Spatial Distribution and Multiscale Transport Characteristics of PM_{2.5} in China

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ABSTRACT

The Weather Research and Forecasting (WRF) model coupled with the Community Multiscale Air Quality (CMAQ) model was used to simulate the temporal and spatial distribution and the multiscale transport patterns of PM_{2.5} in 2 large regions (“NORTH” and “SOUTH”), 7 small regions (“North,” “Northeast,” “East,” “Center,” “South,” “Southwest,” and “Northwest”) and 31 provinces in China during January and July of 2015. The simulated PM_{2.5} concentrations were compared with the observed values to assess the accuracy of WRF-CMAQ. During January and July, local emissions contributed the majority of the PM_{2.5}, with their percentage exceeding 60% in 7 of the regions. External transport was the dominant source of PM_{2.5} in some of the provinces, such as Shanghai and Qinghai, whereas local emission was the main contributor in others, such as Hebei and Xinjiang. We also identified the primary PM_{2.5} source in each region, the results of which indicated distinctly different regional transport patterns among the provinces and regions of China.

Keywords: Particulate matter; Regional transport; WRF-CMAQ model; Multiscale regions.

INTRODUCTION

With the development of urbanization and industrialization in China, smog has become the focus of environmental and social issues. It has serious impacts on human health, visibility and climate change. Fine particulate matter (PM_{2.5}) is the main cause of haze formation, and consists of primary particles and secondary particles (Hu *et al.*, 2015). Primary particles can damage the respiratory system, and increase morbidity and mortality. Secondary particles have an impact on the climate by destroying the balance of Earth’s radiation. Natural and anthropogenic emissions are the main sources of PM_{2.5}, with the latter contributing the most, including incomplete combustion of fossil fuels, transport emissions, agricultural activities and domestic emissions (Cheng *et al.*, 2014). The chemical formation and transformation of PM_{2.5} is different due to different sources and chemical compositions (Zhang *et al.*, 2014).

According to the announcement of the World Health Organization (WHO) in 2015, there are 6 cities of China among the 30 most polluted cities in the world. Among the 211 Chinese cities recorded by WHO, there are 162 cities whose annual concentration of PM_{2.5} exceed the standard value (35 µg m⁻³). Therefore, attention should be paid to air quality in China. There are 4 economically developed

and heavily polluted regions in China, which are the Beijing-Tianjin-Hebei (BTH) region, the Yangtze River Delta (YRD) region, the Pearl River Delta (PRD) region and the Sichuan-Chongqing Economic Zone. With different kinds of topography and meteorological conditions in the 4 economic zones, PM_{2.5} pollution has various spatial and temporal distribution characteristics and transport interactions.

Severe regional air pollution frequently occurs in Beijing. The secondary particles are mostly contributed by regional transport mainly from Hebei Province and Tianjin Municipality (He *et al.*, 2017). Hebei is one of the most polluted provinces in China, characterized by extremely high concentrations of PM_{2.5} in some of its cities which are prone to transport northwards to Beijing (Wang *et al.*, 2015). Furthermore, the concentrations of PM_{2.5} in the YRD region on hazy days are 1.6–2.4-fold higher than the values on clear days (Wang *et al.*, 2016). The dominant contributors are local accumulation or long-range regional transport from north of the YRD region, such as Shandong Province and Hebei Province in winter (Ming *et al.*, 2017). With air mass from northern China importing substantial fugitive soils to the YRD region in spring, the PM_{2.5} concentration is enhanced due to the long-range transport (Du *et al.*, 2017). The PRD region is not seriously polluted compared with the YRD region and the BTH region (Zhou *et al.*, 2016). In addition, severe haze pollution episode sometimes happens originating from various sources, such as vehicle emission, coal combustion and the industrial emission (Zhou *et al.*, 2017). Especially, the pollutants from north of PRD region play important roles in dry seasons (Cui *et al.*, 2015). Ma *et al.* (2017) studied five haze

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episodes in Beijing in 2014, which indicated that regional transport from the south is major cause of the initial-stage increase of $\text{PM}_{2.5}$. Studies of regional transport's contribution to $\text{PM}_{2.5}$ have also been performed during the APEC Economic Leaders' 2014 Summit in Beijing. Regional transport plays an important role in the heavy $\text{PM}_{2.5}$ pollution under the strict regional air emission controls (Hua *et al.*, 2016). In summary, the formation of haze pollution should be attributed to both local and long-range transport.

