

# **Regional transport of PM<sub>2.5</sub> and O<sub>3</sub> based on complex network method and source apportionment technology in the Yangtze River Delta, China**

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## **Key Points:**

- PM<sub>2.5</sub> decreased while O<sub>3</sub> increased in the YRD over the years from 2015 to 2018.
- The complex network method was applied to investigate regional transport of PM<sub>2.5</sub> and O<sub>3</sub> between grid cells.
- Regional transport played significant roles in PM<sub>2.5</sub> and O<sub>3</sub> pollution in the YRD.

## **Abstract**

Ground-level ozone(O<sub>3</sub>) and atmospheric fine particulate matter (PM<sub>2.5</sub>) pollution are the major challenges for continually improving air quality in the Yangtze River Delta (YRD) region of China (Lu et al., 2020; Wei et al., 2020). Understanding regional transport pattern of PM<sub>2.5</sub> and O<sub>3</sub> pollution is essential for the development of regional cooperative prevention and control strategies. This study shows annual concentration of PM<sub>2.5</sub> in the YRD decreased by 18.5% from 2015 to 2018, while average of daily maximum 8-h average(MDA8) O<sub>3</sub> concentration during March to October increased by 16.3%. A complex network method is developed to investigate regional transport of PM<sub>2.5</sub> and O<sub>3</sub> in different grid cells (nodes). Source apportionment technology in regional transport model is applied for comparing with the results from the complex network method. Interregional and intraregional transportation play an important role in

both  $\text{PM}_{2.5}$  and  $\text{O}_3$  over the YRD. The northern part of the YRD contributes much more than other areas for  $\text{PM}_{2.5}$  in winter, which accounts for about 60%, while the central part of the YRD, especially the northern part of Zhejiang, is the largest contributor of  $\text{O}_3$  in the YRD in summer, which accounts for about 70%. Intraregional transport played major role in  $\text{PM}_{2.5}$  than  $\text{O}_3$ . The study focusing on heavily pollution episodes not only show results between source apportionment technology and complex network method are comparable, and also reveals both two methods pose great potential in understanding transport pattern and air pollution relationship, which is the foundation for emission mitigation in the YRD region.

## 1 Introduction

Ground-level ozone ( $\text{O}_3$ ) and atmospheric fine particulate matter ( $\text{PM}_{2.5}$ ) are the major air pollutants affecting human health, climate and ecosystems (Ainsworth et al., 2012; Daellenbach et al., 2020; Hollaway et al., 2012; J. Fuhrer 1997; Lefohn et al., 2017; Yue et al., 2017).  $\text{O}_3$  is a secondary air pollutant formed nonlinearly by photochemical reactions of volatile organic compounds and nitrogen oxides (Atkinson 2000; CRUTZEN 1973; Kleinman et al., 2002; Sillman 1999; Thompson 2016). Some studies have shown that even low levels of  $\text{O}_3$  are associated with increased risk of premature mortality (Anenberg et al., 2010; Bell et al., 2006; Logan et al., 1981; Wu and Xie 2017).  $\text{PM}_{2.5}$  is considered one of the five greatest health risks worldwide, alongside high blood pressure, smoking, diabetes and obesity (Chan and Yao 2008; Chen et al., 2020; Cohen et al., 2017; Forouzanfar et al., 2015; Horowitz 2006; Mauzerall and Wang 2001).

$\text{PM}_{2.5}$  and  $\text{O}_3$  pollution in China have been received much attention (Ding et al., 2013; Ding et al., 2017; Fu et al., 2010; Nerem 2013; Wang et al., 2017a; Wang et al., 2017b; X. Xu 2008; Xu et al., 2008; Zhou et al., 2016), especially in megacities. The China government has taken a lot of actions to mitigate pollution, including implementation of National Air Quality Standard starting in 2012 and stringent clean air actions starting in 2013 (CSC 2013). As a result of the governmental Clean Air Action, air quality in major regions such as Jing-Jin-Ji, Yangtze River Delta (YRD) and Pearl River Delta (PRD) have been improved significantly.  $\text{PM}_{2.5}$  decreased by 30-40% across China during 2013-2017 (Dang and Liao 2019; Ma et al., 2019; Vu et al., 2019). However, annual concentration of  $\text{PM}_{2.5}$  in more than half of major cities in China are still much higher than NAAQS Grade 2 annual standard of  $35 \mu\text{g}/\text{m}^3$ . In addition,  $\text{O}_3$

pollution worsened over the same period, regional  $\text{PM}_{2.5}$  and  $\text{O}_3$  severe episodes still occur frequently (An et al., 2015; Dang and Liao 2019; Li et al., 2020; Li et al., 2019a; Liu et al., 2019; Lu et al., 2020; Ma et al., 2019; Wei et al., 2020).

YRD is not only one of the most developed metropolitan economical regions in China, and also a substantial emission per unit area region. Flat geographic feature and subtropical monsoon climate make local transport of pollutants between cities as a predominant factor affecting air quality (Gao et al., 2016; Tang et al., 2013). Gao et al.(2016) reported that YRD was mainly affected by upwind source regions with high concentration of  $\text{O}_3$  and its precursors. Some researches revealed that both interregional and intraregional transport play a significant role in regional  $\text{PM}_{2.5}$  pollution in YRD (Li et al., 2015; Shu et al., 2019). The Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) and chemical transport models have been extensively applied to investigate regional transport patterns and source distribution (Liu et al., 2018; Stein et al., 2015; Sun et al., 2017; Yang et al., 2014; Zhao et al., 2020). Source apportionment model was used to reveal interaction among sources and regions but this approach is limited by the number of source and regions due to expensive calculation (Shu et al., 2020; Wu et al., 2017a). Hence, interaction among grid cells is rarely performed.

In this study, we developed a complex network method to investigate regional transport of  $\text{PM}_{2.5}$  and  $\text{O}_3$  with respect to grid cells (nodes). A more detailed description of a node is given in section 2.3. Key findings were presented in Section 3. Section 4 provided concluding remarks as well as future directions.

## **2 Materials and Methods**

### **2.1 Observational data of $\text{PM}_{2.5}$ and $\text{O}_3$**

The observational data come from the ambient air monitoring stations of the national environmental monitoring network, which updates and releases hourly concentrations of  $\text{PM}_{2.5}$  and  $\text{O}_3$ . These ambient air monitoring stations are established by the Environmental Monitoring of China and they station across all provinces in mainland China, which indicates the quality of the data used in this paper is authoritative and guaranteed by the government.

## 2.2 The HYSPLIT model

The HYSPLIT model (<https://www.arl.noaa.gov/hysplit/hysplit/>) computes simple air parcel trajectories, as well as complex transport, dispersion, chemical transformation, and deposition simulations. HYSPLIT is used for back trajectory analysis to determine the origin of air masses and establish source-receptor relationship. HYSPLIT has also been used in various studies such as tracking and forecasting the release of radioactive material, wildfire smoke, windblown dust, pollutants from various stationary and mobile emission sources, allergens and volcanic ash (Fu et al., 2010; Liao et al., 2017; Nie et al., 2013; Sun et al., 2017; Tang et al., 2016; Zhou et al., 2016).

HYSPLIT adopted a hybrid approach based on the Lagrangian approach, using a moving frame of reference for the advection and diffusion calculations as the trajectories or air parcels move from their initial location, and the Eulerian methodology, which uses a fixed three-dimensional grid as a frame of reference to compute pollutant air concentrations. In this study, daily 24 h and 48 h forward and backward trajectories at an altitude of 100 m above ground of all grids in the YRD region and its surrounding areas were computed using HYSPLIT based on the Global Data Assimilation System (GDAS) data.

## 2.3 Source apportionment by NAQPMS

The Nested Air Quality Predicting Modeling System (NAQPMS) is a three-dimensional Eulerian chemical transport model. NAQPMS is able to quantify contribution of emissions from different regions to the cities or regions of interest (Wu et al., 2017b). It uses “region-tagged” method. Emissions from different regions are tagged with specific names (tracers) and then tracked through the transport, chemical transformation, and deposition processes in model simulations.

In this study, we setup 3 domains with horizontal grid resolutions of 27, 9 and 3 km, respectively. As shown in Figure 1a, the first domain covers the whole China, the second domain covers the YRD and surrounding regions, and the third domain covers Shanghai and vicinities. The YRD and its surrounding area in D2 (Figure 1b) are divided into 10 regions with a unique being assigned to each region.

