Regional transport of PM2.5 and O3 based on complex network method and source apportionment technology in the Yangtze River Delta,China

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November 26, 2022

Abstract

Ground-level ozone(O_3) and atmospheric fine particulate matter ($PM_{2.5}$) 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 $PM_{2.5}$ and O_3 pollution is essential for the development of regional cooperative prevention and control strategies. This study shows annual concentration of $PM_{2.5}$ in the YRD decreased by 18.5% from 2015 to 2018, while average of daily maximum 8-h average(MDA8) O_3 concentration during March to October increased by 16.3%. A complex network method is developed to investigate regional transport of $PM_{2.5}$ and O_3 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 $PM_{2.5}$ and O_3 over the YRD. The northern part of the YRD contributes much more than other areas for $PM_{2.5}$ 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 O_3 in the YRD in summer,which accounts for about 70%. Intraregional transport played major role in $PM_{2.5}$ than $O_{3.}$ 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.

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Key Points:

- $PM_{2.5}$ decreased while O_3 increased in the YRD over the years from 2015 to 2018.
- The complex network method was applied to investigate regional transport of $PM_{2.5}$ and O_3 between grid cells.
- Regional transport played significant roles in $PM_{2.5}$ and O_3 pollution in the YRD.

Abstract

Ground-level ozone(O_3) and atmospheric fine particulate matter ($PM_{2.5}$) 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 $PM_{2.5}$ and O_3 pollution is essential for the development of regional cooperative prevention and control strategies. This study shows annual concentration of $PM_{2.5}$ in the YRD decreased by 18.5% from 2015 to 2018, while average of daily maximum 8-h average(MDA8) O_3 concentration during March to October increased by 16.3%. A complex network method is developed to investigate regional transport of $PM_{2.5}$ and O_3 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 $PM_{2.5}$ and O_3 over the YRD. The northern part of the YRD contributes much more than other areas for $PM_{2.5}$ 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 O_3 in the YRD in summer,which accounts for about 70%. Intraregional transport played major role in $PM_{2.5}$ than 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 (O_3) and atmospheric fine particulate matter （$PM_{2.5}$）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). 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 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). $PM_{2.5}$ 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).

$PM_{2.5}$ and 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. $PM_{2.5}$ 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 $PM_{2.5}$ in more than half of major cities in China are still much higher than NAAQS Grade 2 annual standard of 35 $\mu g/m^3$. In addition, O_3

pollution worsened over the same period, regional $PM_{2.5}$ and 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 O_3 and its precursors. Some researches revealed that both interregional and intraregional transport play a significant role in regional $PM_{2.5}$ 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 $PM_{2.5}$ and 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 $PM_{2.5}$ and 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 $PM_{2.5}$ and 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.