Based on the above background, in order to control severe haze pollution and develop effective emission reduction measures, we should recognize that the main contribution of $\text{PM}_{2.5}$ includes not only local emissions, but also transport in surrounding areas. Although there are many investigations related with regional transport of $\text{PM}_{2.5}$ concentrations, most of them focus on small regions which include the BTH region, the YRD region and the PRD region. The regional transport characteristics of $\text{PM}_{2.5}$ in China are yet to be further studied.

In this study, we used the WRF-CMAQ model to calculate meteorological parameters and $\text{PM}_{2.5}$ concentrations in January and July in 2015, and studied the special distribution of $\text{PM}_{2.5}$ and regional transport characteristics in multiscale regions in China.

METHODS

Study Regions

Fig. 1 shows the study area. We selected provincial capital cities of 31 provinces in China, and studied the distribution of $\text{PM}_{2.5}$ concentrations and regional transport among cities in January and July 2015. These two months were chosen for their representativeness of typical winter and summer climatology conditions, and there was a haze-fog weather process with long duration and strong intensity in January

2015.

In order to study the regional transport characteristics of $\text{PM}_{2.5}$ among multiscale regions in China, we divided the study area into 8 parts, viz., (a) Northeast, (b) North, (c) East, (d) Center, (e) South, (f) Southwest, (g) Northwest and (h) Other (other regions in model domain) (Qi *et al.*, 2014). In addition, we defined 2 large regions, viz., NORTH and SOUTH. NORTH region included Northwest, North and Northeast, and SOUTH region included Southwest, South, Center and East. Each region had different topography and economy, which had an important impact on the emission, diffusion and transport of $\text{PM}_{2.5}$.

Model Description and Configuration

WRF-CMAQ is commonly used in calculation of atmospheric parameters and pollution transport. WRF is a next-generation mesoscale numerical weather prediction system designed for both atmospheric research and operational forecasting needs (Su *et al.*, 2017). CMAQ is an Eulerian photochemical air quality model in which complex interactions between atmospheric pollutants on urban, regional and hemispheric scales are treated in a consistent framework (Chemel *et al.*, 2014). These models have been proven to reasonably perform in most regions in China, and be able to reproduce the concentrations, spatial and temporal variations of pollutants (Zhang *et al.*, 2013; Qin *et al.*, 2015). WRF version 3.4 and CMAQ version 5.0.2 were applied in this study.

WRF-CMAQ simulations were performed over the domain covering East Asia with a grid resolution of 36×36 km, and the grid number was 173×136 . It was divided into 14 layers in the vertical direction in CMAQ. January and July 2015 were simulated with a 5-day spin-up time before the start, which were aimed to minimize the influence of initial conditions.

Fig. 1. Sampling field of China.

The initial and boundary conditions for WRF simulations are prepared using the $1^\circ \times 1^\circ$ resolution NCEP FNL Operational Model Global Tropospheric Analyses dataset (available at <https://dss.ucar.edu/datazone/dsszone/ds083.2/>). Land use data are based on the MODIS 30s resolution WRF input dataset (available at http://www2.mmm.ucar.edu/wrf/users/download/get_sources_wps_geog_V3.html).

Lambert projection coordinate system was used in the simulation area in CMAQ. The domain center was 34°N , 110°E , and the Carbon Bond (CB05) gas-phase chemistry with the AERO6 aerosol module was selected. Input parameters for CMAQ simulations were default clean profile for the initial (ICON) and boundary (BCON) chemical conditions for the domain (Wang et al., 2010). The anthropogenic emissions were based on an inventory developed by Tsinghua University (Zhao et al., 2013a, b). The emission inventory consists of sulfur dioxide (SO_2), nitrogen oxides (NO_x), PM_{10} , $\text{PM}_{2.5}$, black carbon aerosol (BC), organic carbon aerosol (OC), non-methane volatile organic compounds (NMVOC) and ammonia (NH_3) for China with a resolution of $36 \text{ km} \times 36 \text{ km}$ (the same as CMAQ) (Wang et al., 2014; Zhao et al., 2019). NMVOC is categorized into 16 subspecies to match the CB05 chemical mechanism utilized in CMAQ (Yarwood et al., 2005). These pollutants come from 6 sectors including power plants, industrial combustion, domestic combustion, transportation, agriculture and other processes. An “emission factor method” is used to calculate air pollutant emissions, as described in detail in Wang et al. (2011). The detailed methods of estimating each pollutant are introduced in previous studies (Zhao et al., 2013a, b) and the emission inventory is updated to 2015 in a study of Zhao et al. (2018). According to the emission factor method, emissions from 6 sectors are distributed to the $36 \text{ km} \times 36 \text{ km}$ domain, and get the emission file for CMAQ input, which include $\text{PM}_{2.5}$ precursors, such as PEC, POC, PMC, PNO₃, PSO₄, PNH₄ and other pollutants with a total of 42 variables. Biogenic emissions are generated by the Model of Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther et al., 2012).

