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ORIGINAL PAPER

Common source areas of air pollution vary with haze intensity in the Yangtze River Delta, China

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Abstract
Rapid development of China's industrialization and urbanization in the past decades has highly decreased air quality. For instance, the Yangtze River Delta, a major economic area in China, is incurring strong haze pollution, yet precise pollution sources are unknown. Here, we hypothesized that sources of haze pollution might be the same in nearby cities within the region. To test this hypothesis, we studied sources in four major cities, Hefei, Hangzhou, Nanjing, and Shanghai, during the strong haze period from November 28 to December 10, 2013. This period was divided into four periods according to air $PM_{2.5}$ concentrations (PM: particulate matter): slight haze, moderate haze, heavy haze, and severe haze periods. Common pollution source areas were identified for the first time by backward trajectories and concentration weighted trajectory maps of $PM_{2.5}$. Results show that all cities contain air masses transported from the northwestern and northeastern regions. Emissions came mainly from northern and central China during the moderate haze period and from adjacent provinces during the severe haze period. During the heavy haze period, common sources were mainly located in the Anhui province, while during the severe haze period, common sources were mainly located in the northeastern part of the Anhui province and the western part of the Jiangsu province. Overall, our findings show that areas of pollution sources vary with the intensity of haze pollution. Our mapping method should thus provide more precise information to control air pollution at the regional scale.

Keywords Yangtze River Delta · Common source area · Regional transport · Severe haze · $PM_{2.5}$

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Introduction

With the rapid development of economy and urbanization in China during the past three decades, ever-worsening air quality in the megacities has occurred. Haze, a type of weather condition that occurs when atmospheric visibility is less than 10 km with relative humidity less than 80% (Chan and Yao 2008; Long et al. 2014), is a threat to human health and climate change (Chen et al. 2012; Zhang et al. 2015a; Guo et al. 2014; Yu et al., 2014; Rosenfeld et al., 2019). Fine particulate matter with an aerodynamic diameter less than 2.5 µm ($PM_{2.5}$) has been identified as the leading contributor to regional haze in China (Yu et al. 2014; Chan and Yao 2008; Wang et al. 2015; Gao et al. 2015; Wu et al. 2016).

In order to guarantee good air quality during major international events in China, such as the 2008 Beijing Olympic Games, the 2014 Beijing APEC summit, and the 2016 Hangzhou G-20 summit, temporary stringent emission control measures were instituted for local and surrounding areas (Xing et al. 2011; Liu et al. 2016; Li et al. 2017).

These measures include temporary closure of factories and power plants, limitation of motor vehicles, and even reduction of residential activities (Liu et al. 2016; Li et al. 2017). Although these measures have achieved notable successes, they are accompanied with inevitable shortcomings. First, these measures are difficult to be implemented on a large scale and for an extended time (Xing et al. 2011; Liu et al. 2016; Li et al. 2017). In addition, the target areas for emission reductions are simply derived on the basis of administrative divisions or the limited experience of local government, rather than a clear understanding of regional haze formation mechanisms. Actually, there were not enough observation data with high spatial and temporal resolutions to support related research before 2013. Although a nationwide observation network was gradually established in China since 2013, too many attentions have been paid to haze formation in isolated cities instead of cities facing concurrent regional haze. Thus, in order to help local governments to make more precise policies on regional haze control, pinpointing potential sources for regional haze formation for the cities in a region and identifying the common source areas are a key need (Yu et al. 2018).

Hybrid receptor models have become widely used in detecting potential air pollution sources for the receptor cities in China, like Hangzhou (Yu et al. 2014), Beijing (Li et al. 2015c), Shanghai (Zhang et al. 2017), Zhengzhou (Wang et al. 2017a),Wuhan (Wang et al. 2017b), Lin'an (Zhang et al. 2017), and Tianjin (Guo et al. 2018). Potential source areas for a single city have been clearly identified in these studies. However, the common source areas for regional haze formation within a severely polluted region remain to be investigated.