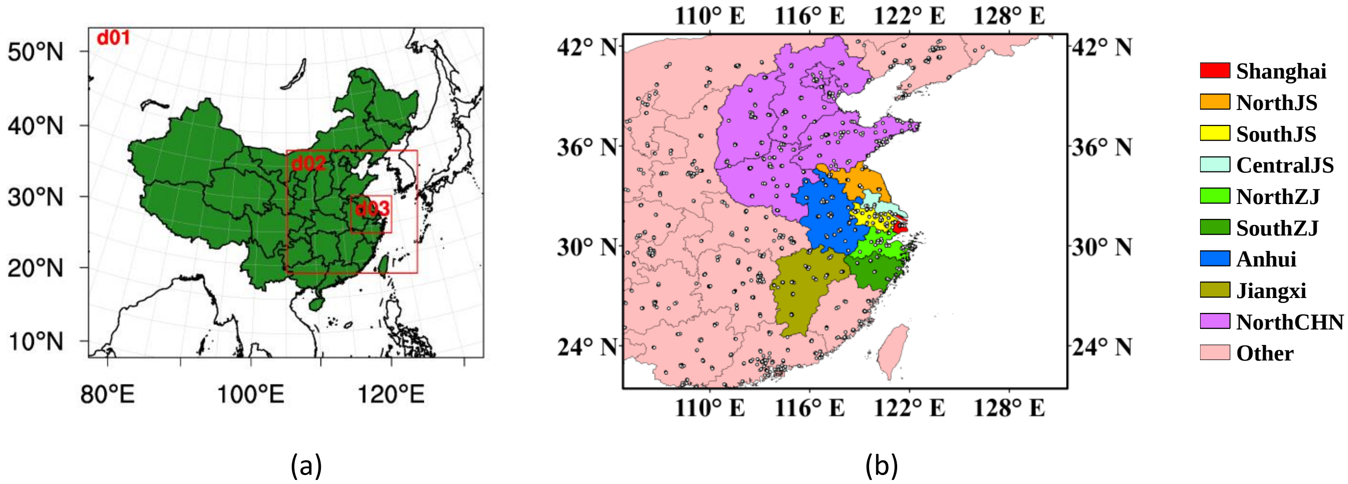


Figure 1. (a) The 27-km (D1), 9-km (D2) and 3-km(D3) domains in the NAQPMS simulation. (b) Subdivided source regions applied in D2 domain. The grey circle symbols represent air quality stations (NorthJS:Xuzhou-Lianyungang-Suqian-Huaian-Yancheng; SouthJS: Nanjing-Zhenjiang-Changzhou-Wuxi-Suzhou;CentralJS:Yangzhou-Taizhou-Nantong;NorthZJ:Hangzhou-Huzhou-Jiaxing-Shaoxing-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 metholody

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 trajecoties 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 tranjectories information of each grids 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 netword 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

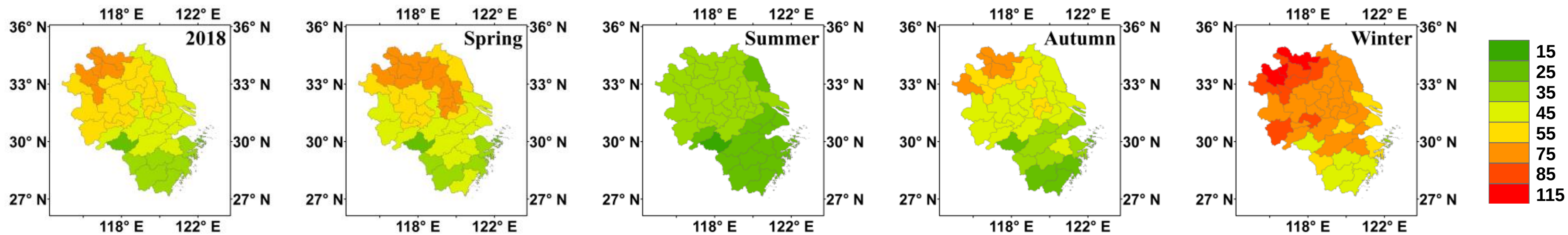
3.1 Characteristics of $PM_{2.5}$ and O_3 in YRD

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 emimssion 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.

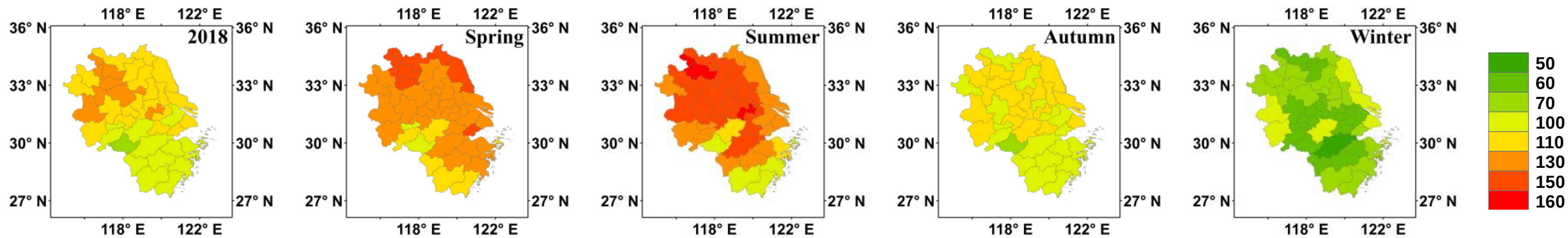
The seasonal spatial distribution show $PM_{2.5}$ concentration in winter is the highest in four seasons. This is mainly because the YRD region is heavily affected by the northerly and westerly wind. $PM_{2.5}$ concentration is also much higher in spring and fall than summer, especially in the northern 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

O_3 production. In autumn and winter, the coastal areas O_3 level is slightly higher than the other areas. Winter time O_3 production drops significantly, elevated level of O_3 in the coastal areas is likely due to incoming O_3 from the ocean by onshore breeze.