Zero-out Method

Brute force zero-out method is a sensitivity analysis tool that removes a source to estimate its impact (Cho et al., 2012). Minoura et al. (2016) used the method to evaluate source contributions of 7 sectors to O_3 by completely removing the source. The major concern of using zero-out method is the nonlinear response between emissions and $\text{PM}_{2.5}$ concentrations (Zhao et al., 2015). Wang et al. (2008) proposed that if the basic $\text{PM}_{2.5}$ concentrations were similar with the sum of $\text{PM}_{2.5}$ concentrations from local and regional emissions of zero-out cases, the uncertainties from results of nonlinear response would be small, and confirmed that there are small uncertainties of $\text{PM}_{2.5}$ and PM_{10} . So the results of using zero-out method has small deviation.

In the present study, CMAQ simulations are conducted separately for the base case and the zero-out scenarios. The contribution of that source is assessed by evaluating the

change in ambient air quality, which is measured by difference of sensitivity case and base case:

$$\mu_i = \frac{\text{Base} - \text{Case}_i}{\text{Base}} \quad (1)$$

where *Base* represents the simulated value of the basic situation, and *Case_i* represents the simulated value of zero-out situation for *i* region.

RESULTS AND DISCUSSION

WRF Model Validation

Meteorological conditions have an important impact on the formation, diffusion and sedimentation of atmospheric pollutants. Errors of meteorological parameters will lead to greater errors in pollutant prediction. Therefore, it is necessary to evaluate the performance of WRF. Several meteorological parameters, such as temperature, relative humidity, wind speed and wind direction, are chosen to evaluate performance of WRF. The evaluation indexes are the correlation coefficient (*R*), normalized mean bias (*NMB*) and normalized mean error (*NME*), as shown in Eqs. (2)–(4):

$$R = \sqrt{\frac{\left[\sum_{i=1}^N (S_i - \bar{S})(O_i - \bar{O}) \right]^2}{\sum_{i=1}^N (S_i - \bar{S})^2 \sum_{i=1}^N (O_i - \bar{O})^2}} \quad (2)$$

$$NMB = \frac{\sum_{i=1}^N (S_i - O_i)}{\sum_{i=1}^N O_i} \times 100\% \quad (3)$$

$$NME = \frac{\sum_{i=1}^N |S_i - O_i|}{\sum_{i=1}^N O_i} \times 100\% \quad (4)$$

where S_i represents the simulated value; $\bar{S}$ represents the average value of simulated values; O_i represents the observed value; $\bar{O}$ represents the average value of the observed values; *R* is the correlation coefficient and represents the extent to which simulated values match observed values; *NMB* and *NME* are normalized mean bias and normalized mean error, which represent the degree of deviation between simulated values and observed values.

Observed meteorological data from ground-based measurements are obtained from the University of Wyoming website (<http://www.weather.uwyo.edu/surface/meteorogram/seasia.shtml>), which include hourly temperature, relative humidity, wind speed and wind direction. We choose 23 capital cities to compare the simulated data and observed data by calculating the evaluation indexes.

The evaluation indexes of meteorological parameters are listed in Table 1. The average *R* values of 4 meteorological parameters in January and July are 0.84, 0.78, 0.72 and 0.67, respectively. Most of the *NMB* and *NME* values are

Table 1. Evaluation indexes of meteorological parameters in 2015.

