SCIENCE CHINA Earth Sciences

SPECIAL TOPIC: The Severe Fog and Haze Event over Eastern China in January 2013 • RESEARCH PAPER • January 2014 Vol.57 No.1: 3–13 doi: 10.1007/s11430-013-4793-0

Modeling study of regional severe hazes over mid-eastern China in January 2013 and its implications on pollution prevention and control

WANG ZiFa^{1*}, LI Jie¹, WANG Zhe¹, YANG WenYi¹, TANG Xiao¹, GE BaoZhu¹, YAN PinZhong¹, ZHU LiLi¹, CHEN XueShun¹, CHEN HuanSheng¹, WAND Wei¹, LI JianJun², LIU Bing², WANG XiaoYan², WAND Wei², ZHAO YiLin², LU Ning² & SU DeBin³

¹ State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029, China;
² China National Environmental Monitoring Center, Beijing 100012, China;

³ Beijing Meteorological Bureau, Beijing 100089, China

Received October 16, 2013; accepted November 27, 2013; published online December 6, 2013

The Nested Air Quality Prediction Model System (NAQPMS) was used to investigate the temporal and spatial variations of $PM_{2.5}$ over tropospheric central eastern China in January 2013. The impact of regional transport and its implications on pollution prevention and control were also examined. Comparison between simulated and observed $PM_{2.5}$ showed NAQPMS was able to reproduce the evolution of $PM_{2.5}$ during heavy haze episodes. The results indicated that regional transport of $PM_{2.5}$ played an important role in regional haze episodes in the city cluster including Hebei, Beijing and Tianjin (HBT). The cross-city clusters transport outside HBT and transport among cities inside HBT contributed 20%–35% and 26%–35% of $PM_{2.5}$ as compared with local emission, in HBT respectively. To meet the Air Quality Standards for Grade II, 90%, 90% and 65% of emissions would have to be cut down in Hebei, Tianjin and Beijing, if non-control strategy was taken in the surrounding city clusters of HBT. This implicated that control of emissions in one city cluster is not sufficient to reduce regional haze events, and joint efforts among city clusters are essential. Besides regional transports, two-way feedback between boundary-layer evolution and $PM_{2.5}$ also significantly contributed to the formation of heavy hazes, which contributed 30% of monthly average $PM_{2.5}$ concentration in HBT.

regional hazes, trans-boundary transport, feedback between boundary-layer evolution and PM_{2.5}, pollution prevention and control

Citation: Wang Z F, Li J, Wang Z, et al. 2014. Modeling study of regional severe hazes over mid-eastern China in January 2013 and its implications on pollution prevention and control. Science China: Earth Sciences, 57: 3–13, doi: 10.1007/s11430-013-4793-0

In the last decades, China has undergone an accelerated process of urbanization, manifested by urban population growth, expansion of existing cities, and the rapid emergence of new city centers. Super city clusters in China are being formed one after another (i.e., Hebei-Beijing-Tianjing Region (HBT), Yangtze River Delta, and Pearl River Delta) and heavy polluting industries moved from core cities of city clusters to their neighboring cities. This has caused a remarkable change in the distribution pattern of atmospheric pollutant emissions. As a result of this change, regionalscale severe and complicated air pollution (coal-burning pollution, photochemical smog, acid deposition, haze, etc.) is frequently found and getting worse and worse in East

^{*}Corresponding author (email: zifawang@mail.iap.ac.cn)

[©] Science China Press and Springer-Verlag Berlin Heidelberg 2013

China (Chang et al., 2009; Shao et al., 2006; Wu, 2012), although sulfur dioxide (SO₂) emission gross has been reduced by 10% and its concentration in urban region has obviously decreased during the past five years. Annual PM_{2.5} (fine particulate matter) concentration in China even exceeded 3-5 times of that in developed centuries (Chang et al., 2009; Shao et al., 2006). In particular, an unprecedented severe haze occurred over mid-eastern China in January 2013. In Beijing, the maximum of hourly PM_{2.5} concentration was beyond 600 μ g m⁻³ (Zhang et al., 2013). Its cause and control strategy had aroused broader concerns of the international community. How to improve air quality in the rapid economic development and make optimized control strategies have become not only the most concerned frontier focus in international communities in atmospheric chemistry but also the scientific basis to conducting synergistic control for atmospheric pollution.

Besides the extremely stable boundary layer (Zhang et al., 2013), trans-boundary transport was thought to be another important contributor to regional haze resulting from regional pollutant emissions around urban clusters. Wang et al. (2011) found pollutants produced in HBT can be transported to Northeast China and greatly promoted the formation of haze there, and Wu et al. (2011) found the contribution of emissions over surrounding areas of Beijing to the O₃ concentration over Beijing was close to 50%. However, the quantitative impact of regional transport on the haze under extreme weather condition (e.g., the haze over mid-east China in January 2013) remains unclear. In particular, particles experienced an intensive aging process in China. The aged particles were found to be more hydrophobic than fresh ones and easily converted to cloud condensation nuclei (e.g., MTX2006, BECAPEX, CAREBeijing, PRIDE-PRD2004 and Hachi 2009–2010). The interaction between aerosols with different oxygenation levels in the cross-cities transport during the haze period perplexed this quantitative evaluation on regional transport. Therefore, studies for its quantitative estimation are difficult but necessary for learning its impact on formation of regional haze.