The Yangtze River Delta (YRD) region is regarded as one of the most important economic centers in China with a population of 198 million (http://www.stats.gov.cn), contributing 24% Gross Domestic Product of the entire country. In the past decades, numerous haze episodes have occurred within this region. The annual average concentration of $PM_{2.5}$ in 2013 was 67 μg m^{-3} in this region, significantly exceeding the National Ambient Air Quality Standard in China (NAAQS, 35 μg m^{-3}). Numerous studies have been carried out on the formation mechanisms and source apportionments of haze pollution within the YRD region. These include the chemical composition, vertical distribution patterns, and regional transport of $PM_{2.5}$ (Yang et al. 2012; Peng et al. 2015; Zhang et al. 2015b, 2017; Qiao et al. 2016; Xiao et al. 2017; Li et al. 2018). For example, Du et al. (2017) reported that air masses originating from northern China brought massive fugitive dust to the YRD region in spring and increased the contribution of combustion emissions through long-range transport in winter. Li et al. (2015a) used the WRF-CMAQ model to quantify the contributions from regional transport and local emissions to $PM_{2.5}$ concentrations over the YRD region, which revealed that Anhui, southern Shandong, and northern Jiangsu contributed 50% of the $PM_{2.5}$ in the YRD region. Li et al. (2015b) applied comprehensive air quality model (CAMx) coupled with the particulate matter source apportionment technology (PSAT) method to study the source contributions to $PM_{2.5}$ at six receptors in the YRD region. However, all these studies have not revealed the common source areas of haze pollution in the YRD region.

Our hypothesis is that the potential source areas of haze pollution should be similar in nearby cities during a severe haze episode, but there is actually few knowledge on that. To confirm our hypothesis, the common source areas of haze pollution were identified in four major cities in the YRD region. Target four major cities are Hefei (capital of Anhui province), Hangzhou (capital of Zhejiang province), Nanjing (capital of Jiangsu province), and Shanghai, reflecting the current situation of the YRD region. With a hybrid receptor model, the potential $PM_{2.5}$ sources of these four cities during a severe haze episode in the winter of 2013 were identified. Common source areas of $PM_{2.5}$ for these four cities were accurately detected. This novel design provides a scientific and precise approach of regional haze control by allowing for pinpointing common source areas for formation of haze pollution. The method can also be further applied in controlling regional haze in other city clusters of China.

Experimental

Observational data

Hourly average concentrations of $PM_{2.5}$ from November 28 to December 10, 2013, in four cities (Hefei, Nanjing, Hangzhou, and Shanghai) were obtained from the National Environment Protection Web site (http://www.mee.gov.cn/). Four haze periods were grouped based on hourly $PM_{2.5}$ concentrations as follows: slight haze ($75 \leq PM_{2.5} < 115$ μg m^{-3}), moderate haze ($115 \leq PM_{2.5} < 150$ μg m^{-3}), heavy haze ($150 \leq PM_{2.5} < 250$ μg m^{-3}), and severe haze periods ($PM_{2.5} \geq 250$ μg m^{-3}).

Hybrid single-particle Lagrangian integrated trajectory (HYSPLIT) model

The HYSPLIT model from the National Oceanic and Atmospheric Administration (NOAA) was used to do the backward trajectory analyses and concentration weighted trajectory (CWT) analyses. The HYSPLIT model can be applied to study atmospheric pollution episodes or conduct climatological analyses with gridded meteorological data, using a hybrid calculation method including Eulerian and Lagrangian approaches (Draxler and Hess 1998).

To identify the transport pathways of $PM_{2.5}$, 48-h back trajectories were calculated at 100 m arrival height above the ground level (Wang et al. 2017a, b). The calculations were carried out every 3 h at starting times of 00:00, 03:00, 06:00, 09:00, 12:00, 15:00, 18:00 and 21:00 (UTC time) during the study period (Yan et al. 2015; Wang et al. 2017a, b). Meteorological data with a high spatial resolution of 0.5°×0.5° were obtained from the NOAA Web site (ftp://arlftp.arlhq.noaa.gov/pub/archives/gdas0p5).

A CWT method was also used to evaluate the contributions of potential sources to the haze formation in the receptor cities (Yan et al. 2015). The method calculates the CWT_{ij} values in the grid cell (*i*,*j*) as follows (Hsu et al. 2003; Yan et al. 2015):

$$CWT_{ij} = \frac{1}{\sum_{l=1}^{M} \tau_{ijl}} \sum_{l=1}^{M} C_l \tau_{ijl}$$

where l and M represent the index and the total number of the trajectories in the grid cell, respectively. C_l is the concentration monitored at the receptor site on the arrival of trajectory l. τ_{ijl} is the time that trajectory l spent in the grid cell (*i*,*j*). A high CWT_{ij} value indicates that the air masses traveling across the grid cell (*i*,*j*) have high potential contributions to pollutant concentrations at the receptor site (Yan et al. 2015). The size of a CWT grid cell was set to be 0.25° × 0.25° in this study (Yan et al. 2015; Wang et al. 2017a, b).

