Study on the Influence of Regional Transportation on PM_{2.5} Based on the RAMS-CMAQ Model in Weihai, a Typical Coastal City of Northern China

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ABSTRACT

Weihai, on the east coast of China, is the only city in Shandong Province that the air quality has reached the Chinese National Ambient Air Quality Class II Standard. However, heavy pollution events still occurred in winter and spring. In this study, the Integrated Source Apportionment Method (ISAM), a source analysis tool coupled with the Regional Atmospheric Modeling System and the Community Multiscale Air Quality modeling system (RAMS-CMAQ), was used to quantify the contribution of regional transport to PM_{2.5} and its main components in Weihai during February to March in 2018. The results showed that regional transport dominated PM_{2.5} pollution in Weihai, and the contributions of transport outside Weihai in February and March were 55.6–65.5% and 61.3–68.6%, respectively. Different components in PM_{2.5} showed the proportion of pollutants varies enormously. Secondary particles were mainly transported via long distances. The proportion of long-range transportation of nitrate had the highest value, and the contribution of regional transportation outside Weihai was 87.5–92.5% and 88.2–91.7%, followed by sulfate, which was 67.9–80.6% and 80.8–86.2%, and the proportion of ammonium salt is the lowest, contributed for 38.8–70.8% and 41.1–71.9% in February and March, respectively. Black carbon (BC), organic carbon (OC), and ions (ions here include chloride, sodium, magnesium, potassium, calcium, iron, aluminium, silicon, titanium, manganese, and other major nonspecific fine particles) were mainly influenced by short-range transportation and local emissions. The contributions of BC outside Weihai were 26.0–49.8% and 34.2–55.2%, the values for OC were 33.5–52.9% and 41.0–53.0%, those for ions were 37.8–49.9% and 44.1–54.4%, respectively. The trajectories indicate that PM_{2.5} pollution in Weihai is significantly affected by transport from Shandong Province, North China, Northeast China and other regions. Thus, the improvement in air quality in Weihai requires strengthening interregional joint pollution prevention.

Keywords: PM_{2.5}, CMAQ-ISAM, Regional transport, Coastal city

1 INTRODUCTION

The fine particulate matter (PM_{2.5}) in the atmosphere is both generated by primary particulate matter emissions and the transformation of gaseous precursors such as SO₂, NO_x, VOCs and NH₃ in primary emissions (Yang *et al.*, 2000; Kanakidou *et al.*, 2005; Hallquist *et al.*, 2009; Liu *et al.*, 2015). PM_{2.5} is not only enriched in a large number of toxic and harmful substances but also lives

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in the atmosphere for a long time. Moreover, $\text{PM}_{2.5}$ can be transported for a long distance through atmospheric circulation, which is harmful to human health (Harrison and Yin, 2000; Wang *et al.*, 2000; Poschl, 2005).

Heavy aerosol pollution has been frequently observed in North and central eastern China before 2014, with $\text{PM}_{2.5}$ as the major pollutant in most cities and regions (Wang *et al.*, 2014). At present, Researches related to $\text{PM}_{2.5}$ in China are mainly carried out in the Beijing-Tianjin-Hebei region (BTH) (Liu *et al.*, 2005; Han *et al.*, 2016; Xing *et al.*, 2018; Zhao *et al.*, 2019), the Yangtze River Delta (Li *et al.*, 2014; Wu *et al.*, 2018; Feng *et al.*, 2019), the Pearl River Delta (Fan *et al.*, 2015; Huang *et al.*, 2015), and the Chengyu region (Qiao *et al.*, 2019; Xu *et al.*, 2020). The aforementioned studies mainly focused on the emission reduction strategies, the effectiveness of precursor control, the further exploration of the nonlinear relationship between the emissions of different components of $\text{PM}_{2.5}$ and human health. In this study, the study range was chosen as Shandong Province, which borders the BTH region in North China. The BTH is also one of the regions that have highest pollutant emissions and worst air quality in China (Zhang *et al.*, 2019). To our knowledge, research carried out in Shandong Province mainly focus on the components, distribution characteristics, industry contribution and emission source identification of $\text{PM}_{2.5}$ in polluted cities (Yang and Christakos, 2015; Xiong *et al.*, 2016; Gao *et al.*, 2017; Yan *et al.*, 2017; Zhang *et al.*, 2018; Luo *et al.*, 2019; Yao *et al.*, 2019). However, studies on transport processes in coastal areas with low pollution levels are lacking. Weihai city is located in the easternmost part of Shandong Province, surrounded by the sea on three sides, and it has jurisdiction over 8 major economic areas, including Wendeng (WD), Rongcheng (RC), Rushan (RS), Huancui (HC), Lingang (LG), Gaoqu (GQ), Jingqu (JQ) and the coastal area (YH). Currently, it is the only city in Shandong Province whose annual mean values of all criteria pollutant are below the second-level standard of China's national environmental air quality standard (GB 3095-2012) (Li *et al.*, 2020).