Meteorological parameters	Sampling point	R		NMB (%)		NME (%)	
		Jan.	Jul.	Jan.	Jul.	Jan.	Jul.
Temperature	Shijiazhuang	0.72	0.77	12.07	−28.11	12.07	−13.42
	Changchun	0.81	0.9	3.92	3.42	−14.52	5.19
	Hangzhou	0.81	0.95	6.75	−1.71	20.12	4.41
	Wuhan	0.8	0.86	14.23	1.23	29.23	4.38
	Guiyang	0.89	0.86	−5.85	−1.84	22.31	4.44
	Guangzhou	0.87	0.81	−4.64	0.01	8.22	3.05
Relative humidity	Shijiazhuang	0.81	0.89	−41.43	−27.41	41.43	27.95
	Changchun	0.73	0.73	19.05	−10.74	19.45	12.18
	Hangzhou	0.88	0.85	0.69	12.12	12.26	13.03
	Wuhan	0.87	0.61	−21.23	−12.20	21.57	13.71
	Guiyang	0.66	0.73	−4.10	−2.76	11.61	6.21
	Guangzhou	0.86	0.72	−6.18	−7.57	9.97	8.52
Wind speed	Shijiazhuang	0.71	0.81	1.46	−2.57	22.98	24.75
	Changchun	0.65	0.79	15.53	18.96	28.95	30.36
	Hangzhou	0.77	0.86	16.86	16.47	27.32	32.19
	Wuhan	0.61	0.68	−13.80	−16.32	36.87	35.8
	Guiyang	0.81	0.47	21.65	37.55	26.9	38.9
	Guangzhou	0.8	0.66	11.83	4.5	25.14	26.1
Wind direction	Shijiazhuang	0.68	0.47	−2.16	−10.84	24.44	30.03
	Changchun	0.62	0.61	1.77	−8.16	21.68	20.09
	Hangzhou	0.69	0.74	23.54	2.26	34.33	27.34
	Wuhan	0.81	0.75	5.14	15.52	29.49	33
	Guiyang	0.52	0.73	1.73	27.63	35.02	36.46
	Guangzhou	0.76	0.71	−21.37	−4.31	30.36	31.67

less than 30%, which shows good performance of WRF. Fig. 2 indicates the scatters of daily average of temperature, relative humidity, wind speed and wind direction of 23 meteorological observation sites in January and July. The scatters show that the observed and simulated values have good consistency and small deviation, which further indicates the accuracy of WRF model.

CMAQ Model Validation

The mean fractional error (MFE) and the mean fractional bias (MFB) are chosen as the statistical indicators for evaluation of CMAQ. Boylan and Russell (2006) propose 2 levels of criteria to evaluate the PM model, which are “performance goals” and “performance bias”. They prove that the model “performance goal” is met when MFB is less than or equal to $\pm 30\%$ and MFE is less than $+50\%$. The model “performance bias” is met when the MFB is less than or equal to $\pm 60\%$ and MFE is less than or equal to 75% . MFB and MFE were defined as:

$$MFB = \frac{2}{n} \sum_{i=1}^N \frac{S_i - O_i}{S_i + O_i} \times 100\% \quad (5)$$

$$MFE = \frac{2}{n} \sum_{i=1}^N \frac{|S_i - O_i|}{S_i + O_i} \times 100\% \quad (6)$$

Observations of hourly average concentrations of $PM_{2.5}$ are obtained from Ministry of Environmental Protection

(<http://www.zhb.gov.cn/>) in China collected from monitoring stations around China, and the data are published by the ministry.

Fig. 3 is the values of MFE and MFB for $PM_{2.5}$ concentrations in 31 provinces. Some values of MFE and MFB are between $+50\%$ and -30% , which indicates the accurate performance of CMAQ in simulating $PM_{2.5}$ concentration. Most of the indicators are in the acceptable range, and the model performance satisfies the criteria.

Fig. 4 shows the scatter plots of simulated and observed daily average $PM_{2.5}$ concentrations for 31 provinces. There are 930 data points in January and July and R^2 are 0.43 and 0.41 respectively. From the scatters we find that the simulations underestimate $PM_{2.5}$ concentrations. The underestimation is most likely caused by missing pathways in secondary aerosol formation and uncertainties in WRF simulations. Fig. 5 shows the time series of $PM_{2.5}$ daily average concentration in January and July. 7 cities are chosen to represent 7 regions in China. We compare simulated values and observed values in January and July. Most of the cities show good performance with reasonable values of R, MFB and MFE. We calculate the mean R, MFB and MFE of each region and choose the cities with similar parameters with mean values of their regions, which can show an average level of CMAQ model performance. $PM_{2.5}$ concentrations in January are higher than those in July. The highest concentration is up to $300 \mu g m^{-3}$ in Beijing. Despite these uncertainties, the simulation result from CMAQ is generally acceptable and able to represent the local and regional transport patterns of $PM_{2.5}$ in China.