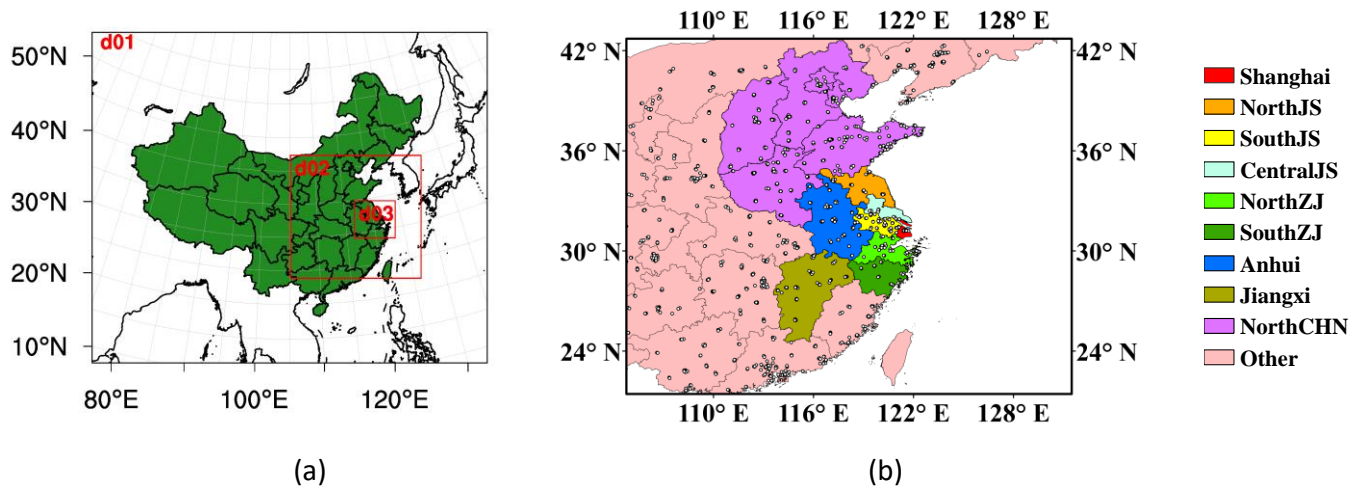


Figure 1. (a) The 27-km (D1), 9-km (D2) and 3-km(D3) domains in the NAQPM simulation. (b) Subdivided source regions applied in D2 domain. The grey circle symbols represent air quality stations (NorthJS:Xuzhou-Lianyungang-Suqian-Huai'an-Yancheng; SouthJS: Nanjing-Zhenjiang-Changzhou-Wuxi-Suzhou; CentralJS: Yangzhou-Taizhou-Nantong; NorthZJ: Hangzhou-Huzhou-Jiaxing-Shaoxing-Ningbo; SouthZJ: Quzhou-Jinhua-Taizhou-Lishui-Wenzhou; NorthCHN: Beijing-Tianjin-Hebei-Henan -Shandong; Other: the remaining areas of the outer domain excluding 9 regions).

### 2.3 Transport analysis by complex network methodology

Network representation of the air pollution transport in a specific region makes it possible to identify important nodes by one of the ranking algorithms. The two founders of Google, Larry Page and Sergey Brin invented an internet search algorithm called PageRank (Brin and Page 2012) to rank the important pages on the internet. Google's success story started from this ranking algorithm. PageRank algorithm has no weight in this ranking analyzing, since page A having a link or 100 links to Page B has the same meaning on the internet. But when we talk about the air masses within a specific area, for instance, node A having one trajectory of air mass or 100 trajectories of air masses to node B, the weight of air masses does matter.

Detailed description of PageRanking algorithm can be found in these studies (Duhan et al., 2009; Franceschet 2011; Medo 2013; Page et al., 1999; Tacchella et al., 2012). Complex network analysis composes of two major steps. The first step is to establish a network to represent research objects. The second step is to find an optimal algorithm to analyze the

constructed network. In this study, PageRank algorithm based on air mass trajectories was applied to create a complex network. The YRD region and its surrounding areas as shown in Figure 1b were subdivided into 1965 grids with respect to the D1 domain grid setting in NAQPM. The trajectories information of each grid were used as the initial matrix. Moreover,  $PM_{2.5}$  and  $O_3$  concentration gradient between node A and node B could impact transport between these two nodes. As a result, complex network method was adjusted by coupling air-mass-weight (AM-Weight) and pollution-transport-weight (PT-Weight) to enhance pollution output importance score (POIS) and pollution input importance score (PIIS) calculations. Ultimately this method identifies key nodes in the YRD region.

### 3 Results and discussion

#### 3.1 Characteristics of $PM_{2.5}$ and $O_3$ in YRD

Figure 2 depicts spatial distribution of  $PM_{2.5}$  and daily maximum 8-h average (MDA8)  $O_3$  concentration in YRD for 2018: annual average and seasonal average. The annual spatial distribution of  $PM_{2.5}$  and MDA8  $O_3$  concentrations illustrated a very similar pattern, with higher concentration in the northern part and lower in the southern part of the YRD region. Higher concentration in the northern part of YRD could be due to local emission and inflow of pollutant from the highly polluted northern part of China such as Shandong, Henan and Hebei provinces. The  $PM_{2.5}$  concentration in the central, southern and coastal areas, including Lianyungang, Yancheng, Nantong, Shanghai, Suzhou, is lower than the northern part of YRD. In contrast, the  $O_3$  concentration in the coastal of northern and central cities, such as Lianyungang, Yancheng, Nantong, Wuxi, Changzhou, Shanghai, is comparable to those in the northern part of YRD.

The seasonal spatial distribution show  $PM_{2.5}$  concentration in winter is the highest in four seasons. This is mainly because the YRD region is heavily affected by the northerly and westerly wind.  $PM_{2.5}$  concentration is also much higher in spring and fall than summer, especially in the northern cities in YRD due to high relative humidity, cooler air mass and low wind speed.  $PM_{2.5}$  concentration in summer is much lower, because of the summer monsoon, a typical maritime inflow, which dissipates pollutants concentration (Tang et al., 2013).

On the  $O_3$  side, Figure 2b showed that the  $O_3$  was much higher in spring and summer. This is consistent with the fact that meteorology is the driving force in these season contributing

166 O<sub>3</sub> production. In autumn and winter, the coastal areas O<sub>3</sub> level is slightly higher than the other  
167 areas. Winter time O<sub>3</sub> production drops significantly, elevated level of O<sub>3</sub> in the coastal areas is  
168 likely due to incoming O<sub>3</sub> from the ocean by onshore breeze.