In this study, a list of localized pollution sources was established by investigating the emissions of primary particulate matter and gaseous precursor pollutants. The RAMS-CMAQ weather-air quality model simulation system was established based on local terrain, land and sea conditions and meteorological data. In addition, the monitoring data of routine monitoring stations were combined to evaluate the simulation accuracy. The contribution of various components of $\text{PM}_{2.5}$ from local and surrounding areas to Weihai city were calculated through the model, which could provide a reference for the development of localized and efficient fine particulate control measures.

2 DATA AND METHODS

2.1 The RAMS-CMAQ Model System

The RAMS-CMAQ model system is used to simulate the transport, transformation and deposition of aerosols and their precursors. CMAQ includes chemical mechanisms such as gas-phase chemistry and gas-solid conversion, horizontal and vertical diffusion transport, liquid-phase chemistry and dry-wet settlement, physical and chemical processes such as nucleation, collision and growth of aerosols. The required initial concentration field, boundary conditions, photolysis rate of pollutants and other data need to be obtained by numerical modules such as the initial field diagnosis module, boundary processing module and photolysis rate calculation module (Byun and Ching, 1999). Currently, the most widely used weather modules in CMAQ are MM5 (fifth-generation Penn State/NCAR Mesoscale Model) or WRF (Weather Research and Forecasting). Since most atmospheric pollutants, heat, water vapor and their fluxes come from the bottom of the atmospheric boundary layer, and the vertical shear of these quantities is very obvious in the atmospheric boundary layer, showing multi-extremum or multi-centre distributions, the region with the most drastic changes in atmospheric stability is also in the bottom of the atmospheric boundary layer (Zhang, 2005). RAMS is adopted to provide a three-dimensional meteorological driving field for CMAQ. In the outer domain, we use the latest China multiresolution emission inventory (MEIC) for the 2016 baseline (He, 2012). The monthly emission list includes 8 types of pollutants with a spatial resolution of $0.25^\circ \times 0.25^\circ$. The inner domain uses the local source emission inventory of Weihai. The local emissions inventory of Weihai city in 2018 was established according to the "Technical Guidelines for the Compilation of Primary Source Emissions Inventory of Atmospheric Fine Particulate Matter (Trial)" in China. The MEIC was established based on the emissions in 2016.

The local emissions inventory of Weihai was established based on the emissions in 2017. Due to the comprehensive ultra-low transformation of the thermal power plant in Weihai in 2017, the ultra-low emission transformation or replacement of coal-fired boilers with more than 10 tons has been completed. Coal-fired boilers of 10 tons or less have been eliminated. "Scattered polluted" enterprises have been cleaned up or treated. Besides, the local emissions inventory of Weihai investigated that marine fuel and heavy oil accounts for a relatively large proportion, which led to an increase of nitrogen oxide particles from non-road mobile sources. Fig. 1 shows that except for NO_x , BC and NH_3 , the other pollutants emissions in the MEIC are larger than that in the local emissions inventory of Weihai. The local emission source is divided into 10 categories, including dining lampblack, waste disposal, storage and transportation, biomass combustion, dust, agricultural, solvent, mobile, process and fossil fuel combustion. The resolution of local emission sources is $1 \text{ km} \times 1 \text{ km}$. The emission source in MEIC has five classes (industry, electric power, transportation, residents and agriculture ammonia). Monthly biomass source emissions include forest fires with a spatial resolution of $0.5^\circ \times 0.5^\circ$, savanna fires and slash-and-burn agriculture inventories from the global fire emission database (GFED v4) (van der Werf *et al.*, 2010). The Asian regional emission inventory (<http://www.jamstec.go.jp/frsgc/researchd4/emission.htm>) provides nitrogen oxide and ammonia emissions from agriculture and NO_x emissions from the world of aircraft and lightning discharge atmospheric research database (Olivier *et al.*, 1994).