(a)

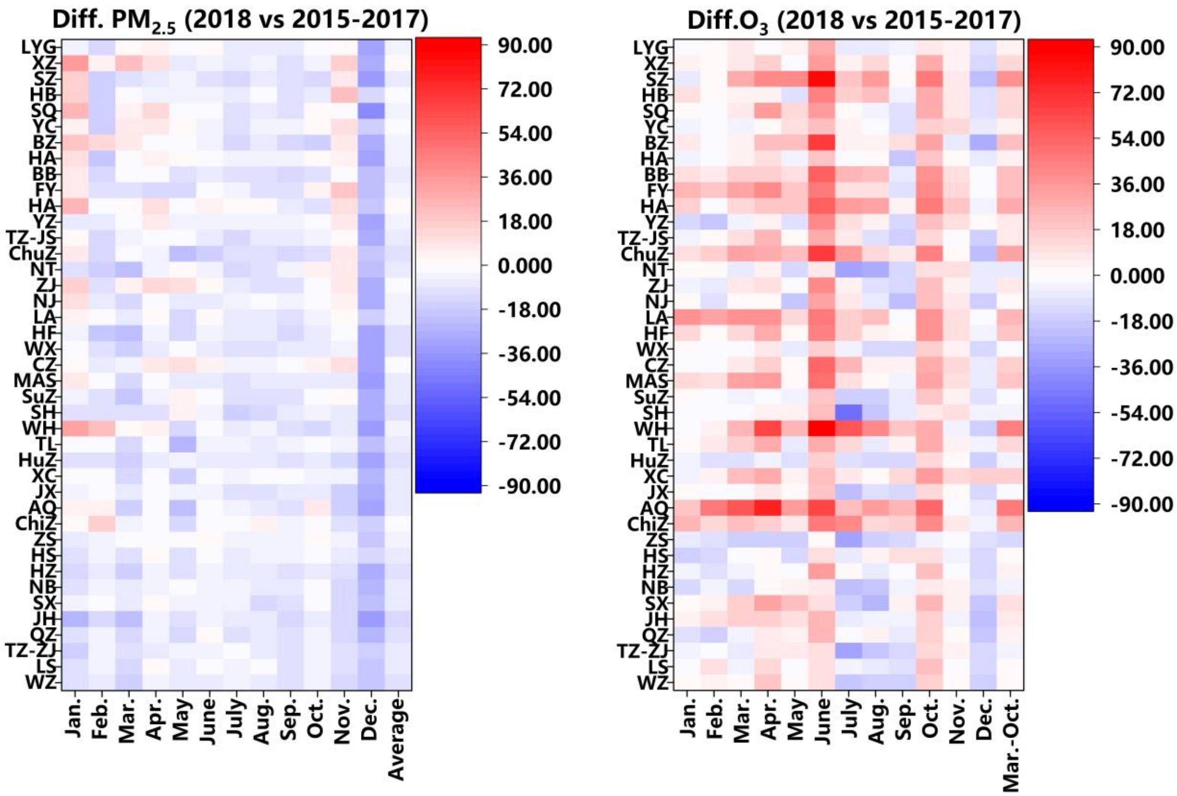


(b)