Calculation of common source areas for haze formation

The potential pollution source areas for haze formation in Hefei, Hangzhou, Nanjing, and Shanghai during different haze periods can be clearly identified on the basis of CWT analyses. To detect the common source areas for haze formation, Esri's ArcGIS software was used. For each haze period, the CWT maps of $PM_{2.5}$ for these four cities were overlapped using the ArcGIS software, in order to get the intersecting portions of the four CWT maps. The intersecting portions can show locations of potential pollution sources that contribute to haze formation in these four cities. Therefore, the intersecting portions are considered to be common source areas for haze formation in the region. During different haze periods, the common source areas of these four cities were calculated using this method.

Results and discussion

Statistical characteristics of haze episode and backward trajectory analyses

The Yangtze River Delta (YRD) region and target four cities, i.e., Hefei, Hangzhou, Nanjing, and Shanghai, are shown in Fig. 1a. An extremely severe haze episode occurred in the region in the winter 2013. During the entire study period (November 28–December 10, 2013), the maximum values of hourly $PM_{2.5}$ concentrations in Shanghai, Nanjing, Hangzhou, and Hefei were 604, 465, 465, and 470 μg m^{-3}, respectively (Fig. 1b). The highest daily average $PM_{2.5}$ concentrations in these four cities were all above 330 μg m^{-3}, exceeding the NAAQS, i.e., 75 μg m^{-3}, by around 4 times. As shown in Fig. 1b, the hourly $PM_{2.5}$ concentrations in these four cities increased sharply and decreased rapidly. From November 28 to December 1, the hourly concentrations in these four cities increased gradually and reached a relatively high level in the early morning of December 2 simultaneously. At that time, the $PM_{2.5}$ hourly concentration in Shanghai reached 297 μg m^{-3}, with values around 200 μg m^{-3} in the other three cities (Fig. 1b).

Before December 8, there was little rain within the region and the average wind speed was very low (1.65-2.11 m s^{-1}) (Li et al. 2015a), which was favorable for accumulation of air pollutants. The $PM_{2.5}$ hourly concentrations in Nanjing reached the peak (465 μg m^{-3}) rapidly on December 4, followed by Hefei (on December 5), Shanghai (on December 6), and Hangzhou (on December 7). With a strong wind speed (> 6 m s^{-1}) on December 9, the pollution episode ended gradually (Li et al. 2015a). The $PM_{2.5}$ hourly concentrations in Shanghai decreased quickly as soon as the peak value was reached on December 7 similar to those in the other three cities, showing a significant decrease on December 9. The sharp increases and decreases of $PM_{2.5}$ concentrations indicate that the haze occurrences in the region can be attributed to regional transport. It should be pointed out that these four cities might be affected by similar regional transport sources since the $PM_{2.5}$ hourly concentrations synchronously rose and fell as analyzed before.

Backward trajectories can reveal the movements of air masses. Based on the calculation of backward trajectories, different clusters were classified by the cluster analysis algorithm (Wang et al. 2017a). The backward trajectories and clusters are shown in Fig. 1c, with average $PM_{2.5}$ concentrations and percentages of trajectories for each trajectory cluster summarized in Table 1. Table 1 shows that air masses originating from northwest directions, i.e., Northwest-north, Northwest-west, and Northwest, had the