Due to the wide sources of air pollutants, clarifying the sources of urban pollutants and their proportions has become a very important subject for environmental management and scientific decision-making. The formation mechanism of secondary pollutants is more complex than that of primary pollutants: the complex physical (mixing) and chemical processes involved in transport (gas and liquid phase, especially heterogeneous phase) increase the difficulty of accurately quantifying the effects on transboundary transport. Therefore, it is necessary to identify and quantify the major pollution sources of secondary pollutants, reveal its formation mechanism of air pollution in urban agglomeration. The results can provide evidence on regional air quality prediction, regional air pollution control and prevention.

The comprehensive analytical method ISAM is based on the developed CMAQ. By introducing the reactivity tracer, it realizes the online source resolution of $\text{PM}_{2.5}$ and O_3 . In this study, the ISAM module is used to track $\text{PM}_{2.5}$ and its precursors in different emission areas (Chen *et al.*, 2017; Han *et al.*, 2018; Chang *et al.*, 2019). It is implemented based on tagged species source analysis (TSSA) (Wang *et al.*, 2009), which is a method of species labelling. ISAM can track the contribution of primary and secondary inorganic $\text{PM}_{2.5}$ from initial conditions, boundary conditions and labelled emission sectors and regions (Kwok *et al.*, 2013). ISAM supports 6 kinds of $\text{PM}_{2.5}$ labels, including elemental carbon (EC), primary organic carbon (POC), sulfate (SULFATE), ammonium nitrate (NITRATE), ammonium (AMMONIUM), and $\text{PM}_{2.5}$ _ions (ions) tags (Li *et al.*, 2019). ions here include chloride, sodium, magnesium, potassium, calcium, iron, aluminium, silicon, titanium, manganese, and other major nonspecific fine particles. After each chemical or physical process, the tagged species are

Fig. 1. Comparison of the emissions between the MEIC and the local emissions inventory of Weihai.

updated by assigning changes to the corresponding species. Compared to the Comprehensive Air Quality Model with Extensions (CAMx) and Particulate Source Apportionment Technology (PSAT), CMAQ and ISAM also considers blown dust and metal ions, as well as detailed classification of EC and ions (Li *et al.*, 2019). In addition, the module improves the computing efficiency.

To evaluate the influence of mesoscale weather processes, complex surfaces on local atmospheric circulation, atmospheric boundary layer structure and air mass transport on pollutant concentration, the simulated regions were selected on behalf of Weihai as shown in Fig. 2. The simulation region of the coarse grid of the model was with a horizontal resolution of $16 \text{ km} \times 16 \text{ km}$ and a grid point of 94×90 . The fine grid of the model covered the whole areas of Weihai and other neighbouring cities, with a horizontal resolution of $1.5 \times 1.5 \text{ km}$ and a grid point of 101×80 . The grid across the boundary is assigned to the districts and counties with most of the area. In the vertical direction, the height of the top of the model is approximately 15 km, and it is divided into 15 layers, but the thickness of each layer is different. The resolution of the near-ground layer is the highest (the height of the first whole layer is approximately 50 m), and it gradually decreases upward. Nearly half of the 15 layers lie within the atmospheric boundary layer to distinguish the vertical structure of the atmospheric boundary layer.

Model evaluation formulas are listed as Eq. (1) and Eq. (2) (Boylan and Russell, 2006):

FB (fractional bias):

$$FB = 100\% \times \frac{2}{N} \sum_{i=1}^N \frac{(M_i - O_i)}{(M_i + O_i)} \quad (1)$$

FE (fraction error):

$$FE = 100\% \times \frac{2}{N} \sum_{i=1}^N \frac{|M_i - O_i|}{(M_i + O_i)} \quad (2)$$

where FB is the fractional bias, FE is the fraction error, N is the number of simulated and observed values paired with time, and M_i and O_i are the simulated and observed values paired with time.

Acceptable accuracy of mode application: $FE \leq 75\%$ & $|FB| \leq 60\%$.