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 $PM_{2.5}$ and MDA8 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 $PM_{2.5}$ 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 $PM_{2.5}$ between 2018 and the average of 2015 to 2017 in Jinhua shows the largest decreasement,which is 13.9%, while the concentration difference in Xuzhou show the largest increasement, which is 1.7%. Annual concentration of $PM_{2.5}$ in the YRD decreased by 18.5% with a rate of 3.3 $\mu g/m^3$ per year from 2015 to 2018, while average concentration of daily maximum 8-h average(MDA8) O_3 during March to October increased by 16.3% with a rate of 5.7 $\mu g/m^3$ per year. $PM_{2.5}$ pollution episodes still occur frequently under adverse meteorological conditions with dominent emission sources in the YRD. It is noted that concentration of O_3 increased significantly in many cities in 2018, however $PM_{2.5}$ has shown a decrease. This is probably related to emission mitigation strategy which focused on NOx, SO_2 and primary particulate matter reduction, started in 2013. Research has shown anthropogenic NOx emission in China decreased by 20% during 2013 to 2017, while VOC emissions increased by 2%, O_3 increase could be driven by the $PM_{2.5}$ decrease, due to the role of $PM_{2.5}$ as scavenger of hydroperoxy (HO_2) radicals that would otherwise react with nitric oxide (NO) to produce ozone (Ma et al., 2019). From Figure 3, the MDA8 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 $\mu g/m^3$. In summer, the difference was much smaller, and MDA8 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 O_3 and $PM_{2.5}$ concentration in the YRD, particularly in four different cities: Hefei, Nanjing, Shanghai, and Hangzhou (Figure S3 – S6). Figure S3 to S6, showed faily strong correlation between $PM_{2.5}$ and 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:Jiaxing;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.

Sesson 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 $PM_{2.5}$ precusors, 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 NOx emission