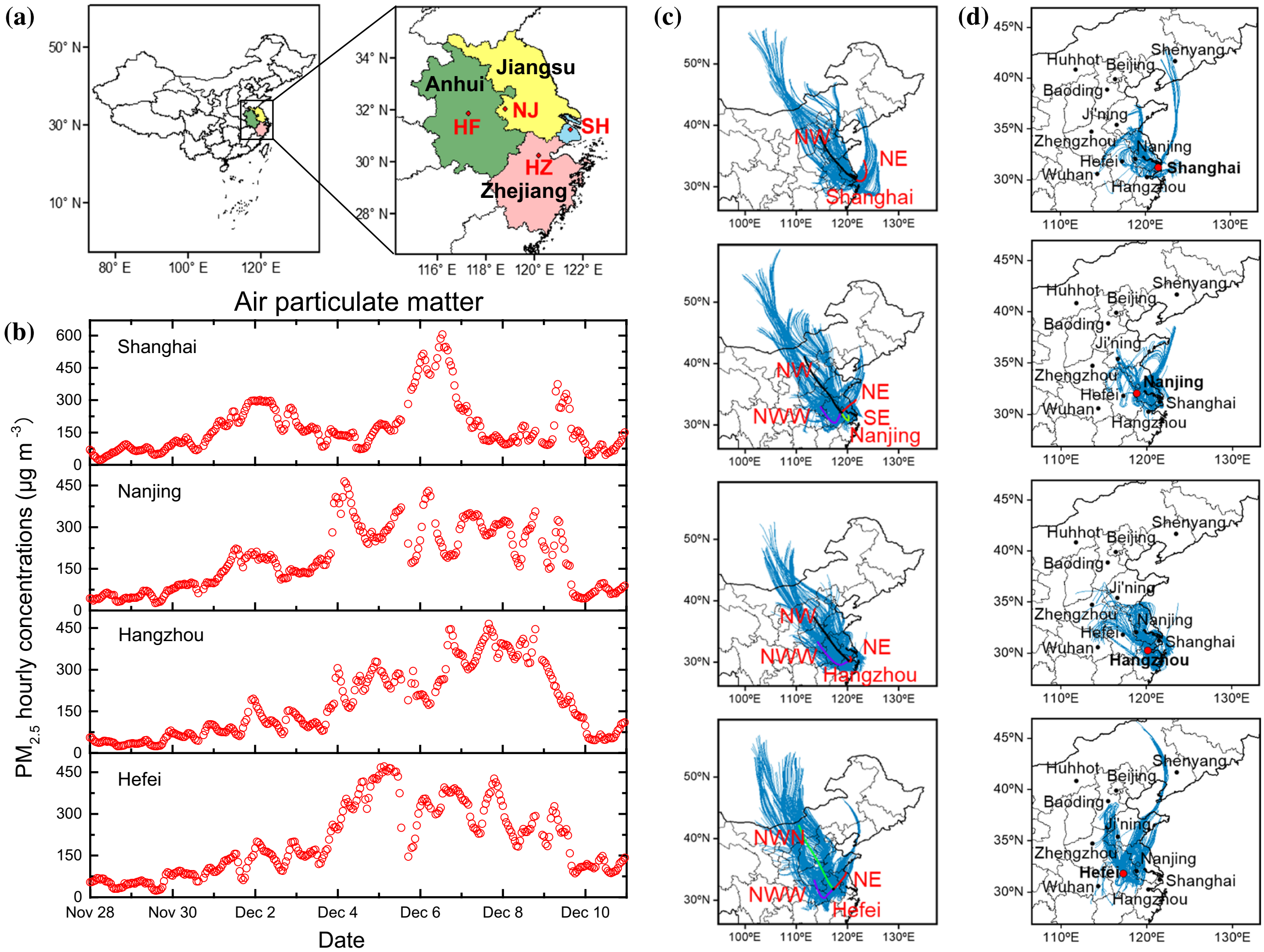

Fig. 1 **a** Map of China showing eastward the Yangtze River Delta (YRD) region with districts Jiangsu, Anhui, Shanghai (SH) and Zhejiang and locations of Shanghai (SH), Nanjing (NJ), Hangzhou (HZ), and Hefei (HF) cities. **b** Time series of $PM_{2.5}$ hourly concentrations in Shanghai, Nanjing, Hefei, and Hangzhou during 28 November–10 December, 2013. **c** 48-h air mass back trajectories and clusters during the entire study period (November 28–December 10, 2013). **d** 48-h back trajectories in severe haze period ($PM_{2.5} \geq 250$ μg m^{-3}). The red dots denote receptor cities. It can be seen that the maximum concentrations of $PM_{2.5}$ were extremely high (465-604 μg m^{-3}) in these four cities and the haze episode lasted for more than 10 days. The back trajectories and clusters indicate these four cities were all affected by northwestern and northeastern air masses. During heavy haze period, neighboring provinces had high contribution to pollution in the YRD region

largest portion, accounting for 63.7–69.4% in all trajectories of each city. For Hefei, Hangzhou, and Nanjing, the $PM_{2.5}$ concentrations of air masses originating from the Northwest directions (86.9-135.5 μg m^{-3}) were significantly lower than those from the northeast and southeast directions (266.5-319.7 μg m^{-3}). However, Shanghai where the average $PM_{2.5}$ concentrations from NW and NE direction were 184.3 and 153.6 μg m^{-3}, respectively, were somewhat different from those of the other three cities. This difference may be attributed to the factories in cities located in Northwestern Shanghai, such as Anhui and Jiangsu provinces, which emitted a lot of air pollutants (Zhang et al. 2015a). The backward trajectory results of these four cities are similar to those of previous studies (Zhang et al. 2015a, 2018; Shu et al. 2017; Zhu et al. 2018).