Fig. 2. Scatters of daily average values (temperature, relative humidity, wind speed and wind direction) of 23 cities in January and July.

Fig. 3. Mean fractional error (MFE) and mean fractional bias (MFB) of $PM_{2.5}$.

Fig. 4. Daily average $PM_{2.5}$ concentration scatter plot of 31 provincial capitals in January and July.

$PM_{2.5}$ Distribution

Fig. 6 shows the distribution of monthly average concentration of $PM_{2.5}$ in January and July in China. An obvious regional $PM_{2.5}$ concentration characteristic with higher $PM_{2.5}$ concentration in winter and lower concentration in summer is depicted. In January, the most polluted regions are Beijing, Hebei, Jilin, Chongqing and Shanghai with the monthly average concentration of $PM_{2.5}$ exceeding $130 \mu g m^{-3}$. Dense population distribution and fossil fuel combustion are the main causes of high $PM_{2.5}$ concentrations. In July, the monthly average concentrations of $PM_{2.5}$ in most regions are less than $60 \mu g m^{-3}$. Only the $PM_{2.5}$ concentrations in Chongqing and Sichuan are still high, which may be related to the basin geomorphology. From a regional perspective, monthly concentration of $PM_{2.5}$ in eastern China is higher than that in the west. With the population growing and economic development, the urbanization and industrialization process in eastern China

is faster than that in the western region, which leads to excessive emissions and a high level of $PM_{2.5}$ concentration.

Inter-provincial Transport of $PM_{2.5}$

In order to analyze the characteristics of inter-provincial transport among the 31 provinces in China, we use the method of zero-out to set the emission inventory to zero in a certain province and calculate the distributions of that province on the other provinces. In total, 31 scenarios are simulated for January and July, respectively. This paper focuses on 4 key areas of the Beijing-Tianjin-Hebei region, the Yangtze River Delta, the Pearl River Delta and the Sichuan-Chongqing Economic Zone, which are densely populated and economically developed regions.

Fig. 7(a) shows the provincial transport contribution matrix of $PM_{2.5}$ concentration in January. The inter-provincial transport presents different characteristics in summer and winter, which results from the different prevailing wind in

Fig. 5. Time series of daily average PM_{2.5} concentration in January and July in 8 cities.

January and July. In January, the contributions of PM_{2.5} from external regions to Beijing, Tianjin and Hebei are 44.81%, 61.48% and 14.52%, respectively. Change *et al.* (2019) had similar result with 38% contribution from regional transport in Beijing in January. Hebei is the

dominant external contributor to PM_{2.5} in Beijing and Tianjin with the contributions of 24.30% and 38.05%, respectively. Shandong (2.56%), Shanxi (2.40%) and Inner Mongolia (2.33%) become the major external source contributors in Hebei, in which local emission is the

Fig. 6. Monthly average concentration of $PM_{2.5}$ in January and July 2015.

biggest contribution to $PM_{2.5}$ concentration. As we know, Hebei is one of the most polluted province in the Beijing-Tianjin-Hebei region. In the Yangtze River Delta, external transport is the major source of $PM_{2.5}$ concentration in Shanghai, Zhejiang and Jiangsu, which generally accounts for 90.65%, 62.69% and 46.48%, respectively. Emissions from Jiangsu contribute as much as 20% of $PM_{2.5}$ concentration in Shanghai and Zhejiang. The external province contributing most to Jiangsu is Anhui with the contribution of 11.47%. We find that inter-provincial transport in the Yangtze River Delta and transport from northern China are the major sources of $PM_{2.5}$ concentrations in Shanghai, Zhejiang and Jiangsu Provinces. In the Pearl River Delta, external transport is the major source in Guangdong and Guangxi accounting for 65.48% and 62.68% of $PM_{2.5}$, respectively. Wu *et al.* (2013) used CAMx model and PSAT technology to simulate the spatial resources of $PM_{2.5}$ in major cities in the Pearl River Delta, among which 69.6% of the $PM_{2.5}$ concentration in Guangzhou come from outside the PRD. The result was similar to ours. Meanwhile, the external sources only contribute 30.94% and 34.78% to Sichuan and Chongqing, and they are the largest contributor to each other. Besides the provinces in these 4 regions, Inner Mongolia, Heilongjiang and Jilin are less affected by external sources, with the contributions of 17.92%, 23.18% and 21.70%, respectively. Hainan, Fujian and Jiangxi are significantly affected by external sources with the contributions of 97.18%, 88.86% and 77.71%. In summary, there is distinct inter-provincial transport among the 31 provinces, and in some provinces external transport is the major source.