Table 1 shows the comparison between the $PM_{2.5}$ simulation and observation data. Except for Zhangcun site ($FE = 55.9\%$; $FB = 93.8\%$), all other sampling sites meet the requirements of simulation effect evaluation ($FE \leq 75\%$ & $|FB| \leq 60\%$). The higher FB of Zhangcun station is associated with the higher systematic systematic bias (Fig. 3). Generally, the simulation results on pollutants are slightly higher than observed concentration. However, they have about the same standard deviation and correlation coefficient. Since the dispersion degree of the simulated

Fig. 2. (a) The study area of the RAMS-CMAQ model. The simulation area is shown in the red box. (b) The administrative divisions of the study city of Weihai including eight regions and seven observation sites are shown in the figure.

Table 1. Comparison of PM_{2.5} simulation and measurement.

Station	N	M	O	σM	σO	r	FE (%)	FB (%)
HuaXia	765	25.6	27.4	18.6	14.4	0.64	46.3	-0.4
LanTian	750	27.6	26.7	18.2	12.9	0.52	44.2	15.2
MuGong	761	21.0	21.8	16.0	15.5	0.72	49.3	22.8
ShanDa	758	25.4	23.6	16.3	14.0	0.55	46.7	29.1
ShiZhan	763	24.9	26.2	15.9	13.1	0.56	44.4	4.8
WD	765	24.7	24.8	20.3	17.3	0.40	57.7	24.0
ZhangCun	723	23.0	19.4	15.8	14.1	0.61	55.9	93.8

N is the number of simulated and observed values paired with time; *M_i* and *O_i* are the simulated and observed values paired with time, and *M* and *O* are the mean values of the simulated and observed values, respectively. σM and σO are the standard variance evaluation parameters of the simulated and observed values. *r* is the correlation coefficient between simulated and observed values. *FB* is fractional bias, *FE* is fraction error.

value is slightly higher, the standard deviation of the simulated value is slightly higher than the observed value. Overall, the PM_{2.5} concentration simulated by CMAQ well fitted with the observed value in 2018 (Fig. 3).

2.2 Cluster Analysis of Back-trajectory Airmass

The Hybrid Single-Particle Lagrangian Integrated Trajectory Model (HYSPLIT) has been widely used to determine the individual backward trajectory of each air mass transport for tracing pollutant sources (Draxler and Hess, 1998). The Meteorological data was from 1° × 1° global data assimilation system (GDAS) of the National Centers for Environmental Prediction (NCEP). The 24-h back trajectories at 500 m above ground level (AGL) in Weihai (122.07°E, 37.22°N) were calculated in February and March 2018 with 24 trajectories approaching Weihai at 00:00–23:00.

Cluster analysis was performed to obtain the average trajectory that can represent multiple trajectories (Pu *et al.*, 2015). In this study, the euclidean distance between two reverse trajectories was used (Fang *et al.*, 2018). Cluster analysis is shown in Eq. (3):

$$\begin{aligned} D &= \sqrt{\sum_{j=0}^t d_j^2} \\ SPVAR &= \sum_{i=1}^x \sum_{j=0}^t D_{ij}^2 \\ TSV &= \sum SPVAR \end{aligned} \quad (3)$$

where *i* is the number of the trajectories, *j* is the number of passing points, *t* is the movement time of the airflow, *d_j* is the distance between the *j*th point of the two trajectories, *x* is the number of trajectories in the cluster, *D* is the distance between the trajectories, so *D_{ij}* represents the distance between the *j*th passing point in the *i*th trajectory and the corresponding point on the average trajectory, *SPVAR* is the space variation of each group of trajectories, and *TSV* is the total space variation. Stepwise cluster analysis method can divide the adjacent points in a large number of statistical samples into the same category, and then select similar trajectories to class. The more clusters are classified, the closer they are to the real situation, and the smaller the error of the results (Meng *et al.*, 2020). In this study, the cluster number is set as 8.

3 RESULTS AND DISCUSSION

3.1 Spatial Distribution and Regional Transport of PM_{2.5} in the 8 Districts and Counties in Weihai

RAMS-CMAQ was used to simulate the spatial distribution of PM_{2.5} and the concentration of each component in Weihai (Fig. 4). The average mass concentration of PM_{2.5} in Weihai in February and March 2018 was higher in the western part of the urban area (far from the coastal areas) in

Fig. 3. Comparison of $PM_{2.5}$ concentration between model simulation and monitoring data in the districts and counties of Weihai.

