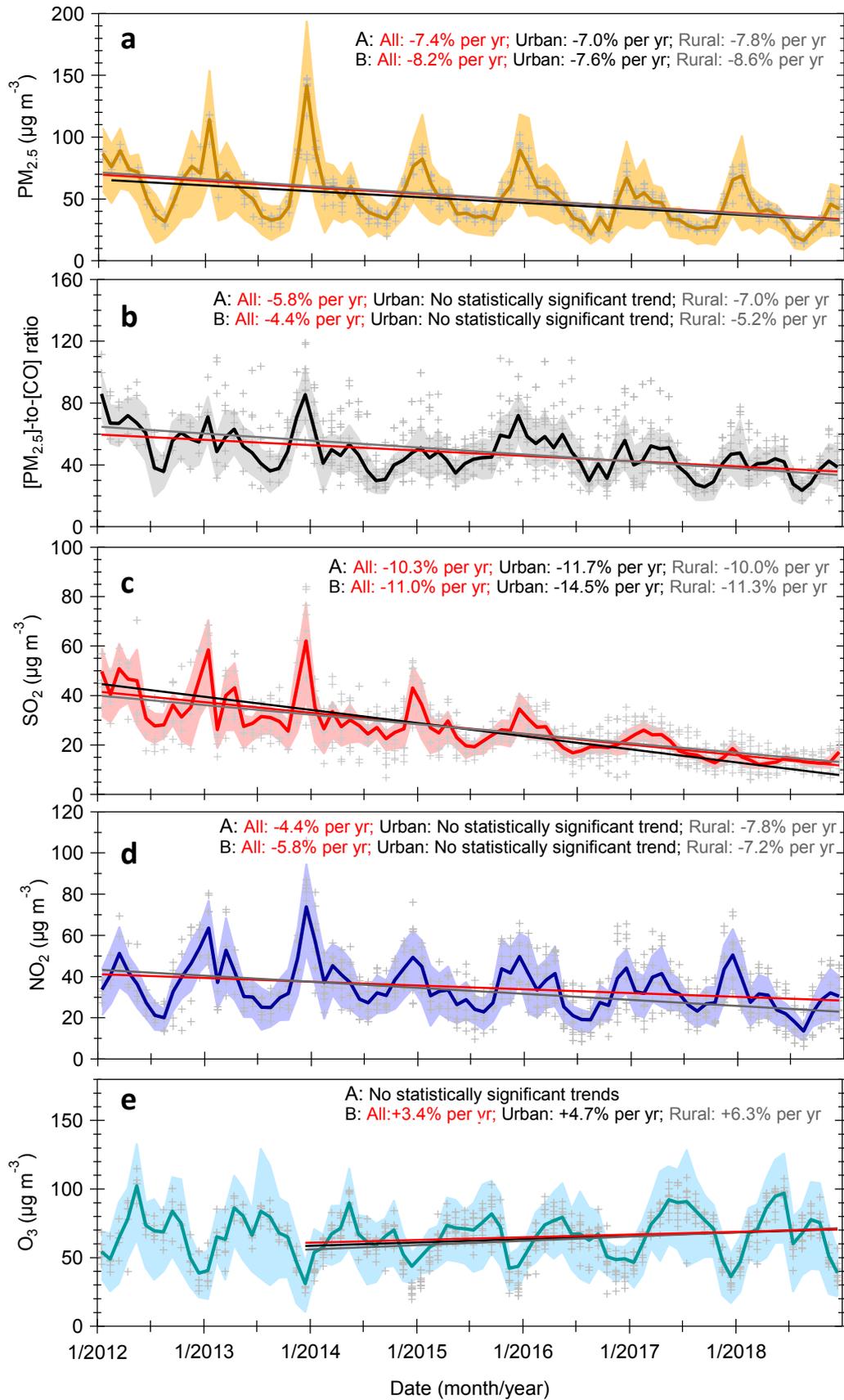


Figure 2.