Fig. 7(b) illustrates the source contribution of inter-provincial transport of $PM_{2.5}$ in July 2015. From the contribution matrix, we find that the inter-provincial transport shows a different characteristic. In the Beijing-Tianjin-Hebei region, $PM_{2.5}$ concentration mainly comes from local emissions with the contribution of 69.03% and the dominant provinces are Hebei, which is the most severe polluted provinces in North China. $PM_{2.5}$ concentration in Tianjin is also mainly contributed by Hebei and Shandong

Provinces with the rates of 29.5% and 22.5%. Meanwhile, local contribution is the major source in Hebei Province accounting for 69.05% of $PM_{2.5}$ concentration in Hebei. In the Yangtze River Delta, the predominant sources of $PM_{2.5}$ in Shanghai are from external transport with the contribution of 86.39%, and $PM_{2.5}$ in Zhejiang and Jiangsu provinces are mainly from local emissions. In the Pearl River Delta, the external contributions of $PM_{2.5}$ are 32.47% and 33.02% in Guangdong and Guangxi, respectively. Emissions from Jiangxi Province contribute most to Guangdong Province with the contribution of 9.99% and Hunan is the biggest contributor to Guangxi Province accounting for 6.81% of $PM_{2.5}$. In the Sichuan-Chongqing Economic Zone, local sources are the major contributor to $PM_{2.5}$, and external transport of $PM_{2.5}$ accounts for 25.02% and 23.72% in Sichuan and Chongqing. The results from Xue *et al.* (2019) show that emissions within Sichuan and Chongqing are the major contributors to $PM_{2.5}$ in both seasons (~80%) and emissions outside the region contribute approximately 20%. Furthermore, Qinghai and Tibet are also more affected by external sources with the contributions of 71.20% and 75.71%, respectively.

Seasonal variations of regional transport can result from meteorological conditions or the contribution of emission-intensive sources (Yan *et al.*, 2016). We can identify the key $PM_{2.5}$ transport pathways of northwest-southeast in January and southeast-northwest in July brought by winter and summer monsoon, respectively (Change *et al.*, 2018). For example, the regional transport of $PM_{2.5}$ concentrations in Anhui Province mainly come from Shandong Province (12.87%) and Henan Province (5.64%) in January and Jiangsu Province (26.47%) in July. Regional transport of $PM_{2.5}$ concentrations in Henan Province are mainly from Hebei Province (10.96%) and Shanxi Province (12.71%) in January and Shandong Province (10.12%) in July. These major transport directions are the same with monsoon directions in winter and summer respectively, and the nearby upstream city is always one of the largest contributors besides local emissions (Chang *et al.*, 2019). Furthermore, the domestic heating demand in winter causes high

anthropogenic emissions, which leads to a higher level of $PM_{2.5}$ concentrations than those in summer. The relatively stable atmospheric condition in winter inhibits the diffusion of pollutants (Zhi *et al.*, 2015; Sun *et al.*, 2014). Therefore, the concentrations from regional transport in winter are larger in absolute value and smaller in percentage. For example, the regional transport of $PM_{2.5}$ concentrations in Beijing are mainly from Hebei Province with a percentage of 24.30% in January and 27.45% in July. The contributions are similar, but the $PM_{2.5}$ concentrations in January in Beijing are larger than those in July. In Sichuan Province,

regional transport of $PM_{2.5}$ are mainly from Chongqing with 10.76% in January and 10.91% in July.

In summary, there are significant $PM_{2.5}$ regional transport phenomena in all provinces in January and July, 2015. Due to the influence of the terrain and the meteorology, the transport characteristics vary among different provinces. Neighboring provinces are generally major external sources of each province. Moreover, northwest wind dominates in January and southeast wind dominates in July. The regions in the downwind are generally affected by the regional transport of $PM_{2.5}$ from the regions in upwind areas.

Fig. 7. Source contribution (%) of inter-provincial transport of $PM_{2.5}$ (from Province1 to Province2) in (a) January and (b) July 2015.