February, and the concentration gradually decreased from west to east. The concentration range was between 40 and 60 $\mu\text{g m}^{-3}$ in the RS and WD areas and the lowest concentration range was between 25 and 40 $\mu\text{g m}^{-3}$ in the coastal area of RC in the east. The concentration in March was significantly higher than that in February, and the concentration range in the western part of the city was 50–75 $\mu\text{g m}^{-3}$, with the highest concentration in RS. The concentration range of the RC

Fig. 4. The mass contribution ($\mu\text{g m}^{-3}$) of emissions in Weihai, Shandong Province, China, to PM_{2.5}, BC (black carbon), OC (organic carbon), and ions during February (Feb) and March (Mar) 2018.

coastal area with the lowest concentration was $30\text{--}55\ \mu\text{g m}^{-3}$. The average wind speed of Weihai in February and March was respectively $3.49\ \text{m s}^{-1}$ and $4.33\ \text{m s}^{-1}$ in 2018. The higher concentration in March compared to February is due to the dust caused by higher wind speeds in March. Moreover, the suspension of production in February for the Lunar New Year holiday may respond to the lower concentration.

In February, the BC concentration gradually decreased from west to east and from the coast to the interior, with the highest concentration in RS, which was above $2.2\ \mu\text{g m}^{-3}$ and above $3.0\ \mu\text{g m}^{-3}$ in some areas. In March, the concentration in the YH was the highest, and the concentration distribution was similar to that in February, and the increase trend was observed from west to east and from the coast to the interior. The concentration on the eastern coast increased significantly, with a concentration range of $3.0\text{--}3.6\ \mu\text{g m}^{-3}$ and a relatively small contribution to other regions, with a concentration range of $1.8\text{--}3\ \mu\text{g m}^{-3}$. The BC concentration rising in YH was related to the increase in fishing boats, passenger and cargo ships after the Spring Festival in March. In February and March, the OC concentration showed a decreasing trend from west to east, while in February, it ranged from 4.0 to $9.0\ \mu\text{g m}^{-3}$ in the west, and in March, it ranged from 7.5 to $9.5\ \mu\text{g m}^{-3}$ in the west.

In February, the concentration of ions in the northwest of RS and the central area of WD was slightly higher than other regions, above $12\ \mu\text{g m}^{-3}$. In most areas, the concentration ranged from 6 to $12\ \mu\text{g m}^{-3}$. The concentration was the lowest in the eastern YH, below $6\ \mu\text{g m}^{-3}$. In March, the concentration in the central part of RS and the central and southern parts of WD increased significantly, with the concentration above $14\ \mu\text{g m}^{-3}$ and the range significantly expanded, while the concentration in the southern parts of JQ and RC ranged from 10 to $14\ \mu\text{g m}^{-3}$.

Fig. 5 shows the transport contributions of primary component of $\text{PM}_{2.5}$ in the 8 districts and counties of Weihai in February and March 2018. In February, pollutant emissions from WD, RC, RS and JQ made a significant contribution to the monthly average concentration of local $\text{PM}_{2.5}$, which reached above $14\ \mu\text{g m}^{-3}$. Pollutant emissions from other regions contributed insignificant part to the monthly average concentration of local $\text{PM}_{2.5}$, which was below $7\ \mu\text{g m}^{-3}$. In February, the prevailing wind direction was northwest and the wind speed is relatively high. The pollutants discharged by each district and county were obviously transported downwind. In March, the pollutant emissions in all regions of Weihai contributed to RS, WD, LG, HC, RC, etc., with a relatively low and in the range of $3\text{--}15\ \mu\text{g m}^{-3}$. Compared with February, $\text{PM}_{2.5}$ pollution was widely distributed and aggravated in March.

Fig. 5. Contribution of transport of $\text{PM}_{2.5}$, black carbon (BC), organic carbon (OC), and ions in Weihai in February (Feb) and March (Mar) 2018. OTHR: contribution of emission source short-range transportation in simulated nested areas except Weihai; BCON: influence of boundary conditions to characterize long-range transport outside the nested area; ICON: influence of initial conditions.

Fig. 5. (continued).