Trajectories with hourly $PM_{2.5}$ concentrations exceeding 250 μg m^{-3} were selected to represent the air mass transport pathways for the severe haze period (Fig. 1d). Results show that the region were mainly affected by air masses coming from neighboring provinces, such as Zhejiang, Jiangsu, Anhui, and Henan. In addition, some trajectories arriving at Hefei and Shanghai originated from Inner Mongolia and Liaoning provinces in northeast China by transporting across the Yellow Sea, as shown in Fig. 1d. These results indicate that the air masses originating from the northwestern region areas were relatively clean, while those from the northeastern and southeastern region areas were heavily polluted.

Table 1 Number of backward trajectories and average concentrations of $PM_{2.5}$ in different clusters in Hefei, Hangzhou, Nanjing, and Shanghai during the study period

Cluster	Number (percentage) of trajectories	Mean ± SD (μg m^{-3})	Number (percentage) of polluted trajectories*	Mean ± SD (μg m^{-3}) (polluted trajectories)
Hefei				
NW-N	363 (43.3%)	118.3 ± 83.4	235 (28.0%)	157.2 ± 79.5
NE	273 (32.5%)	319.7 ± 95.5	272 (41.1%)	320.6 ± 94.5
NW-W	203 (24.2%)	135.5 ± 52.1	174 (26.4%)	147.8 ± 45.6
Hangzhou				
NW-W	185 (20.8%)	86.9 ± 30.9	111 (12.5%)	104.5 ± 27.2
NE	272 (30.6%)	285.8 ± 116.2	266 (30.0%)	290.7 ± 112.6
NW	431 (48.5%)	125.7 ± 110	224 (25.2%)	204.2 ± 101.4
Nanjing				
NW	374 (47.7%)	121.9 ± 93.7	201 (25.6%)	184.4 ± 87.9
SE	80 (10.2%)	266.5 ± 66.5	80 (10.2%)	266.5 ± 66.5
NW-W	148 (18.9%)	129.2 ± 48.8	133 (17.0%)	136.6 ± 45.7
NE	182 (23.2%)	292.7 ± 76.6	182 (23.2%)	292.7 ± 76.6
Shanghai				
NW	428 (63.7%)	184.3 ± 139.3	315 (46.9%)	231.7 ± 133.4
NE	244 (36.3%)	153.6 ± 80.1	236 (35.1%)	156.7 ± 79.6

*Polluted trajectories: trajectories with hourly $PM_{2.5}$ concentrations higher than 75 μg m^{-3}

PM particulate matter

Source contributions from the concentration weighted trajectories

Concentration weighted trajectory (CWT) analyses provide information on the relative contributions of each potential source area to the high concentrations in the receptor areas (Zhang et al. 2015a). To locate the potential source areas that contributed to the high $PM_{2.5}$ concentrations in the region, CWT analyses were conducted. As shown in Fig. 2, for the entire study period, potential source areas for haze formation in Hefei were located in the western part of Jiangsu province, southwest of Shandong province, and some portions of Liaoning and Inner Mongolia, whereas those of Hangzhou were located in the northeast Zhejiang province, Shanghai, and Jiangsu provinces. Air masses originating from Liaoning province transported across the Yellow Sea could potentially carry considerable pollutants, resulting in $PM_{2.5}$ accumulations in Nanjing and Shanghai.

For the four haze periods, it was found that potential source areas became more concentrated as air quality deteriorated (Fig. 2). For Hefei, air masses from Inner Mongolia, northwestern Jiangsu and northeastern Anhui carried high concentrations of $PM_{2.5}$. In contrast with Hefei, long-range transport occurred only during clean periods in Hangzhou during the haze periods, while short-range transport from adjacent provinces played a leading role for $PM_{2.5}$. Both Nanjing and Shanghai were affected by air masses transported from the Yellow Sea.

Figure 2 shows that the potential source areas vary from one haze period to another. For example, in relatively less polluted periods, i.e., slight and moderate haze periods, air masses were transported long distance, originating from places as far as Mongolia and Inner Mongolia. However, during heavily polluted periods, i.e., heavy and severe haze periods, potential source areas are located in neighboring provinces, like Henan, Shandong, and Jiangxi provinces (Fig. 2). This difference may be attributed to different weather patterns. During heavily polluted periods, the region was trapped under stable weather conditions due to the significant downward motions of air masses, thereby hindering the long-range transport of air pollutants (Shu et al. 2017).