Fig. 7. (continued).

Regional Transport of PM_{2.5}

Additionally, 31 provinces are divided into 7 regions. We use the method of zero-out to investigate PM_{2.5} transport characteristics among 7 regions. The relative difference of PM_{2.5} concentrations between basic scenario and zero-out scenario is the contribution of the region.

Table 2 displays the regional transport contribution matrix of PM_{2.5} in January 2015. The matrix demonstrates that PM_{2.5} mainly comes from external sources in the South region with the contribution of 59.18%, while PM_{2.5} concentrations mainly come from local sources in the other 6 regions with the contributions exceeding 65%. In terms

of regional transport, the East region contributes most to the North region. The North region contributes most to the Northeast, East, Center and Northwest region. The Center region contributes most to the South and Southwest region. In summary, local sources of PM_{2.5} are the major contributors to PM_{2.5} concentration, and there exists the regional transport of PM_{2.5} in each region.

Similarly, the contribution matrix of regional transport in July 2015 is listed in Table 3. It presents that local sources are the major contributors in each region in July. In terms of regional transport, the East region contributes most to the North, Northeast and Center regions. The Center

Table 2. Contribution matrix of PM_{2.5} regional transport in January 2015.

	North	Northeast	East	Center	South	Southwest	Northwest
North	93.56%	4.96%	13.25%	14.08%	8.85%	2.15%	6.68%
Northeast	0.35%	93.99%	1.67%	0.50%	0.39%	0.69%	0.02%
East	3.00%	0.81%	68.37%	12.34%	17.20%	1.13%	0.60%
Center	1.18%	0.10%	7.49%	68.77%	19.65%	5.53%	3.25%
South	0.00%	0.00%	0.22%	0.68%	40.82%	0.80%	0.05%
Southwest	0.06%	0.07%	0.62%	0.60%	4.93%	84.67%	2.07%
Northwest	1.79%	0.06%	1.85%	3.04%	4.72%	5.03%	86.99%

Table 3. Contribution matrix of PM_{2.5} regional transport in July 2015.

	North	Northeast	East	Center	South	Southwest	Northwest
North	79.57%	6.16%	5.62%	7.93%	1.03%	2.43%	7.99%
Northeast	3.48%	85.17%	0.75%	0.61%	0.12%	0.11%	0.15%
East	17.62%	7.38%	74.13%	19.42%	6.91%	3.59%	5.87%
Center	3.09%	1.05%	8.20%	73.81%	8.40%	5.93%	7.76%
South	0.04%	0.08%	1.17%	0.64%	77.46%	0.34%	0.01%
Southwest	0.10%	0.10%	0.69%	2.32%	2.94%	86.84%	2.57%
Northwest	1.77%	0.40%	0.79%	1.30%	0.64%	3.72%	76.18%

region contributes most to the East, South and Southwest regions. The North region contributes most to the Northwest region. Compared with January, the contribution characteristics are different because of the different prevailing wind directions. In January, the prevailing wind is the north wind. The emissions in the North region are the major regional transport sources of other regions. In July, the prevailing wind is southeast wind. The contributions of the East region to the North, South, Center and Northeast region have increased.

Analysis of Contribution to PM_{2.5} in Each Small Region

Characteristics of regional transport among provinces and regions are analyzed in the study. Local emission is the major contributor to each region, and the regional transport is mainly from surrounding provinces. To find the province that contributes most in each region, we calculate the contributions of provinces in each region.

Fig. 8 shows the regional contribution of PM_{2.5} in 7 regions in January and July. Hebei Province contributes most to the North region with the contributions of 49% and 44% in January and July, respectively. The contribution of external sources is higher in summer which comes from Eastern Plains.

The Northeast region is located in the Northeast Plain with Changbai Mountains in the east and Greater Khingan Mountains in the west. Depending on the terrain, PM_{2.5} is easily affected by the North region. Jilin contributes the most to PM_{2.5} in the region with the contribution rate of 40%. Liaoning and Heilongjiang contributes almost the same to this region, with the contribution rates of 28% and 26%, respectively.

The East region is distributed in the southeast coast. The figure shows that Shandong and Jiangsu are the main contributors to PM_{2.5} concentration in this region with Shandong contributing most in winter and Jiangsu contributing most in summer. In conclusion, concentration

of PM_{2.5} is higher in the north of the East region and lower in the south of this region.