In March, the $PM_{2.5}$ concentration in the western region was high, and the local contribution was relatively small, accounting for 30%. In the eastern region, the concentration was low, and the local contribution was relatively large, approximately 35%. In February and March, the regional transport outside Weihai accounted for 55.6–65.5% and 61.3–68.6% of the $PM_{2.5}$ contribution in the whole region, respectively. Particularly, the proportion of contributions outside the region increased significantly in March. The average contribution rate to each district and county was above 64.0% (March), followed by the contribution of local pollutant emissions and coastal pollutant emissions to each district and county, with a smaller contribution among districts and counties, indicating that the $PM_{2.5}$ pollution in Weihai is dominated by transport outside the region. In February and March, the local contribution was the highest in JQ (February), accounting for 42.4%, and the lowest in RS (March), accounting for only 30.2%. This result was related to the geographical location of the two regions. RS Mountain is located in the west of Weihai, while the prevailing northwest wind in winter weakens the contribution of the region. JQ is located in the hinterland of Weihai, where the contribution ratio was relatively strong. Among them, the contributions of WD, RC, RS, JQ and the YH played a leading role in intraregional transportation. WD had the largest influence on intraregional contributions and dominates the contributions of HC, LG, GQ and JQ. It is worth noting that WD contributes the largest part of the transportation to GQ in the region, which was higher than the local contribution of GQ. The transportation of WD to LG was also comparable to the local contribution of LG.

3.2 Transport Contribution of Primary Particulate Matter Components

To evaluate the contribution of specific emission sources to the whole region, the transport matrices of various components in $PM_{2.5}$, including BC, OC, ions, sulfate, nitrate and ammonium salt were calculated. Fig. 5 shows the transport contribution matrices of primary particles, including BC, OC and ions.

Fig. 5 showed that the total contribution of local pollutant emissions to the average monthly BC concentration in Weihai in February and March was relatively large, and the contribution of local sources in YH can reach 65–75%. In the eastern region, the contribution from local sources was more than 50%, while in the western region, the contribution from local sources was less than 40%.

In February and March, the total contribution of local pollutant emissions to the monthly average OC concentration in Weihai was less than that of BC, and the contribution ratio of local emissions in the eastern part of Weihai to OC was higher than that in the western part, ranging from 45 to 65%, while the contribution of local sources in the western part was less than 50%. The local contribution ratio of BC and OC in Weihai was 10%–20% higher than the total $PM_{2.5}$. Except for RS, ship emissions in YH dominated the contribution of BC, accounting for more than 20%–50%.

Compared with BC and OC, ions are more susceptible to the influence of outside transport, and the local contribution ratio was relatively low. The local pollutant emissions in Weihai contributed more to RS, WD, HC, JQ and GQ, reaching more than $6 \mu g m^{-3}$ in March, but less to other areas. Also all districts and counties in Weihai contributed a high concentration of pollutants to RS and part of WD, with a concentration between 12 – $20 \mu g m^{-3}$, which was mainly related to the local emissions and geographical location of the two regions. The concentrations of RC, HC and other parts of the region contributed less, with a concentration range of 6 – $11 \mu g m^{-3}$. In February, local pollutant emissions contributed approximately 45–60% to the ions in most areas in the eastern part of Weihai, while the local contribution was relatively low in parts of RS in the western part of Weihai. In March, the contribution rate of local emissions to the whole region of Weihai was 45–55%.

3.3 Regional Transport of Secondary Particulate Matter Components

Different from primary particles, the transport of secondary particles includes both secondary fine particles and their gaseous precursors (He *et al.*, 2013). Considering the differences in emission source characteristics, formation mechanism and life cycle, different secondary particles show different transport characteristics. Fig. 6 shows the transport contribution matrix of nitrate (NO_3^-), sulfate (SO_4^{2-}) and ammonia (NH_4^+) in Weihai. The results indicated that nitrate contributed the largest part of long-range transport $PM_{2.5}$, which was identified as the strongest transport

Fig. 6. Contribution of transport of nitrate (NO_3^-), sulfate (SO_4^{2-}), and ammonia (NH_4^+) in Weihai in February and March 2018. Here A stands for aerosol, and ANO_3 , ASO_4 and ANH_4 are total mass concentration of i-mode (Aitken) and j-mode (accumulation) of nitrate, sulfate and ammonia aerosols.

capacity of all PM_{2.5} components in this study. The contribution from outside the region exceeded 87.5%. The YH contributed the most part of the transport within the region, and the mobile source emissions of the marine industry dominated the localization contribution, with no significant change between February and March.

Fig. 7 shows the distribution of nitrate, sulfate and ammonium salt mass concentrations in Weihai in February and March, 2018. The concentration of nitrate in the west of Weihai was higher, while the concentration in the east was lower. The local contribution of nitrate in Weihai in February was less than 20%. In March, the contribution from local sources was further reduced to approximately 10%, indicating that the nitrate in Weihai mainly came from transport outside the region, and the local contribution was mainly from the YH, contributing 4.5–9.4%.