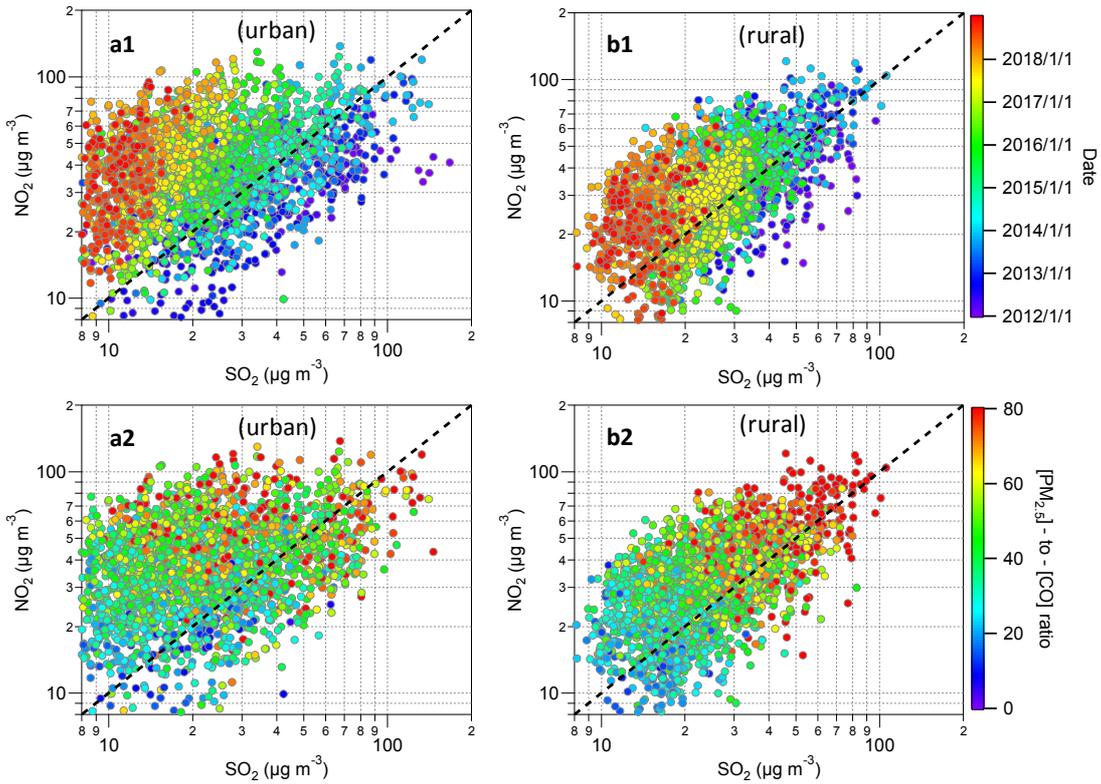


Figure 3.

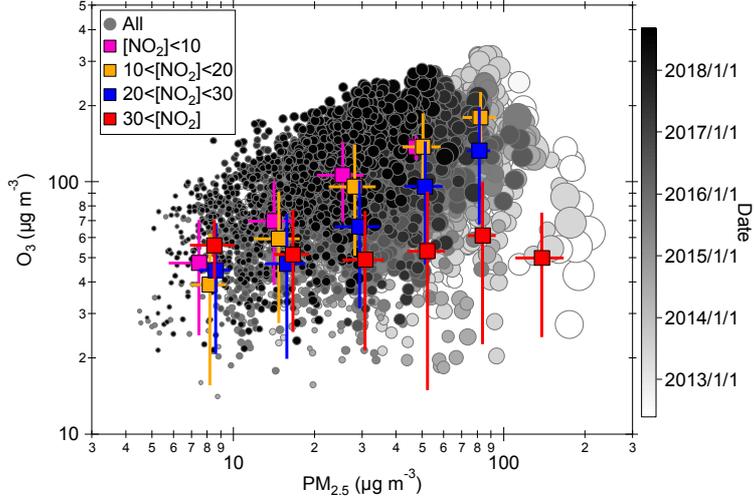


Figure 4.