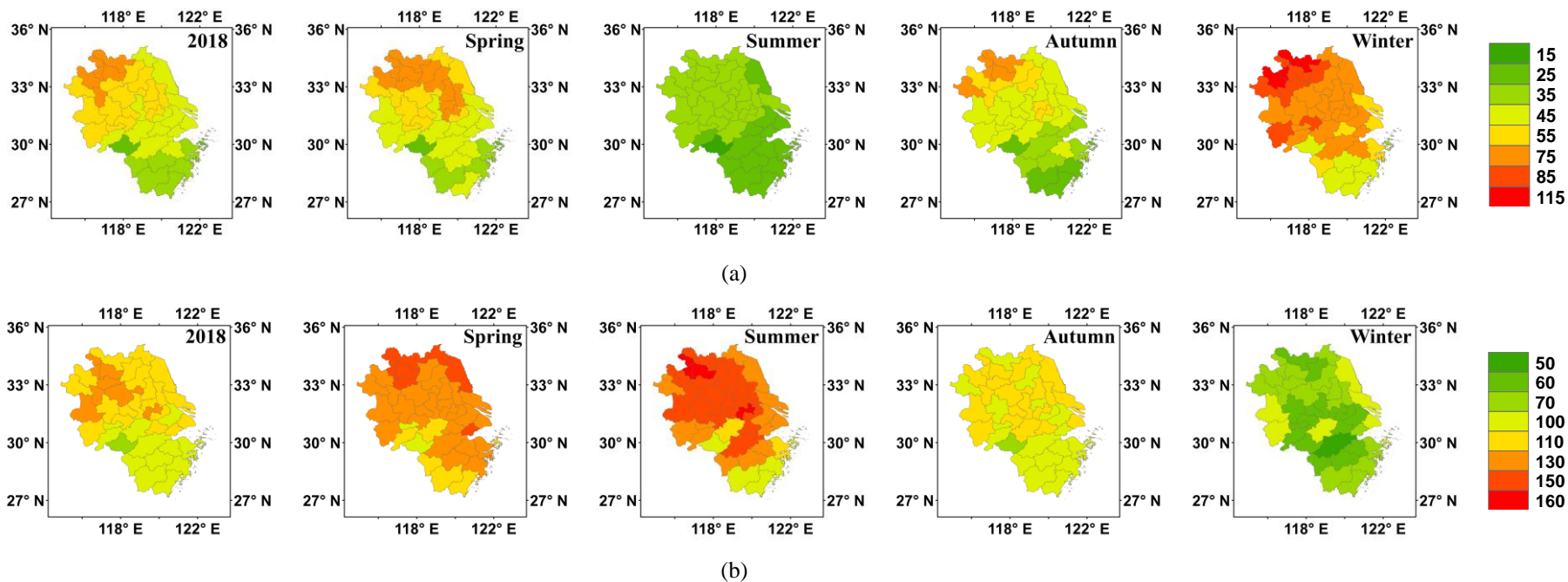
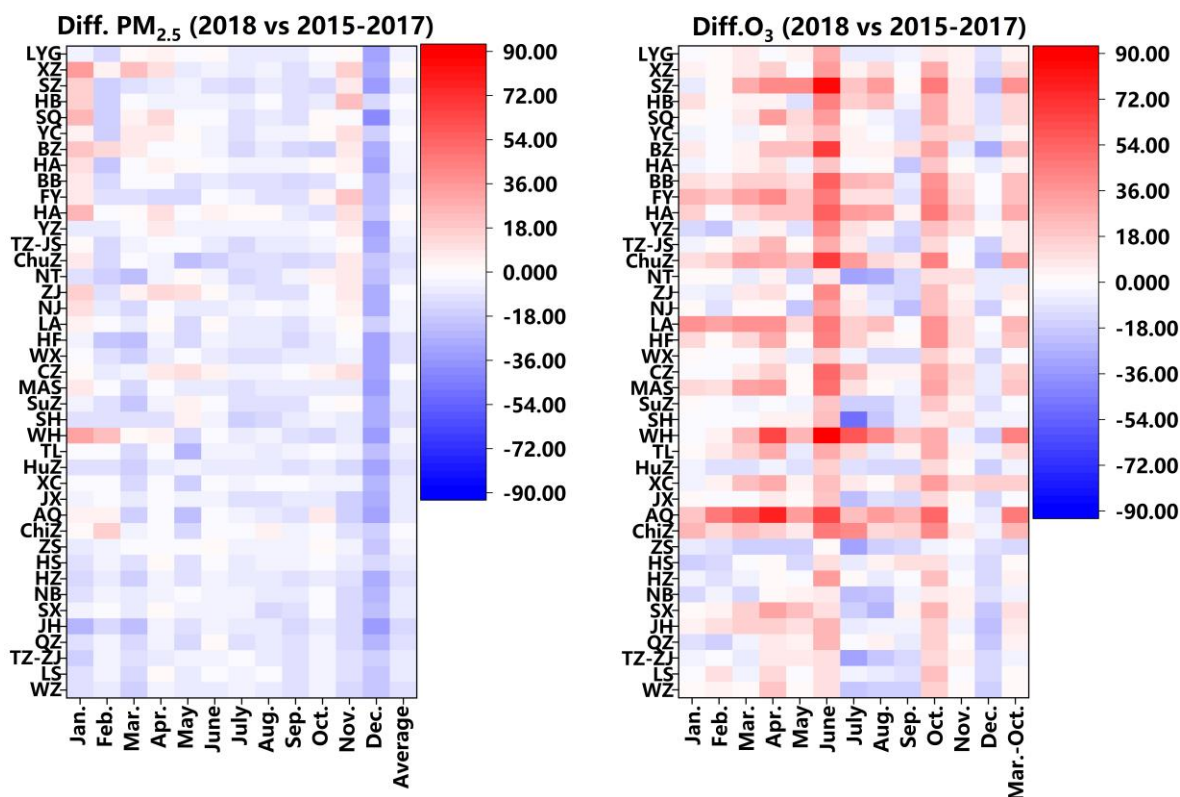


Figure 2. Spatial distribution of  $PM_{2.5}$  and MDA8  $O_3$  concentration in the YRD during 2018 and four seasons.(a: $PM_{2.5}$ ;b: $O_3$ ).Spring includes March, April and May.Summer includes June, July and August.Autumn includes September,October and November.Winter includes December,January and Feburary.



Figure 3. shows the differences in monthly average of  $\text{PM}_{2.5}$  and MDA8  $\text{O}_3$  between 2015-2017 and 2018 over the YRD region. The list of the cities in the figures from top to down is arranged with respect to city location in latitude. It's worth noting that although  $\text{PM}_{2.5}$  has shown a decrease during most of the months, it also shows an increase in the northern cities in January, especially in 2017 and 2018 (Figure S1 and S2). Annual concentration difference of  $\text{PM}_{2.5}$  between 2018 and the average of 2015 to 2017 in Jinhua shows the largest decrease, which is 13.9%, while the concentration difference in Xuzhou shows the largest increase, which is 1.7%. Annual concentration of  $\text{PM}_{2.5}$  in the YRD decreased by 18.5% with a rate of  $3.3 \mu\text{g}/\text{m}^3$  per year from 2015 to 2018, while average concentration of daily maximum 8-h average (MDA8)  $\text{O}_3$  during March to October increased by 16.3% with a rate of  $5.7 \mu\text{g}/\text{m}^3$  per year.  $\text{PM}_{2.5}$  pollution episodes still occur frequently under adverse meteorological conditions with dominant emission sources in the YRD. It is noted that concentration of  $\text{O}_3$  increased significantly in many cities in 2018, however  $\text{PM}_{2.5}$  has shown a decrease. This is probably related to emission mitigation strategy which focused on  $\text{NO}_x$ ,  $\text{SO}_2$  and primary particulate matter reduction, started in 2013. Research has shown anthropogenic  $\text{NO}_x$  emission in China decreased by 20% during 2013 to 2017, while VOC emissions increased by 2%,  $\text{O}_3$  increase could be driven by the  $\text{PM}_{2.5}$  decrease, due to the role of  $\text{PM}_{2.5}$  as scavenger of hydroperoxy ( $\text{HO}_2$ ) radicals that would otherwise react with nitric oxide ( $\text{NO}$ ) to produce ozone (Ma et al., 2019). From Figure 3, the MDA8  $\text{O}_3$  concentration in 2018 was much higher in the month of March to June, especially in June, with the difference ranging from 20-90  $\mu\text{g}/\text{m}^3$ . In summer, the difference was much smaller, and MDA8  $\text{O}_3$  concentrations in several cities over the southern part of YRD were even lower than in 2015-2017.

Substantial rainfall in 2018 summer lowered  $\text{O}_3$  and  $\text{PM}_{2.5}$  concentration in the YRD, particularly in four different cities: Hefei, Nanjing, Shanghai, and Hangzhou (Figure S3 – S6). Figure S3 to S6, showed fairly strong correlation between  $\text{PM}_{2.5}$  and  $\text{O}_3$  in spring and summer.



(a)

(b)

Figure 3. Monthly concentration difference of  $PM_{2.5}$  and  $O_3$  between 2018 and the average of 2015 to 2017 of prefectural cities in YRD (a:  $PM_{2.5}$ ; b:  $O_3$ ; LYG: Lianyungang; XZ: Xuzhou; SZ: Suzhou in Jiangsu; HB: Huaibei; SQ: Suqian; YC: Yancheng; BZ: Bozhou; HA: Huaian; BB: Bengbu; FY: Fuyang; HA: Huaian; YZ: Yangzhou; TZ-JS: Taizhou in Jiangsu; ChuZ: Chuzhou in Anhui; NT: Nantong; ZJ: Zhenjiang; NJ: Nanjing; LA: Luan in Anhui; HF: Hefei; WX: Wuxi; CZ: Changzhou; MAS: Maanan; SuZ: Suzhou in Jiangsu; SH: Shanghai; WH: Wuhu; TL: Tongling; HuZ: Huzhou in Zhejiang; XC: Xuancheng; JX: Jiaying; AQ: Anqing; ChiZ: Chizhou in Anhui; ZS: Zhoushan; HS: Huangshan; HZ: Hangzhou; NB: Ningbo; SX: Shaoxing; JH: Jinhua; QZ: Quzhou; TZ-ZJ: Taizhou in Zhejiang; LS: Lishui; WZ: Wenzhou)

### 3.2 Regional $O_3$ and $PM_{2.5}$ transport pattern in YRD

2018 is the first year after implementing the stringent clean air act based on emission mitigation strategy established in 2013. The Chinese government, policymakers and the public would be interested to know the impact of local emission reduction and regional cooperation on

air quality . Understand interaction among regions in terms of  $PM_{2.5}$  and  $O_3$  transport is the key. Li et al. revealed that contribution of transport to  $PM_{2.5}$  level in typical regions in China such as Northern Plain, Yangtze River delta, Pearl River Delta, and Chengyu area (Li et al., 2019b). Other researches examined interregional transport of  $PM_{2.5}$  and  $O_3$  among main cities in YRD (Li et al., 2015; Shu et al., 2020). In this study, intraregional and interregional transport of  $PM_{2.5}$  and  $O_3$  among in those 1965 nodes (Figure 1) is examined by the complex network algorithm during the pollution seasons in 2018.