Different from nitrate, the sulfate transport matrix was similar to the total PM_{2.5}. Apart from the higher contribution of the YH, the transport contribution between districts and counties in the region was comparable, with a high concentration in the western and a low concentration in eastern region; The contribution rate of the local source of sulfate in the whole region of Weihai was between 20% and 30%. In March, the sulfate concentration in Weihai decreased from west to east, and the overall concentration increased significantly compared with that in February.

Fig. 7. Mass concentrations of nitrate, sulfate and ammonia in Weihai in February and March 2018. A refers to aerosol, and ANO3, ASO4 and ANH4 are total mass concentration of i-mode (Aitken) and j-mode (accumulation) of nitrate, sulfate and ammonia aerosols.

Fig. 8. Cluster of air mass backward trajectory of Weihai in (a) February and (b) March, 2018.

The contribution of local sources to the sulfate concentration in Weihai further decreased to less than 20%, while the contribution of off-site transport in February and March was 67.9–80.6% and 88.2–91.7%, respectively, indicating that the main source of sulfate in Weihai in winter and spring was regional transport from outside.

In a suitable environment, the formation of ammonium salt usually takes a few hours, the ammonium salt itself is unstable (Behera and Sharma, 2011), and its life cycle is shorter than nitrate and sulfate. Fig. 6 showed the contribution of ammonium salt was dominated locally. In March, the concentrations in the western and central regions were high, while the concentrations in the coastal and surrounding areas were low. In addition, the local contribution in the western region was more than 50%, mainly because the western region of Weihai had more cultivated land and more ammonium salt emitted from natural sources. Thus, the local contribution accounts for a relative large proportion, and the local contribution in the coastal region was 35–50%. The local contribution of the WD, RC and RS regions was comparable to that of regional transport outside Weihai.

In winter and spring, transport outside the region contributed 40–60% to $PM_{2.5}$ in Shandong Province, including 70–80% of nitrate, 40–45% of sulfate, and 30–35% of ammonium salt (Li *et al.*, 2019). The external contribution of $PM_{2.5}$ in Beijing, which is also located in North China, was 31–38% (Chang *et al.*, 2019). The proportion of $PM_{2.5}$ transported outside the Chengdu area in Southwest China could reach 42%, while that of Chongqing, another capital city, was only 30% (Qiao *et al.*, 2019). This result indicated that the $PM_{2.5}$ pollution in Weihai had very obvious characteristics of external transport, especially for nitrate and sulfate. Weihai is located at the easternmost end of the Shandong Peninsula and is directly affected by the westerly belt in the Northern Hemisphere. Fig. 8 showed that northwest and northeast winds prevailed in winter and spring, which is conducive to the transregional of pollutants, the contributions from North China and Northeast China in February and March accounted for 68.16% and 57.23% of the external transport in Weihai, and the contributions from Shandong province and other regions accounted for 23.51% and 26.39%, respectively. The contribution of residential coal burning and biomass burning to winter haze formation in northeast China is significantly larger than that in the North China Plain (Zhang *et al.*, 2020).

4 CONCLUSIONS

In this study, ISAM, a source analysis tool coupled with the RAMS-CMAQ model, was used to simulate and quantify the contribution of regional transport of $PM_{2.5}$ and the main components in Weihai during February and March, 2018. Regional transport dominated the contribution of

total PM_{2.5} pollution in Weihai. The contributions of transport outside Weihai in February and March were 55.6–65.5% and 61.3–68.6%, respectively. Different components of PM_{2.5} have different transport ratios. Among the primary particulate matter, BC, OC and ions were mainly affected by short-range transport and local contribution. Among the contributions of ammonium salt, the local contributions in the WD, RC and RS regions were comparable to those outside Weihai, and the contribution from transportation outside Weihai dominated in other districts and counties. There were significant differences in the transport capacity of various components in PM_{2.5}, with the highest transport capacity of nitrate, followed by sulfate. OC, BC, and ions where local contributions dominated, which indicated that PM_{2.5} pollution in Weihai was significantly affected by transport in Shandong Province, North China, Northeast China and other regions, and the improvement of air quality in Weihai requires strengthening interregional joint prevention and control.

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