During the whole study period, sulfate and nitrate were the main components of $PM_{2.5}$, followed by ammonium, elemental carbon (EC), and organic carbon (OC), accounting, respectively, for about 30%, 24%, 18%, 12%, and 16% of $PM_{2.5}$ (An 2015). Industrial boilers and power plants contributed mainly to sulfate. Nitrate was mainly from industrial boilers, power plants, and transportation sources. Ammonium was mainly from agricultural emissions. Fugitive dust and transportation contributed mainly to EC, while industrial boilers and fugitive dust contributed mainly to OC. This implicated that emissions from industrial boilers, power plants, and transportation should be controlled to mitigate the pollution situation.

In summary, during the slight and moderate haze periods, long-distance airstreams contributed mostly to pollution in these four cities. By contrast, during heavy and severe haze periods, potential pollution sources were mainly located in the neighboring provinces, like eastern Henan, southwestern Shandong, Jiangsu, and northern Zhejiang.

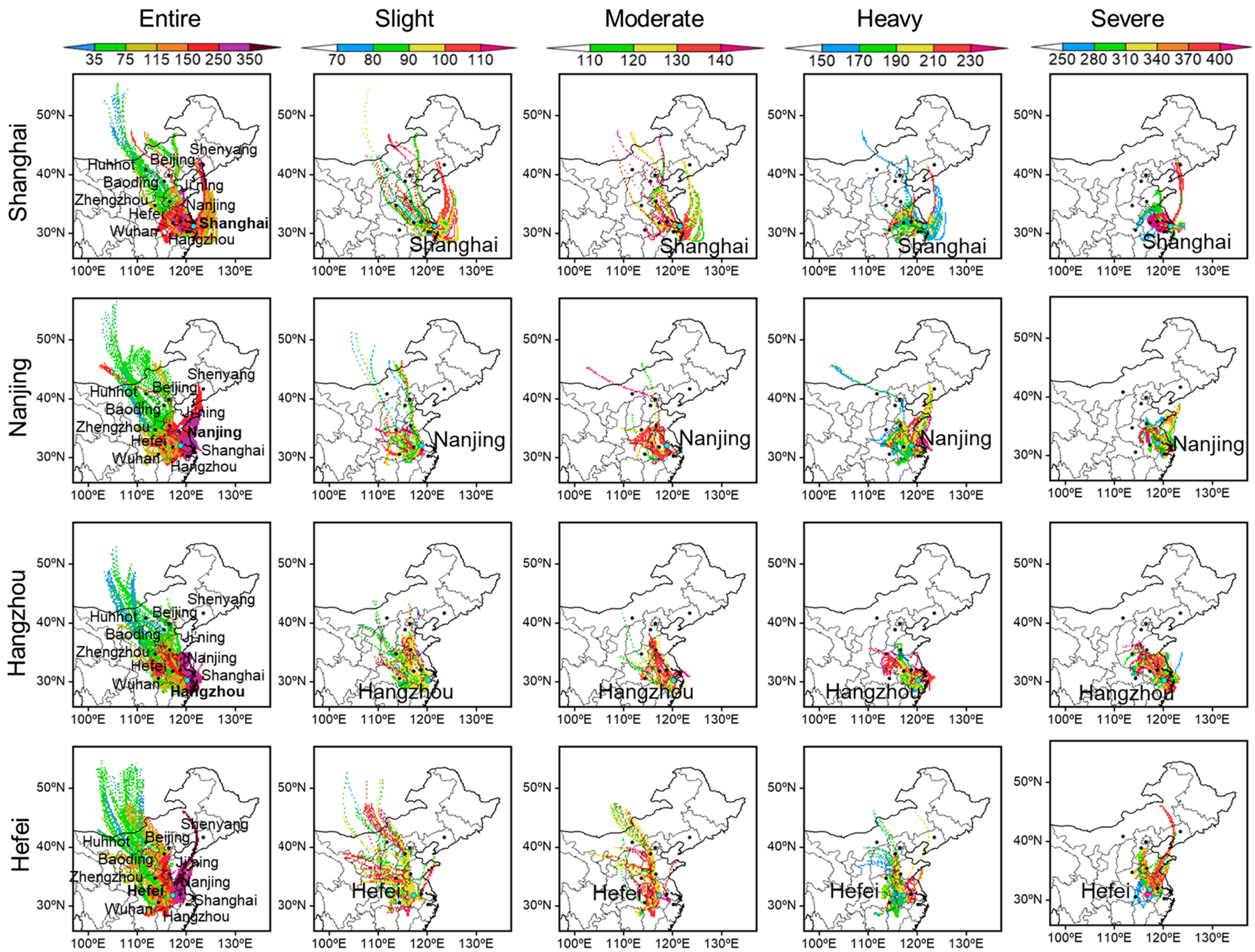

Fig. 2 Concentration weighted trajectory (CWT) maps of $PM_{2.5}$ (particulate matter with aerodynamic diameter less than 2.5 μm) for Hefei, Hangzhou, Nanjing, and Shanghai for the entire study period (November 28–December 10, 2013), slight haze ($75 \le PM_{2.5} < 115$ μg m^{-3}), moderate haze ($115 \le PM_{2.5} < 150$ μg m^{-3}), heavy haze ($150 \le PM_{2.5} < 250$ μg m^{-3}), and severe haze periods ($PM_{2.5} \ge 250$ μg m^{-3}). The light blue dots denote receptor cities. It shows that short-range transport form northwestern YRD region and long-range transport from northeastern YRD region have high contribution to local pollution. During the slight and moderate haze periods, the YRD region was affected by long-range transport pollutants. As air quality got worse, the potential pollution sources were mainly located in neighboring provinces