Session 3.1 reported  $PM_{2.5}$  concentration in winter and  $O_3$  concentration in summer were the highest in four seasons. The POIS and PIIS of  $PM_{2.5}$  in winter and  $O_3$  in summer were computed for the YRD region. Figure 4 shows POIS values of  $PM_{2.5}$  in the northern areas bordering Henan and Shandong and they are much higher than others in winter. This means these areas are main  $PM_{2.5}$  pollution contributors in winter. In addition, the POIS of the cities on the boader of Anhui and Jiangsu is also high. It is consistant with other studies (Chen et al., 2020; Shu et al., 2019; Sun et al., 2020). Combination of present of large amount of  $PM_{2.5}$  precursors, flat terrain and flavoble weather condition in winter,  $PM_{2.5}$  concentration in the northern of YRD is much higher than that in the southern area, and became a contributing source to other areas through transport. The PIIS of  $PM_{2.5}$  in the southwestern areas of Anhui (Huangshan, Anqing and south Chinzhou) and Zhejiang (Lishui and Wenzhou) are higher than other areas in YRD. In other words, these areas are likely influenced by regional transport from other areas.

Figure 4 also shows the POIS and PIIS of  $O_3$  in the YRD region in summer. Unlike  $PM_{2.5}$ , the POIS of  $O_3$  in central areas of YRD is higher than other areas, especially in these cities: Yangzhou, Zhenjiang, Changzhou, Wuxi, Suzhou, Huzhou, Jiaxing. This is mainly because of relatively high  $O_3$  precursors in these areas (Li et al., 2017). The patthern of PIIS for  $O_3$  is quite similar to  $PM_{2.5}$ . The PIIS of  $O_3$  in southwestern areas including Lishui and Huangshan is higher than other areas. It's mainly because of the downwind and relatively low concentration of  $O_3$  in these areas. In addition, the are that borders Hangzhou, Shaoxing and Jinhua is also high, due to close vicinity of the Hangzhou Bay, an area with various and a main source of VOCs and  $NO_x$  emission

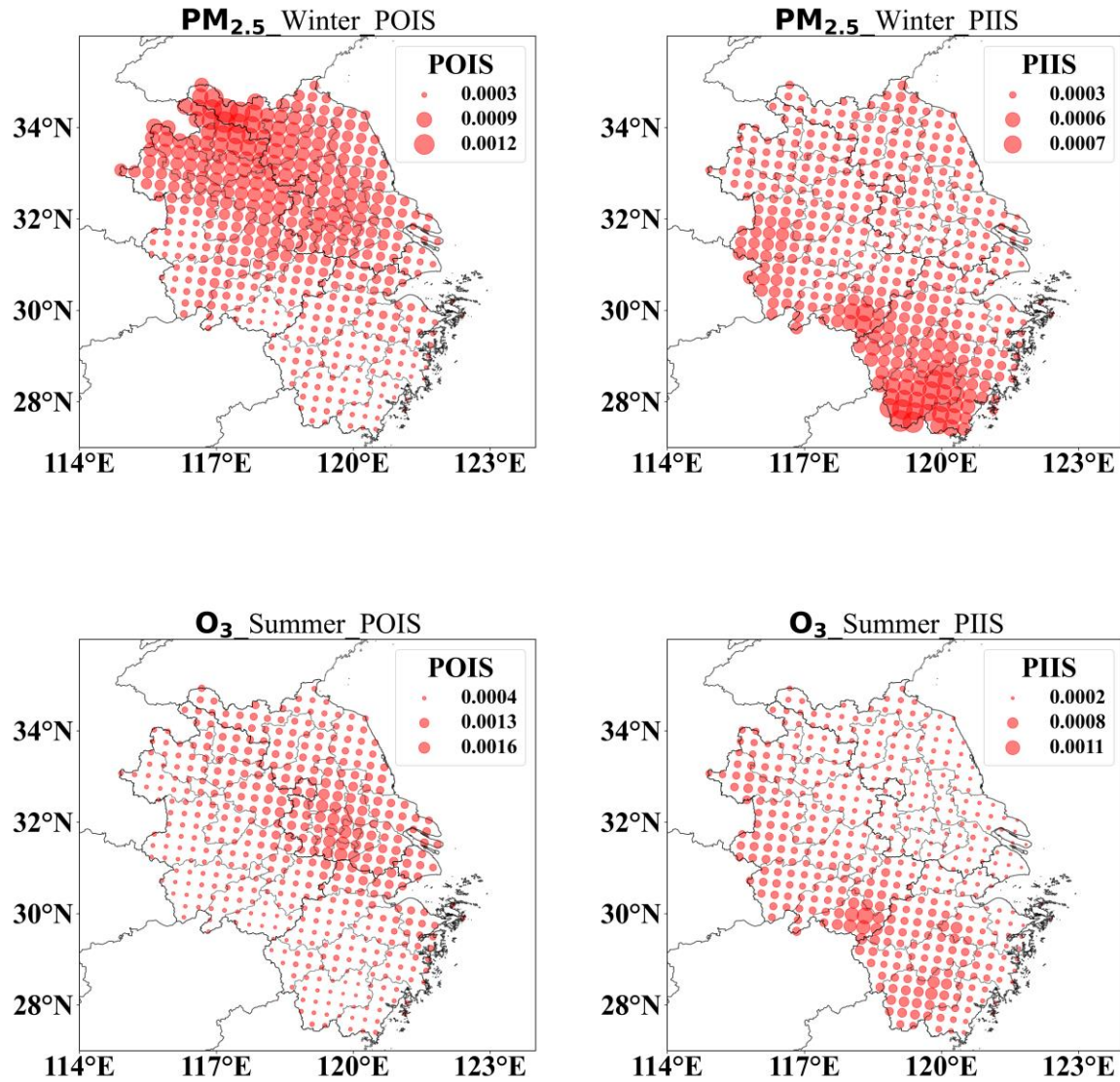


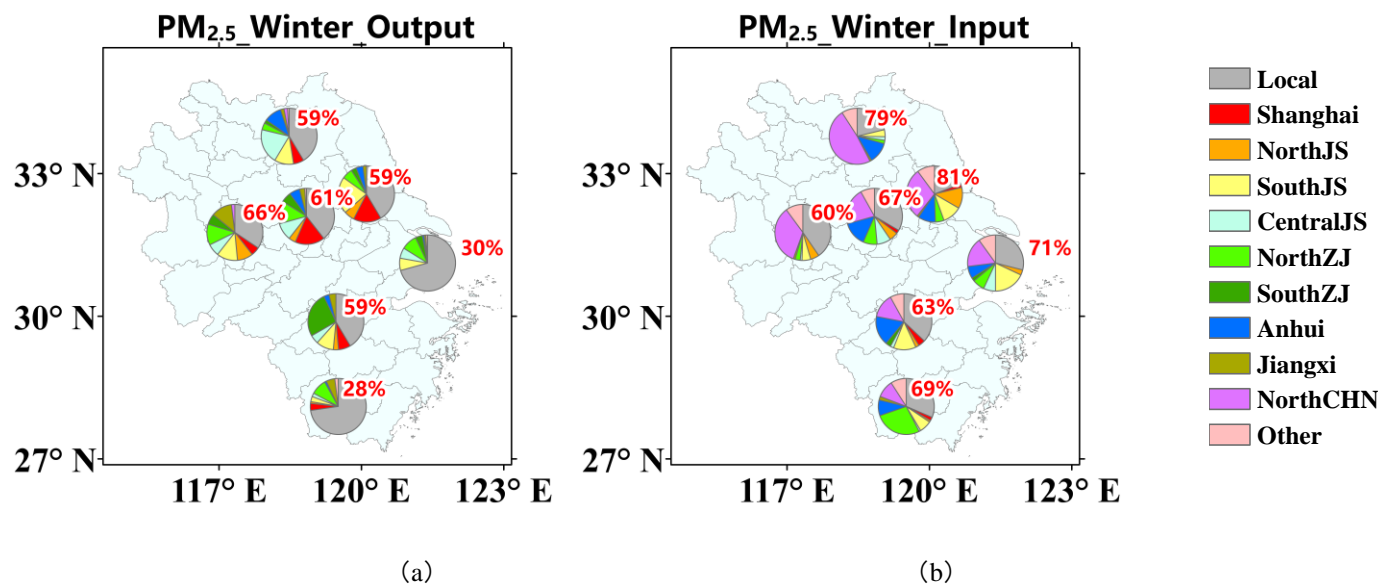
Figure 4. The POIS and PIIS of  $PM_{2.5}$  and  $O_3$  for nodes in YRD during 2018.