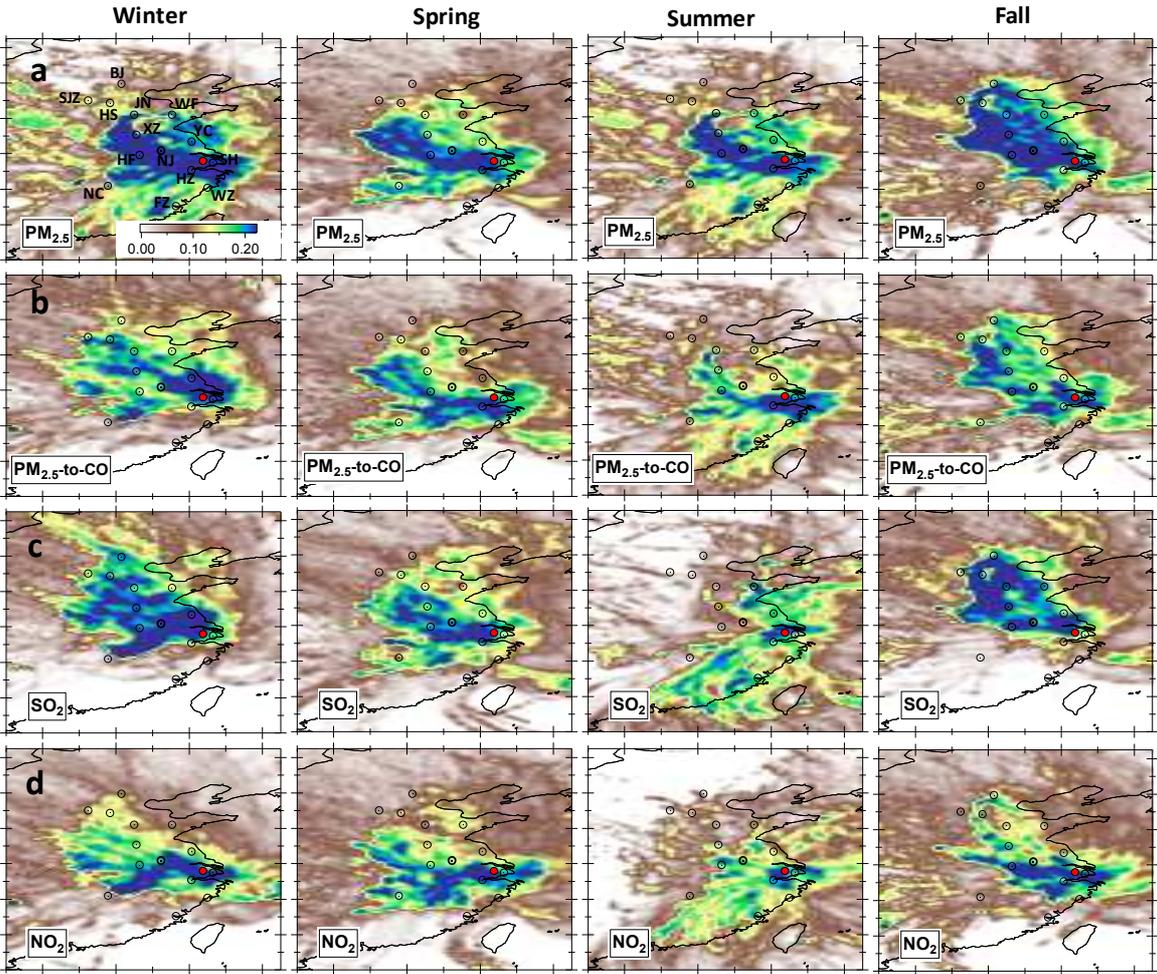


Figure 5.