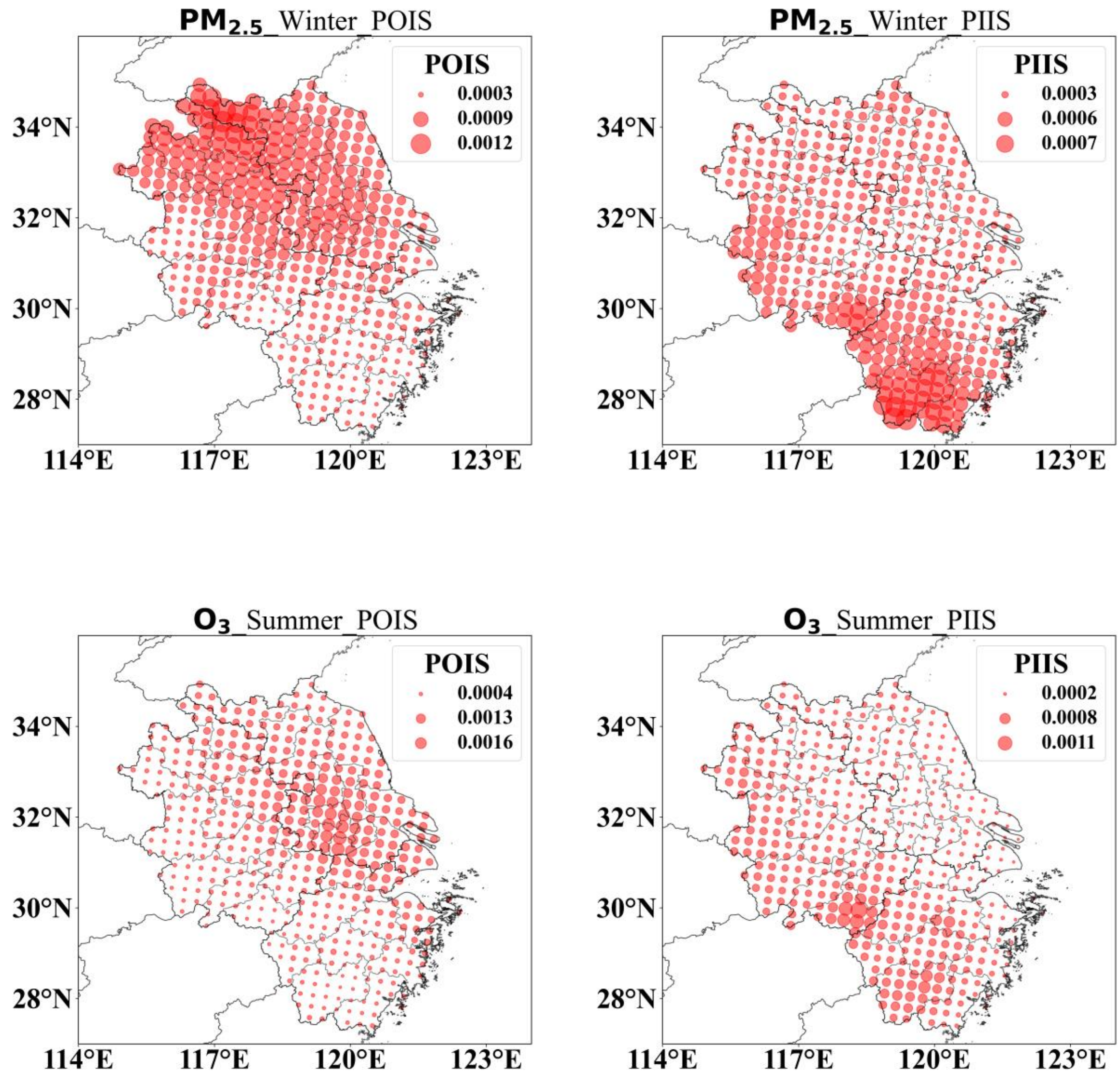
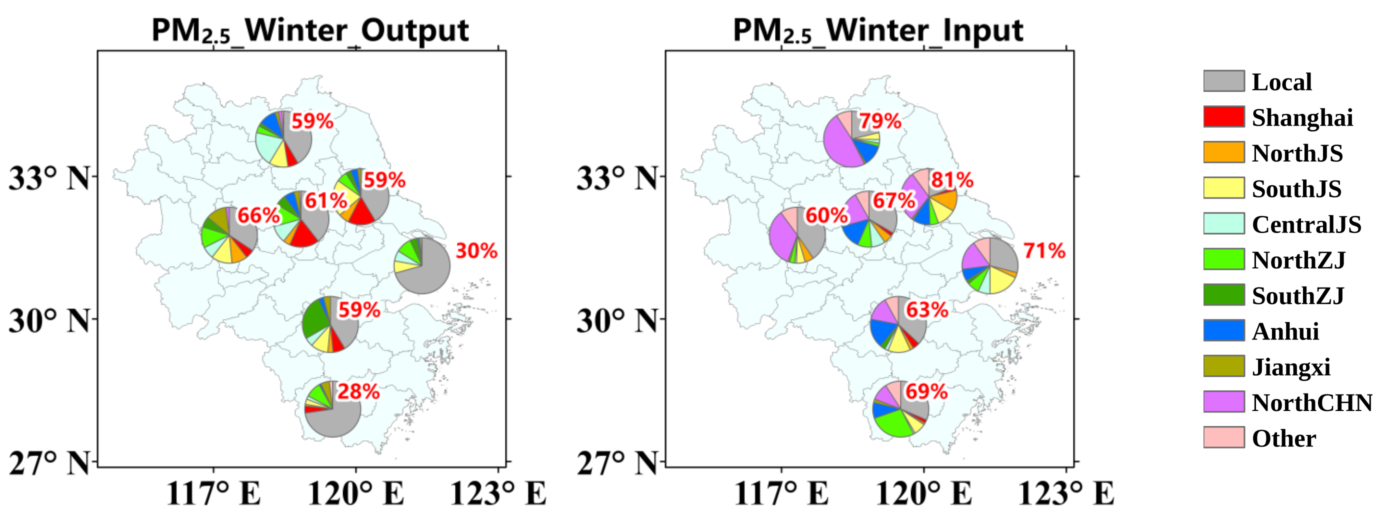


Figure 4. The POIS and PIIS of $PM_{2.5}$ and O_3 for nodes in YRD during 2018.