Figure 5 shows the source apportionment results for  $PM_{2.5}$  in winter and  $O_3$  in summer in the YRD. Regarding to regional transport pattern of  $PM_{2.5}$  and  $O_3$ , the results from both the complex network method and NAQPMS are comparable. The  $PM_{2.5}$  contribution ratio of Jiangsu, Anhui and Northern of Zhejiang to other regions is over 50%, which is much higher than that of other regions in the YRD. This is consistent with the results of the complex network method. The  $PM_{2.5}$  contribution ratio of Northern and central Jiangsu from other regions is the highest in the YRD region, but this does not match with the pattern of PIIS for  $PM_{2.5}$ . This is

probably because of the Northern and central Jiangsu is influenced by the interregional transport accounts for 57% and 39%, respectively from the regions out of YRD primarily. The PIIS of  $PM_{2.5}$  from the regions outside YRD is not in the scope of this study. Southern Zhejiang is mainly impacted by intraregional transport in the YRD region and such statement echoed with the results of the complex network method.

For  $O_3$ , the central regions of YRD including southern Jiangsu, Shanghai and northern Zhejiang accounts for 70%, 69% and 68%, respectively, is the main origin of regional transport in summer. Once again, this finding consistent with the results of the complex network method, which indicated the contribution ratio of northern and southern Zhejiang from the regions in the YRD is the highest, which accounts for 76% and 75%, respectively. Since these regions are the largest contributor of  $O_3$  pollution in the YRD region, they require tougher  $O_3$  pollution control and monitoring.

During pollution seasons, local  $PM_{2.5}$  concentration in each of the YRD region is impacted by interregional transport (22% ~ 58%), however, interregional transport played less important role on  $O_3$  side (24%~45%).



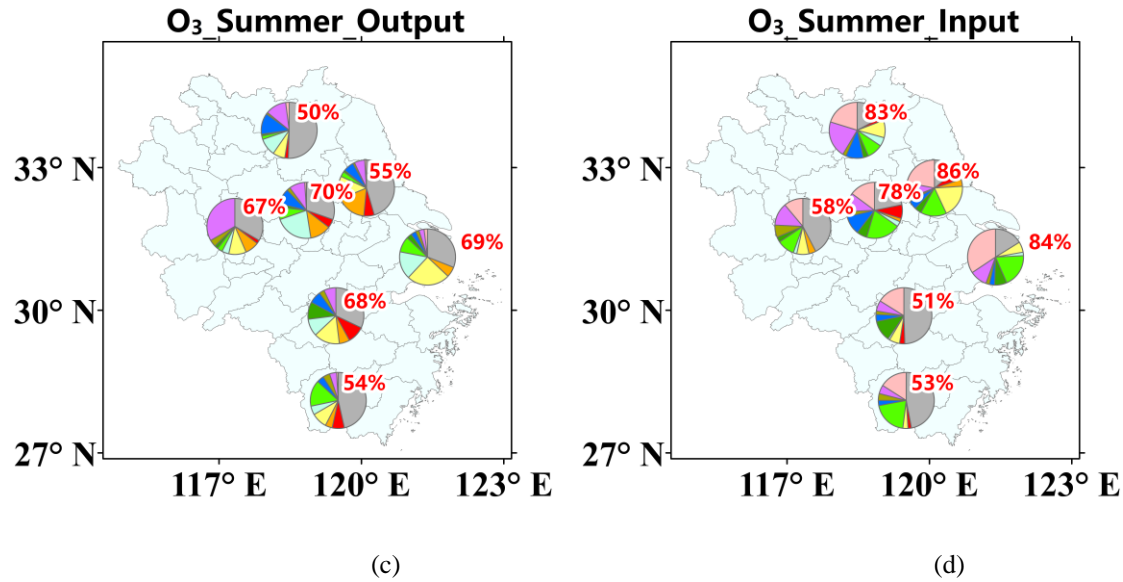


Figure 5. (a)  $PM_{2.5}$  source contribution to different regions in winter 2018. (b)  $PM_{2.5}$  source contribution from different regions in winter 2018. (c)  $O_3$  source contribution to different regions in summer 2018. (d)  $O_3$  source contribution from different regions in summer 2018. Red numbers mean the source apportionment ratio from or to the outside of the local region.

### 3.3 Analysis of $PM_{2.5}$ and $O_3$ pollution episodes

Taking at least three nearby cities with heavily or above pollution as the standard, 8 heavily pollution episodes are selected during 2018 as shown in Table 1. An individual episode of  $PM_{2.5}$  and  $O_3$  was chosen for examining  $PM_{2.5}$  and  $O_3$  regional transport pattern closely (Fig. 6). The results of other episodes can be found in Figure S7 to S12. In terms of  $PM_{2.5}$ , figure 6 shows heavy pollution was situated in the northern and central area of YRD. The highest daily average  $PM_{2.5}$  concentration was found in Xuzhou with  $227 \mu g/m^3$ .

Table 1 Regional pollution characteristics of heavily pollution episodes in YRD during 2018.

Pollutants	Starting date	Ending date	lasting days	peak of daily concentration	numbers	numbers	numbers	total polluted cities
					of cities lightly polluted	of cities medially polluted	of cities heavily polluted	
$PM_{2.5}$	2018/1/13	2018/1/23	11	273	5	5	29	39
	2018/1/28	2018/2/2	6	227	8	7	26	41



O <sub>3</sub>	2018/2/6	2018/2/10	5	177	13	15	11	39
	2018/11/23	2018/12/3	11	251	5	8	22	35
	2018/12/16	2018/12/24	9	181	17	10	6	33
	2018/4/25	2018/4/30	6	281	22	15	3	40
	2018/5/30	2018/6/8	10	281	10	17	9	36
	2018/6/11	2018/6/18	8	272	14	23	3	40

City	PM <sub>2.5</sub>							MDA8 O <sub>3</sub>						
	1/28	1/29	1/30	1/31	2/1	2/2	2/3	4/25	4/26	4/27	4/28	4/29	4/30	
LYG	99	156	122	117	139	72	43	183	175	167	186	180	204	
XZ	170	227	157	129	129	71	58	198	152	198	199	166	157	
SZ	131	190	157	136	88	59	39	196	159	180	212	170	163	
HB	151	195	162	130	91	54	39	193	138	178	194	161	169	
SQ	132	222	124	120	125	60	38	184	153	194	197	178	160	
HC	68	142	173	127	156	102	31	160	172	186	169	194	207	
BZ	138	150	82	95	64	47	35	174	130	170	182	153	151	
HA	104	183	134	126	126	70	32	180	173	197	221	189	181	
BB	111	175	143	129	95	77	31	183	174	152	214	190	153	
FY	120	113	100	97	63	52	36	160	129	167	179	145	144	
HN	124	167	129	132	82	78	46	182	167	175	214	170	150	
YZ	84	145	135	92	111	96	31	159	157	169	214	181	171	
TZ	48	163	185	156	155	129	31	162	175	179	191	217	179	
CZ	72	162	137	125	113	91	28	169	158	169	224	159	165	
NT	42	134	203	171	119	111	26	149	144	191	204	258	219	
ZJ	69	176	196	119	152	95	26	169	162	175	230	164	151	
NJ	60	172	167	132	138	99	28	173	158	181	205	157	126	
LA	72	95	92	85	65	65	30	164	141	172	191	151	146	
HF	90	144	134	127	95	93	35	185	137	197	207	134	132	
WX	44	138	183	205	162	134	35	184	208	232	247	230	175	
CZ	48	180	201	186	191	141	36	188	217	217	240	196	176	
MAS	62	184	182	168	156	115	33	190	177	172	238	183	151	
SZ	46	133	197	222	145	142	36	175	184	235	255	200	189	
SH	22	96	189	173	123	103	29	152	94	229	211	246	226	
WH	68	186	184	200	186	149	39	169	169	199	246	187	147	
TL	67	125	145	135	149	109	43	139	129	157	163	103	100	
HZ	41	126	139	182	141	109	27	177	208	229	281	164	161	
XC	55	133	131	170	122	111	31	146	145	145	177	126	104	
JX	47	133	168	217	157	119	44	217	206	272	268	193	203	
AQ	58	138	150	150	133	113	32	158	161	175	171	103	110	
CZ	71	151	147	172	127	112	41	166	137	189	147	99	115	
ZS	11	69	97	111	94	84	34	153	109	232	226	136	90	
HS	34	68	64	83	84	59	42	103	88	92	112	61	61	
HZ	33	109	128	135	108	102	42	170	225	199	239	118	137	
NB	26	112	120	153	144	110	41	206	207	249	221	175	145	
SX	42	110	153	151	127	117	42	194	240	235	267	146	142	
JH	21	61	99	85	82	89	57	192	187	214	164	140	94	
QZ	20	44	88	76	80	91	62	169	168	145	146	97	76	
TZ	13	71	101	81	83	74	78	201	199	210	221	152	109	
LS	10	41	85	58	69	73	100	167	160	119	165	136	92	
WZ	25	57	81	43	46	51	70	166	195	130	182	117	126	

Figure 6. Daily PM<sub>2.5</sub> and MDA8 O<sub>3</sub> concentration of prefectural cities during a PM<sub>2.5</sub> and O<sub>3</sub> pollution episode.