The Center region consists of Henan, Hubei and Hunan and is located in the intersection of North China Plain and the Yangtze River region. The 3 provinces show almost the same contributions of PM_{2.5} in winter and summer. Contribution of external transport is higher in summer with the contribution of 16% which results from the high topography in the west. PM_{2.5} transport from the west in winter is blocked by the mountains which decreases the contribution of external sources.

The South region is located along the South China Sea coast. The atmospheric pollution in the Pearl River Delta is characterized by the coexistence of soot and photochemical pollutants. The pollutants are transported and converted into each other in the boundary layer. The pollutants from different cities transfer and interact with each other. External transport of PM_{2.5} is a major contributor accounting for 59% and 50% in winter and summer, respectively. The external sources are mainly from the Center and East region. Furthermore, the study of Deng *et al.* (2008) demonstrated that the pollutants emitted by biomass combustion in Southeast Asia were prone to increase aerosol concentration in Guangzhou and thus increase the contribution of external sources.

The Southwest region consists of Sichuan, Chongqing, Guizhou, Yunnan and Tibet, which includes the Sichuan Basin, the Yunnan-Guizhou Plateau and Qinghai-Tibet Plateau. Sichuan and Chongqing Provinces are the major contributors both in winter and summer.

The Northwest region has complex terrain including Western Loess Plateau, Qilian Mountains, Zhungeer Basin and Tarim Basin. Emissions from Shanxi (Xi'an) account for as much as 45% in winter and summer. Due to the dusty weather in the Northwest region, PM_{2.5} concentration is difficult to simulate and observe. Therefore, the simulation in this area is only considered as a reference.

Fig. 8. Contributions of provinces to PM_{2.5} in each region.

Transport of PM_{2.5} between NORTH and SOUTH Region

We divided the 7 small regions (excluding “Other”) into 2 big regions (NORTH and SOUTH) to represent north and south parts of China and simulate the contributions of the 2 regions to PM_{2.5} concentration in each region in winter and summer.

Fig. 9 is the contributions of NORTH and SOUTH regional transport of PM_{2.5} to each region in January and July 2015. In the NORTH region, local contributions are the major sources of PM_{2.5} in January and July, which are 97.80% and 90.86%, respectively. The SOUTH region has very little contribution of PM_{2.5} to the NORTH in January, while it contributes more in July, up to 8.74%. The reason is related to southeast monsoon in summer and northerly wind in winter in China (He *et al.*, 2017). The prevailing wind in summer results in increasing of contribution of the SOUTH to the NORTH. In the SOUTH region, local contributions are also the major sources in winter and summer. The NORTH region has more contributions to PM_{2.5} concentration in the SOUTH region in winter and

summer with the ratio of 16.08% and 9.53%. For the same reason of prevailing wind, the NORTH region makes a bigger contribution to the SOUTH in January than in July.

CONCLUSIONS

WRF-CMAQ was used to simulate the temporal-spatial distribution and multiscale transport patterns of PM_{2.5} during January and July of 2015 in China. We used the brute force zero-out method to characterize the regional transport of PM_{2.5} in 31 provinces, 7 small regions (North, Northeast, East, Center, South, Southwest and Northwest) and 2 large regions (NORTH and SOUTH). Based on the statistical indicators, WRF-CMAQ’s performance was acceptable. Several points are noted below.

The PM_{2.5} was contributed by both internal and external sources in the provinces, with local emissions dominating some areas, e.g., Xinjiang (91.26%), Hebei (85.48%) and Inner Mongolia (82.02%) during January and Xinjiang (91.97%), Chongqing (76.28%) and Sichuan (74.98%)

Fig. 9. Source contribution of large regional transport of PM_{2.5} in January and July 2015.

during July, and regionally transported pollution dominating others, e.g., Qinghai (97.18%), Shanghai (90.65%) and Fujian (88.86%) during January and Shanghai (86.39%), Qinghai (75.71%) and Tibet (71.20%) during July.

However, local emissions were the primary source of PM_{2.5} in every region, with Hebei, Chongqing, Shanxi and Guangdong being the major contributor in the North, Southwest, Northwest and South region, respectively. In the Northeast and the Center, the provinces contributed almost equally to the PM_{2.5} concentrations. Therefore, local emission was also responsible for most of the PM_{2.5} observed in the 2 large regions, the NORTH and the SOUTH, whereas the external contribution accounted for 10% or less and varied by season.

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