Common source areas of the haze formation in the region

Common source areas were determined for the formation of $PM_{2.5}$ in Hefei, Hangzhou, Nanjing, and Shanghai during the four haze periods (Fig. 3). It is clear that the locations of common source areas vary during the different periods. The common source areas of these four cities varied during the different haze periods, with the heavy and severe haze periods > slight haze period > moderate haze period. During the slight haze period, the common areas were mainly located in the central and western parts of Anhui province (such as Fuyang and Hefei), eastern Henan province, and southwestern Shandong province (Fig. 3a). The haze pollution in the region was mainly affected by long-range transport, especially emissions from Northwestern Delta regions. During the moderate haze period, common source areas were the smallest among the four haze periods, distributed sporadically over the surrounding areas of Zhengzhou, Fuyang, Hefei, Ji'ning, and Nanjing (Fig. 3b). This was partly because of the smallest number of backward trajectories during the moderate haze period. Pollution from outside the Delta region decreased, while pollution from local provinces increased. During the heavy haze period, common source areas were located mainly in Anhui province (Fig. 3c). Emissions from local provinces became dominated as haze pollution got worse. At the same time, long-range transport of $PM_{2.5}$ changed to short-range transport due to the slow wind speed and temperature inversion conditions (An 2015; Li et al. 2015a). During the severe haze period, the common