On PM<sub>2.5</sub> side, the highest value of POIS occurred in Anhui and the highest value of PIIS occurred in coastal areas (Figure 7). It illustrated PM<sub>2.5</sub> pollution of the coastal areas was impacted by regional transport from Anhui in this particular episode. This echoed with the results of source apportionment method by NAQPMS. Total contribution from Anhui, which was the largest contributor in the YRD during this period, to southern Jiangsu, Shanghai, northern Zhejiang and southern Zhejiang were 24%, 10%, 37% and 20%, respectively.

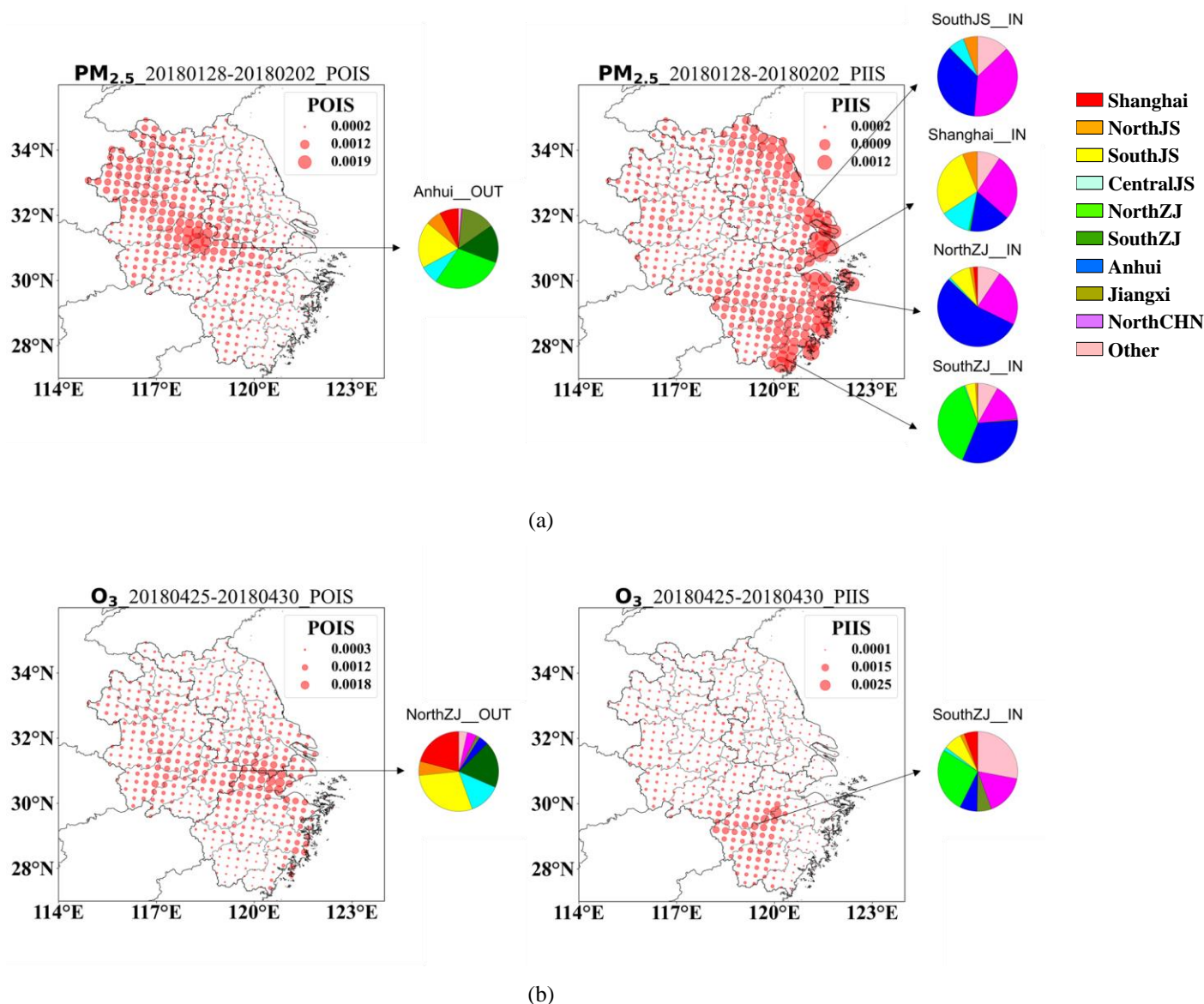


Figure 7. (a) The POIS and PIIS of PM<sub>2.5</sub> and source apportionment in key areas of YRD during the pollution episode 1/13-2/23 in 2018. (b) The POIS and PIIS of O<sub>3</sub> and source apportionment in key areas of YRD during the pollution episode 4/25-4/30 in 2018.

Figure 6 also presents the variation of MDA8 O<sub>3</sub> concentration in prefectural cities in the YRD during the O<sub>3</sub> pollution episode, 4/25 – 4/30 in 2018. The highest daily average concentration occurred in Hangzhou, about 281 μg/m<sup>3</sup>. POIS in the northern areas of Zhejiang and PIIS in southern areas of Zhejiang were higher than other areas. This means northern Zhejiang is the dominant contributor to this O<sub>3</sub> pollution episode (Figure 7). The result from the source apportionment method by NAQPMS echoed the same finding. The contribution ratio



from the northern Zhejiang to the southern Zhejiang accounts for 23%, which is the top contributor in the YRD based on the source apportionment method.

#### 4 Conclusions

Regional transport of air pollutants has been investigated in various studies(Ding et al., 2017; Dong et al., 2020; Fan et al., 2015; Lu and Fung 2016; Sun et al., 2020; Zhu et al., 2011).The Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) and chemical transport models(CTM) have been extensively applied. The HYSPLIT is mainly utilized to focus on the meteorological analysis and generally associated with cluster analysis (CA), potential source contribution function (PSCF) and concentration weighted trajectory (CWT). The CTM coupled with the source apportionment method includes the Particle Source Apportionment Technology (PSAT) and Ozone Source Apportionment Technology (OSAT) in the Comprehensive Air Quality Model with extensions (CAMx), the Integrated Source Apportionment (ISAM) in the Community Multi-scale Air Quality model (CMAQ), and the on-line source-tagged model of the Nested Air Quality Predicting Modeling System (NAQPMS), the complex network analysis is seldom applied to investigate the regional transport of air pollutants, despite it has been widely applied to characterizing a wide range of scientific problem(Tian and Gunes 2014; Wang et al., 2018).

In this study, a complex network method and a source apportionment model were used to analyze regional  $PM_{2.5}$  and  $O_3$  transport pattern of in the YRD region. Complex network method, this new approach not only help identifying major receptor and contributor region, but also present a thorough view in uncovering underlying key nodes associated with regional transport of  $PM_{2.5}$  and  $O_3$  in the YRD region. Our findings in terms of regional transport of  $PM_{2.5}$  and  $O_3$  pattern are consistent with existing research (Ding et al., 2017; Fu et al., 2016; Li et al., 2016; Shu et al., 2020; Sun et al., 2020; Tang et al., 2016).

Source apportionment is a computational intensive method. With this constraint, a typical application setup with a limited number of regions to study source and receptor relationship. A regional could be a combination of nearby provinces or an area with vicinal cities. This work not only has proven complex network methodology produced comparable result as source apportion technique. It can also provide contribution information in grid cell level. Nevertheless,it should be noted that these results has the following limitations: (1) It is not able to discern contribution

from the pollutant perspective, either in pollutant type or individual pollutant species. (2) Local emission sources were not taken into consideration. For future research, a more quantitative uncertainty analyses on the regional transport will be conducted to enhance accuracy of this method.