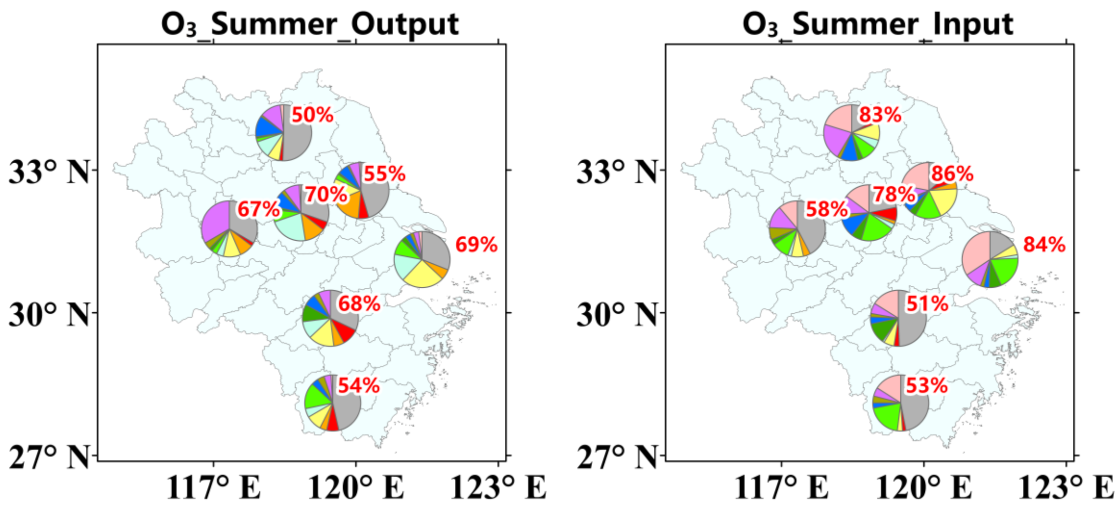
Figure 5 shows the source apportionment results for $PM_{2.5}$ in winter and O_3 in summer in the YRD. Regarding to regional transport pattern of $PM_{2.5}$ and O_3, the results from both the complex network method and NAQPMS are comparable. The PM2.5 contribution ratio of Jiangsu, Anhui and Northern of Zhejiang to other regions is over 50%, which is much higher than that of other regions in the YRD. This is consistant with the results of the complex network method. The PM2.5 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 consistant 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 O3 pollution in the YRD region, they require tougher O3 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%).



(a) (b)



(c) (d)

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 choosen 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 Xuzhouwith 227 μg/m^3.

Table1 Regional pollution characteristics of heavily pollution episodes in YRD during 2018.

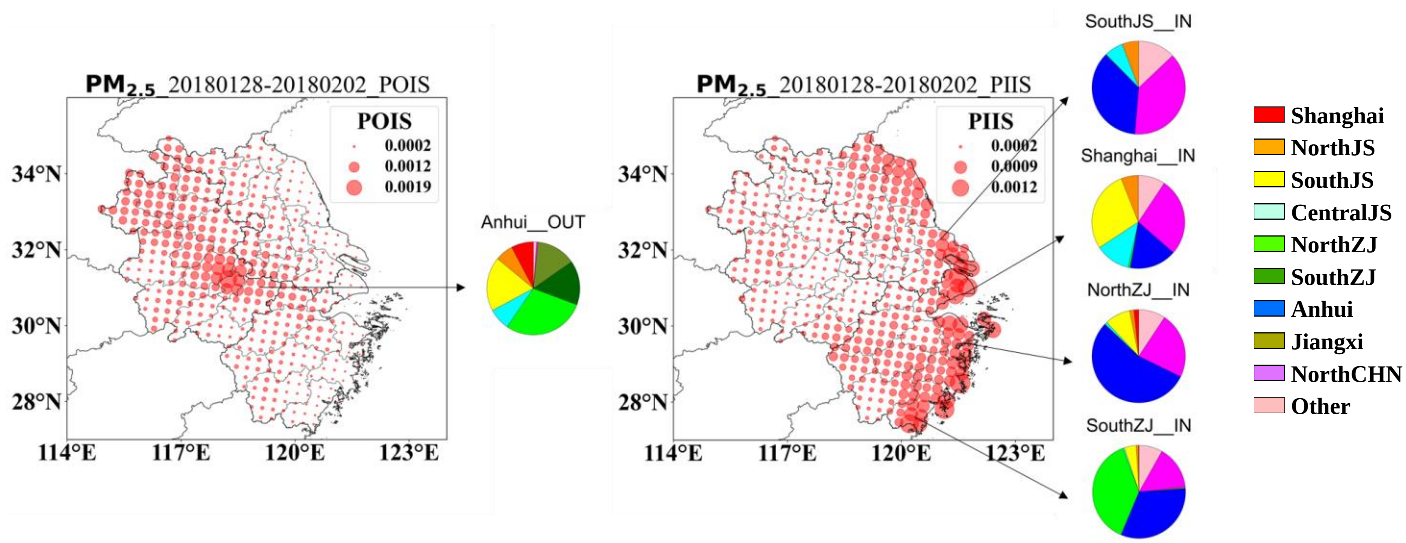
Pollutants	Starting date	Ending date	lasting days	peak of daily concentration	numbers of cities lightly polluted	numbers of cities medially polluted	numbers of cities heavily polluted	total polluted cities
$PM_{2.5}$	2018/1/13	2018/1/23	11	273	5	5	29	39
	2018/1/28	2018/2/2	6	227	8	7	26	41