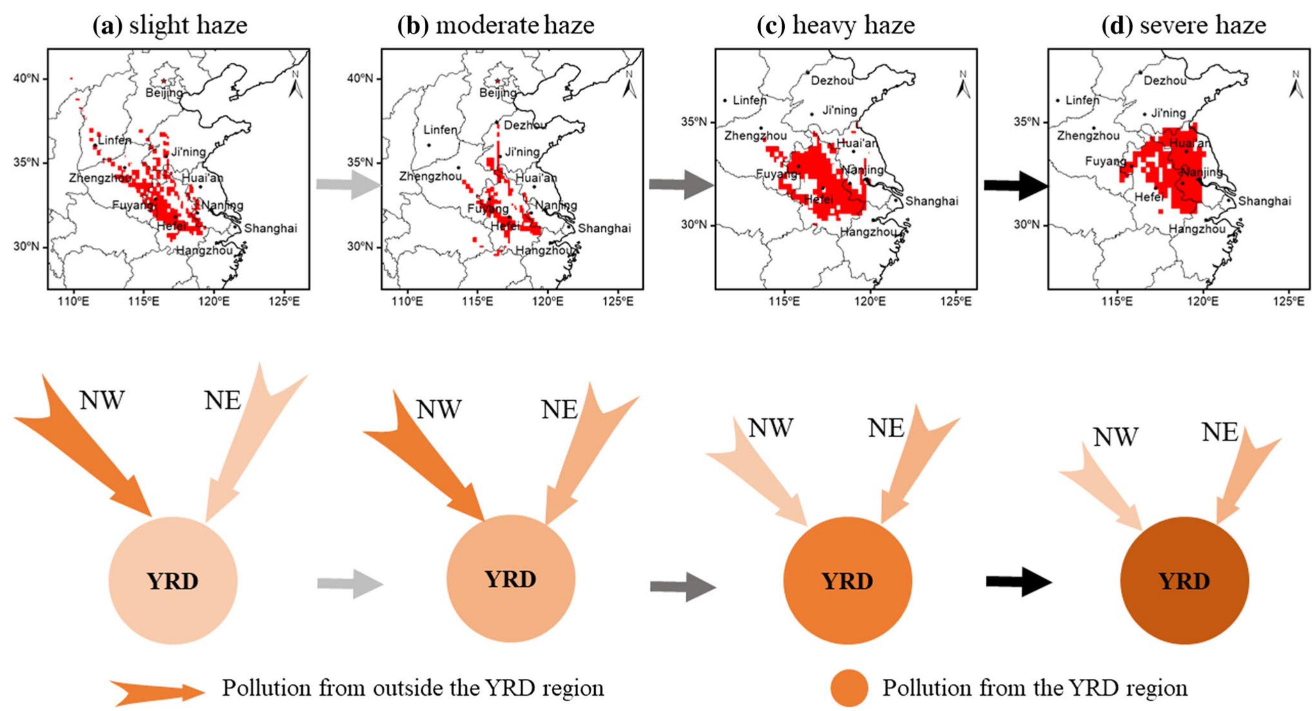

Fig. 3 Common source areas of $PM_{2.5}$ in Hefei, Hangzhou, Nanjing, and Shanghai during the slight (**a**), moderate (**b**), heavy (**c**), and severe (**d**) haze periods using Esri's ArcGIS software. The common source areas are different during different haze periods. During the heavy and severe haze periods, the common source areas are much larger than those during the slight and moderate haze periods. In the bottom animations, the brown arrows show the pollution from outside the Yangtze River Delta (YRD) region, while the brown circles show the pollution from the local YRD region. Longer arrows reflect longer transport distances of $PM_{2.5}$, while darker color reflects relatively greater contribution to the haze pollution of the YRD region. It is shown that emissions from local provinces in the YRD region become dominated as haze pollution gets severe

source areas were located in the northeastern part of Anhui province and the western part of Jiangsu province, such as Huaian and Nanjing, as shown in Fig. 3d. Note that during the heavy and severe haze periods, the common source areas were larger than those during slight and moderate haze periods (Fig. 3), mainly located in neighboring provinces, like Anhui and Jiangsu provinces. Relatively stable weather conditions in the region limited the long-range transport and diffusion of air pollutants during heavy and severe haze periods.

Joint efforts of adjacent areas are essential in controlling regional air pollution (Yan et al. 2015; Zhang et al. 2017; Cheng et al. 2018). As illustrated above, the common source areas for these four haze periods are all much smaller than the area of the entire region. Therefore, controlling emissions in common source areas will be easier and more economical, relative to controlling the entire region. Based on the pinpointed common source areas, local governments can focus on emission reductions in these areas and make more precise air pollution control policies. In addition, this method can be applied to understand the formation of regional air pollution in other city clusters and provides references for policy making. If combined with air quality forecast system, the potential common source origins of regional haze can be detected ahead of time. Then, the results will guide local governments to make scientific emission control policies in targeted regions before severe haze truly occurs (Yu et al. 2018). With the help of three-dimensional atmospheric chemical transport models, the effectiveness of different control strategies can also be quantified and thus guarantee precise decision made by the governments.

Conclusion

The Yangtze River Delta, one of the most important economic centers in China, has suffered from the occurrence of severe haze pollution in recent years. In this study, the HYSPLIT model was used to identify potential source areas for formation of haze pollution in Hefei, Nanjing, Hangzhou, and Shanghai within the YRD region during a prolonged haze episode in the winter of 2013. The 48-h backward trajectories show that these four cities have similar trajectory clusters, i.e., NE and NW. CWT maps of $PM_{2.5}$ for these four cities indicate that potential pollution sources for the regional haze formation differed during different haze periods. The YRD region was affected by long-range transport of pollution during the slight and moderate haze periods, while pollution sources were located mainly in adjacent provinces during the heavily haze periods. The common source areas were detected by using ArcGIS software. Results show that the spatial distributions of common source areas varied during different haze periods. The common source areas of these four cities varied during the different haze periods, with the heavy and severe haze periods > slight haze period > moderate haze period. During the slight and moderate haze periods, the common areas are mainly located in the central and western parts of Anhui province, such as Fuyang and Hefei, eastern Henan province, and southwestern Shandong province. During the severe haze period, the common source areas were located in the northeastern part of Anhui province and the western part of Jiangsu province such as Huaian and Nanjing. These results imply that the common source areas during each haze period were much smaller than those of the entire Yangtze River Delta region.

In summary, controlling emissions in the common source areas is a more precise way relative to traditional policies. Thus, in order to improve air quality in the region, it is necessary to implement strategies to control air pollution in common source areas. This will be easier and more economical, relative to controlling the entire region. The method used in this study could be further applied in haze pollution control in other regions of China.

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