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## References

- Ainsworth EA, Yendrek CR, Sitch S, Collins WJ, Emberson LD (2012) The effects of tropospheric ozone on net primary productivity and implications for climate change *Annu Rev Plant Biol* 63:637-661 doi:10.1146/annurev-arplant-042110-103829
- An J, Zou J, Wang J, Lin X, Zhu B (2015) Differences in ozone photochemical characteristics between the megacity Nanjing and its suburban surroundings, Yangtze River Delta, China *Environ Sci Pollut Res Int* 22:19607-19617 doi:10.1007/s11356-015-5177-0
- Anenberg SC, Horowitz LW, Tong DQ, West JJ (2010) An estimate of the global burden of anthropogenic ozone and fine particulate matter on premature human mortality using atmospheric modeling *Environ Health Perspect* 118:1189-1195 doi:10.1289/ehp.0901220
- Atkinson R (2000) <Atmospheric chemistry of VOCs and NOx.pdf>
- Bell ML, Peng RD, Dominici F (2006) The exposure-response curve for ozone and risk of mortality and the adequacy of current ozone regulations *Environ Health Perspect* 114:532-536 doi:10.1289/ehp.8816
- Brin S, Page L (2012) Reprint of: The anatomy of a large-scale hypertextual web search engine *Computer Networks* 56:3825-3833 doi:10.1016/j.comnet.2012.10.007
- Chan CK, Yao X (2008) Air pollution in mega cities in China *Atmospheric Environment* 42:1-42 doi:10.1016/j.atmosenv.2007.09.003
- Chen X et al. (2020) Common source areas of air pollution vary with haze intensity in the Yangtze River Delta, China *Environmental Chemistry Letters* 18:957-965 doi:10.1007/s10311-020-00976-0
- Cohen AJ et al. (2017) Estimates and 25-year trends of the global burden of disease attributable to ambient air pollution: an analysis of data from the Global Burden of Diseases Study 2015 *The Lancet* 389:1907-1918 doi:10.1016/s0140-6736(17)30505-6
- CRUTZEN P (1973) <A discussion of the chemistry of some minor constituents in the stratosphere and.pdf>
- CSC (2013) Action Plan on Air Pollution Prevention and Control (in Chinese) (Chinese State Council, 2013);. China State Council (CSC). Accessed 1 2021
- Daellenbach KR et al. (2020) Sources of particulate-matter air pollution and its oxidative potential in Europe *Nature* 587:414-419 doi:10.1038/s41586-020-2902-8
- Dang R, Liao H (2019) Radiative Forcing and Health Impact of Aerosols and Ozone in China as the Consequence of Clean Air Actions over 2012–2017 *Geophysical Research Letters* 46:12511-12519 doi:10.1029/2019gl084605
- Ding AJ et al. (2013) Ozone and fine particle in the western Yangtze River Delta: an overview of 1 yr data at the SORPES station *Atmospheric Chemistry and Physics* 13:5813-5830 doi:10.5194/acp-13-5813-2013
- Ding XX et al. (2017) Long-range and regional transported size-resolved atmospheric aerosols during summertime in urban Shanghai *Science of the Total Environment* 583:334-343 doi:10.1016/j.scitotenv.2017.01.073
- Dong Z, Wang S, Xing J, Chang X, Ding D, Zheng H (2020) Regional transport in Beijing-Tianjin-Hebei region and its changes during 2014-2017: The impacts of meteorology and emission reduction *The Science of the total environment* 737:139792 doi:10.1016/j.scitotenv.2020.139792

- Duhan N, Sharma AK, Bhatia KK, Ieee (2009) Page Ranking Algorithms: A Survey. 2009 Ieee International Advance Computing Conference, Vols 1-3. doi:10.1109/iadcc.2009.4809246
- Fan Q et al. (2015) Process analysis of regional aerosol pollution during spring in the Pearl River Delta region, China Atmospheric Environment 122:829-838 doi:10.1016/j.atmosenv.2015.09.013
- Forouzanfar MH et al. (2015) Global, regional, and national comparative risk assessment of 79 behavioural, environmental and occupational, and metabolic risks or clusters of risks in 188 countries, 1990–2013: a systematic analysis for the Global Burden of Disease Study 2013 The Lancet 386:2287-2323 doi:10.1016/s0140-6736(15)00128-2
- Franceschet M (2011) PageRank: Standing on the Shoulders of Giants Communications of the ACM 54:92-101 doi:10.1145/1953122.1953146
- Fu QY et al. (2010) Source, long-range transport, and characteristics of a heavy dust pollution event in Shanghai Journal of Geophysical Research-Atmospheres 115:12 doi:10.1029/2009jd013208
- Fu X, Cheng Z, Wang SX, Hua Y, Xing J, Hao JM (2016) Local and Regional Contributions to Fine Particle Pollution in Winter of the Yangtze River Delta, China Aerosol and Air Quality Research 16:1067-1080 doi:10.4209/aaqr.2015.08.0496
- Gao J, Zhu B, Xiao H, Kang H, Hou X, Shao P (2016) A case study of surface ozone source apportionment during a high concentration episode, under frequent shifting wind conditions over the Yangtze River Delta, China Science of The Total Environment 544:853-863 doi:10.1016/j.scitotenv.2015.12.039
- Hollaway MJ, Arnold SR, Challinor AJ, Emberson LD (2012) Intercontinental trans-boundary contributions to ozone-induced crop yield losses in the Northern Hemisphere Biogeosciences 9:271-292 doi:10.5194/bg-9-271-2012
- Horowitz LW (2006) Past, present, and future concentrations of tropospheric ozone and aerosols: Methodology, ozone evaluation, and sensitivity to aerosol wet removal Journal of Geophysical Research-Atmospheres 111 doi:10.1029/2005jd006937
- J. Fuhrer LSibaMRA (1997) <Critical levels for ozone effects on vegetation in Europe.pdf>
- Kleinman LI, Daum PH, Lee Y-N, Nunnermacker LJ, Springston SR, Weinstein-Lloyd J, Rudolph J (2002) Ozone production efficiency in an urban area Journal of Geophysical Research: Atmospheres 107:ACH 23-21-ACH 23-12 doi:10.1029/2002jd002529
- Lefohn AS, Malley CS, Simon H, Wells B, Xu X, Zhang L, Wang T (2017) Responses of human health and vegetation exposure metrics to changes in ozone concentration distributions in the European Union, United States, and China Atmospheric Environment 152:123-145 doi:10.1016/j.atmosenv.2016.12.025
- Li J et al. (2020) A More Important Role for the Ozone - S(IV) Oxidation Pathway Due to Decreasing Acidity in Clouds Journal of Geophysical Research: Atmospheres 125 doi:10.1029/2020jd033220
- Li K, Jacob DJ, Liao H, Shen L, Zhang Q, Bates KH (2019a) Anthropogenic drivers of 2013-2017 trends in summer surface ozone in China Proc Natl Acad Sci U S A 116:422-427 doi:10.1073/pnas.1812168116
- Li L et al. (2016) Source apportionment of surface ozone in the Yangtze River Delta, China in the summer of 2013 Atmospheric Environment 144:194-207 doi:10.1016/j.atmosenv.2016.08.076
- Li L et al. (2015) Source apportionment of fine particles and its chemical components over the Yangtze River Delta, China during a heavy haze pollution episode Atmospheric Environment 123:415-429 doi:10.1016/j.atmosenv.2015.06.051
- Li M et al. (2017) MIX: a mosaic Asian anthropogenic emission inventory under the international collaboration framework of the MICS-Asia and HTAP Atmospheric Chemistry and Physics 17:935-963 doi:10.5194/acp-17-935-2017
- Li R, Mei X, Wei L, Han X, Zhang M, Jing Y (2019b) Study on the contribution of transport to PM<sub>2.5</sub> in typical regions of China using the regional air quality model RAMS-CMAQ Atmospheric Environment 214 doi:10.1016/j.atmosenv.2019.116856
- Liao TT et al. (2017) Heavy pollution episodes, transport pathways and potential sources of PM<sub>2.5</sub> during the winter of 2013 in Chengdu (China) Science of the Total Environment 584:1056-1065 doi:10.1016/j.scitotenv.2017.01.160
- Liu J et al. (2019) Quantifying the impact of synoptic circulation patterns on ozone variability in northern China from April to October 2013–2017 Atmospheric Chemistry and Physics 19:14477-14492 doi:10.5194/acp-19-14477-2019
- Liu Y et al. (2018) Estimation of biogenic VOC emissions and its impact on ozone formation over the Yangtze River Delta region, China Atmospheric Environment 186:113-128 doi:10.1016/j.atmosenv.2018.05.027
- Logan JA, Prather MJ, Wofsy SC, McElroy MB (1981) Tropospheric chemistry: A global perspective Journal of Geophysical Research 86 doi:10.1029/JC086iC08p07210