	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
O_3	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

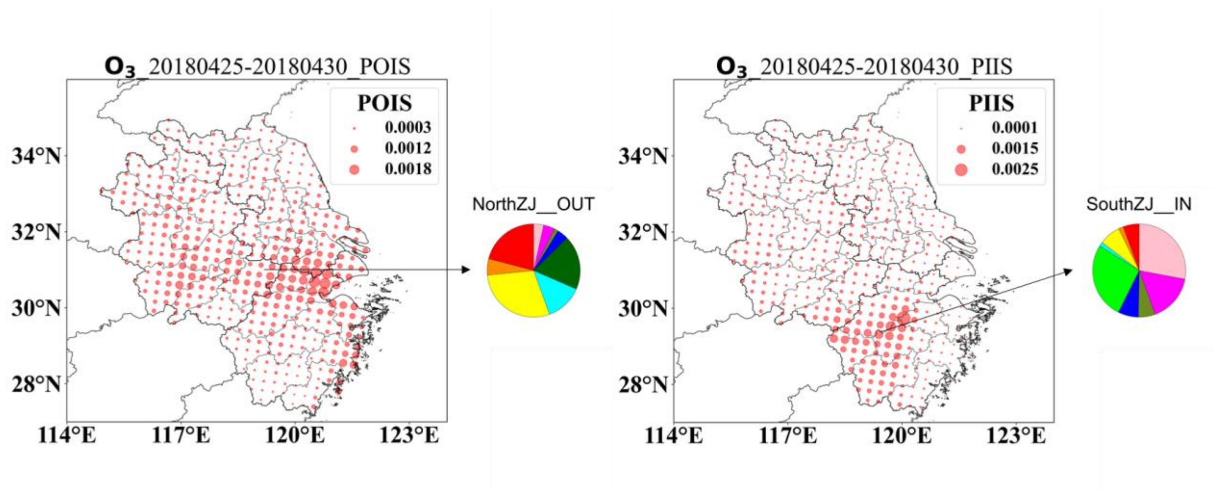
	$PM_{2.5}$							MDA8 O_3					
City	1/28	1/29	1/30	1/31	2/1	2/2	2/3	4/25	4/26	4/27	4/28	4/29	4/30
LYG	99	156	122	117	139	72	43	183	175	167	186	180	204
XZ	170	227	157	129	129	71	58	198	152	198	199	166	157
SZ	131	190	157	136	88	59	39	196	159	180	212	170	163
HB	151	195	162	130	91	54	39	193	138	178	194	161	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 $PM_{2.5}$ and MDA8 O_3 concentration of prefectural cities during a $PM_{2.5}$ and O_3 pollution episode.

On $PM_{2.5}$ side, the highest value of POIS occurred in Anhui and the highest value of PIIS occurred in coastal areas (Figure 7). It illustrated $PM_{2.5}$ 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.



(a)



(b)

Figure 7. (a)The POIS and PIIS of $PM_{2.5}$ 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_3 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_3 concentration in prefectural cities in the YRD during the O_3 pollution episode, 4/25 – 4/30 in 2018. The highest daily average concentration occurred in Hangzhou, about 281 μg/m^3. 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_3 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 focuse 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 persective, 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.

Acknowledgments, Samples, and Data

This work was funded by the Ministry of Science and Technology of China (2018YFC0213804). There are no conflicts of interest in this article. The authors would like to thank Dr. David Wong at the US EPA for his comments on this manuscript.

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