- Lu X et al. (2020) Rapid Increases in Warm-Season Surface Ozone and Resulting Health Impact in China Since 2013 *Environmental Science & Technology Letters* 7:240-247 doi:10.1021/acs.estlett.0c00171
- Lu XC, Fung JCH (2016) Source Apportionment of Sulfate and Nitrate over the Pearl River Delta Region in China *Atmosphere* 7:13 doi:10.3390/atmos7080098
- Ma T et al. (2019) Air pollution characteristics and their relationship with emissions and meteorology in the Yangtze River Delta region during 2014-2016 *Journal of Environmental Sciences* 83:8-20 doi:10.1016/j.jes.2019.02.031
- Mauzerall DL, Wang XP (2001) Protecting agricultural crops from the effects of tropospheric ozone exposure: Reconciling science and standard setting in the United States, Europe, and Asia *Annual Review of Energy and the Environment* 26:237-268 doi:10.1146/annurev.energy.26.1.237
- Medo M (2013) Network-based information filtering algorithms: Ranking and recommendation vol 55. doi:10.1007/978-1-4614-6729-8\_16
- Nerem ACALGMRS (2013) <Climate Change 2013 The Physical Science Basis, Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change.pdf>.
- Nie W, Wang T, Wang W, Wei X, Liu Q (2013) Atmospheric concentrations of particulate sulfate and nitrate in Hong Kong during 1995-2008: Impact of local emission and super-regional transport *Atmospheric Environment* 76:43-51 doi:10.1016/j.atmosenv.2012.07.001
- Page L, Brin S, Motwani R, Winograd T (1999) The pagerank citation ranking: Bringing order to the web. Stanford InfoLab,
- Shu L, Wang T, Han H, Xie M, Chen P, Li M, Wu H (2020) Summertime ozone pollution in the Yangtze River Delta of eastern China during 2013-2017: Synoptic impacts and source apportionment *Environ Pollut* 257:113631 doi:10.1016/j.envpol.2019.113631
- Shu L, Wang TJ, Xie M, Li MM, Zhao M, Zhang M, Zhao XY (2019) Episode study of fine particle and ozone during the CAPUM-YRD over Yangtze River Delta of China: Characteristics and source attribution *Atmospheric Environment* 203:87-101 doi:10.1016/j.atmosenv.2019.01.044
- Sillman S (1999) <The relation between ozone, NO<sub>x</sub> and hydrocarbons in urban and polluted rural environments.pdf>
- Stein AF, Draxler RR, Rolph GD, Stunder BJB, Cohen MD, Ngan F (2015) NOAA's HYSPLIT Atmospheric Transport and Dispersion Modeling System *Bulletin of the American Meteorological Society* 96:2059-2077 doi:10.1175/bams-d-14-00110.1
- Sun JJ, Huang L, Liao H, Li JY, Hu JL (2017) Impacts of Regional Transport on Particulate Matter Pollution in China: a Review of Methods and Results *Current Pollution Reports* 3:182-191 doi:10.1007/s40726-017-0065-5
- Sun P et al. (2020) Impact of air transport and secondary formation on haze pollution in the Yangtze River Delta: In situ online observations in Shanghai and Nanjing *Atmospheric Environment* 225 doi:10.1016/j.atmosenv.2020.117350
- Tacchella A, Cristelli M, Caldarelli G, Gabrielli A, Pietronero L (2012) A New Metrics for Countries' Fitness and Products' Complexity *Scientific Reports* 2 doi:10.1038/srep00723
- Tang H, Liu G, Zhu J, Han Y, Kobayashi K (2013) Seasonal variations in surface ozone as influenced by Asian summer monsoon and biomass burning in agricultural fields of the northern Yangtze River Delta *Atmospheric Research* 122:67-76 doi:10.1016/j.atmosres.2012.10.030
- Tang LL et al. (2016) Regional contribution to PM<sub>1</sub> pollution during winter haze in Yangtze River Delta, China *Science of the Total Environment* 541:161-166 doi:10.1016/j.scitotenv.2015.05.058
- Thompson AM (2016) <The oxidizing capacity of the earth's atmosphere.pdf>
- Tian G, Gunes MH (2014) Complex Network Analysis of Ozone Transport. In: *Complex Networks V. Studies in Computational Intelligence*. pp 87-96. doi:10.1007/978-3-319-05401-8\_9
- Vu TV, Shi Z, Cheng J, Zhang Q, He K, Wang S, Harrison RM (2019) Assessing the impact of clean air action on air quality trends in Beijing using a machine learning technique *Atmospheric Chemistry and Physics* 19:11303-11314 doi:10.5194/acp-19-11303-2019
- Wang T, Xue L, Brimblecombe P, Lam YF, Li L, Zhang L (2017a) Ozone pollution in China: A review of concentrations, meteorological influences, chemical precursors, and effects *Sci Total Environ* 575:1582-1596 doi:10.1016/j.scitotenv.2016.10.081
- Wang TJ, Lam KS, Xie M, Wang XM, Carmichael G, Li YS (2017b) Integrated studies of a photochemical smog episode in Hong Kong and regional transport in the Pearl River Delta of China *Tellus B: Chemical and Physical Meteorology* 58:31-40 doi:10.1111/j.1600-0889.2005.00172.x

- 502 Wang Y, Wang H, Zhang S (2018) A weighted higher-order network analysis of fine particulate matter (PM<sub>2.5</sub>)  
503 transport in Yangtze River Delta *Physica A: Statistical Mechanics and its Applications* 496:654-662  
504 doi:10.1016/j.physa.2017.12.096
- 505 Wei Y et al. (2020) Nocturnal PM<sub>2.5</sub> explosive growth dominates severe haze in the rural North China Plain  
506 *Atmospheric Research* 242 doi:10.1016/j.atmosres.2020.105020
- 507 Wu J-B, Wang Q, Chen H, Zhang Y, Wild O (2017a) On the Origin of Surface Ozone Episode in Shanghai over  
508 Yangtze River Delta during a Prolonged Heat Wave *Aerosol and Air Quality Research* 17:2804-2815  
509 doi:10.4209/aaqr.2017.03.0101
- 510 Wu JB et al. (2017b) Development of an on-line source-tagged model for sulfate, nitrate and ammonium: A  
511 modeling study for highly polluted periods in Shanghai, China *Environ Pollut* 221:168-179  
512 doi:10.1016/j.envpol.2016.11.061
- 513 Wu R, Xie S (2017) Spatial Distribution of Ozone Formation in China Derived from Emissions of Speciated  
514 Volatile Organic Compounds *Environmental Science & Technology* 51:2574-2583  
515 doi:10.1021/acs.est.6b03634
- 516 X. Xu WL, T. Wang, P. Yan, J. Tang, Z. Meng, and Y. Wang (2008) <Long-term trend of surface ozone at a  
517 regional background station in eastern China 1991–200.pdf>
- 518 Xu X, Lin W, Wang T, Yan P, Tang J, Meng Z, Wang Y (2008) Long-term trend of surface ozone at a regional  
519 background station in eastern China 1991-2006: enhanced variability *Atmospheric Chemistry and Physics*  
520 8:2595-2607 doi:10.5194/acp-8-2595-2008
- 521 Yang Y, Liao H, Li J (2014) Impacts of the East Asian summer monsoon on interannual variations of summertime  
522 surface-layer ozone concentrations over China *Atmospheric Chemistry and Physics* 14:6867-6879  
523 doi:10.5194/acp-14-6867-2014
- 524 Yue X et al. (2017) Ozone and haze pollution weakens net primary productivity in China *Atmospheric Chemistry*  
525 *and Physics* 17:6073-6089 doi:10.5194/acp-17-6073-2017
- 526 Zhao N, Wang G, Li G, Lang J, Zhang H (2020) Air pollution episodes during the COVID-19 outbreak in the  
527 Beijing-Tianjin-Hebei region of China: An insight into the transport pathways and source distribution  
528 *Environ Pollut* 267:115617 doi:10.1016/j.envpol.2020.115617
- 529 Zhou JB, Xing ZY, Deng JJ, Du K (2016) Characterizing and sourcing ambient PM<sub>2.5</sub> over key emission regions in  
530 China I: Water-soluble ions and carbonaceous fractions *Atmospheric Environment* 135:20-30  
531 doi:10.1016/j.atmosenv.2016.03.054
- 532 Zhu L, Huang X, Shi H, Cai X, Song Y (2011) Transport pathways and potential sources of PM<sub>10</sub> in Beijing  
533 *Atmospheric Environment* 45:594-604 doi:10.1016/j.atmosenv.2010.10.040