In this study, we use a regional air quality model (NAQPMS) to investigate the spatial and temporal variations of $PM_{2.5}$ and its formation mechanisms in January 2013 over mid-eastern China. We preliminarily evaluate the impact of regional transport and clarify its implications on the air pollution prevention and control. Therefore, we provide constructive suggestions for the protection and management of atmospheric environment in China.

1 Model setup and validation

1.1 Model description

The Nested Air Quality Predicting Modeling System (NAQPMS) is a multi-scale air quality modeling system developed by the Institute of Atmospheric Physics, Chinese

Academy of Sciences (IAP/CAS), targeting at reproducing the transport and evolution of atmospheric pollutants in China. It included modules used for real-time emissions, advection, diffusion, dry and wet deposition, aerosol, gaseous phase, aqueous phase, and heterogeneous atmospheric chemical reactions (Wang et al., 2001). Meanwhile, techniques like tracer-tagging, sensitivity analysis, and data assimilation are applied to run this modeling system. The predominant features of NAQPM are: a two-way nested module for reproducing the interaction between regional and urban scale pollution; a dust deflation mechanism for the transport of Asian mineral dust; a new gaseous chemical mechanism (CBM-Z) involving 71 species and 134 chemical reactions. The aqueous chemistry involving 22 species of gases and aerosols was simulated by the RADM mechanism. As for the simulation of heterogeneous chemical process, 14 species and 28 reactions on dust, sea salt, sulfate, and black carbon are involved. The details about NAQPMS can be found in Li et al. (2011, 2013).

NAQPMS has been used in studies for the formation and its long-range transport of dust, acid rain, and secondary pollutants (ozone and $PM_{2.5}$) (Li et al., 2012). Meanwhile, it has been widely used in routine operation in local Environmental Protection Bureau of Beijing, Shanghai, Guangzhou, and elsewhere. In particular, NAQPMS has made a great contribution to the air quality assurance during the Beijing Olympic Games, Shanghai World Expo and Guangzhou Asia Games (Wu et al., 2010).

1.2 Tracer-tagging

Traditionally, sensitivity analysis (the difference of two model simulations with different emissions yields the contribution of individual source to the concentration of target pollutant) is widely used to quantitatively estimate the impact of regional transport. However, the recent research showed that the sensitivity analysis underestimated the actual contribution of target emissions to secondary pollutants. Take O₃ for example, it has been underestimated 40% in global scale and 10%-20% over East China (Grewe, 2004). It is caused mainly by the non-linearity of chemical reactions in the atmosphere, which means the production efficiency of secondary pollutants in two simulations apparently differs. To avoid this problem, NAQPMS established a new algorithm as an online tracer-tagged method. In this algorithm, source-receptor relationships between various areas can be obtained through only one calculation. More importantly, the production efficiency of secondary pollutants won't be changed, which avoids the model errors caused by the nonlinearity of atmospheric chemistry. More introductions can be found in Li et al. (2008, 2010b) and Wu et al. (2010).

1.3 Model setup

The model domain is composed of two nested domains

(Figure 1). The first domain covers the whole East Asia with 80 km×80 km horizontal resolution and 97×77 grids in the latitudinal and longitudinal direction, respectively. The second domain includes areas like East China (the east part of 110° E), Korean Peninsula and Japan with 20 km×20 km horizontal resolution and 160×148 grids. Vertically, NAPQMS ranges from surface to 20 km a.s.l, with 20 terrain-following layers. There were seven layers within the lowest 1 km above the surface.

Hourly meteorological field is provided by Mesoscale meteorological model WRFv3.5. The anthropogenic emission was obtained from the 2007 bottom-up Regional Emission inventory in ASia (REAS 2.1) data with $0.25^{\circ}\times0.25^{\circ}$ resolution developed by National Institute of Environmental Sciences of Japan. The emission species include SO₂, volatile organic compounds (NO_x), volatile organic compounds (VOC_s), black carbon (BC), organic matter (OM), and primary PM_{2.5} (Kurokawa et al., 2013). The natural NHHC emissions were obtained from The Global Emission Inventory Activity (GEIA) (Guenther et al., 1995). Figure 1 shows the emission rate of primary PM_{2.5} in this study.

The 16th December 2012 to 31st January 2013 was selected as the simulation period with 5-min time step and the first 15 days were spin-up period to exclude the impact of uncertainties of initial conditions. Involved $PM_{2.5}$ species were sulfate, nitrate, ammonium salt, BC, OM, primary $PM_{2.5}$, dust, sea salt and so on. The initial and boundary conditions were taken from the modeling result of a global model (MOZARTv2.4).

For estimating the impact of different areas on PM_{2.5} regional distribution, 34 production regions have been identified by NAQPMS, which were all the provinces in China and the rest regions inside the modeling region (Japan, Korean peninsula, Siberia, Southeastern Asia, Northwestern Pacific, and parts of India). In the following section, the contribution of $PM_{2.5}$ produced in HTB and its surrounding provinces to $PM_{2.5}$ concentration in HTB during the haze episode will be mainly analyzed.

1.4 Model evaluation

For evaluating its reasonability, simulated PM2.5 was compared with observed ones. In this study, observed PM2.5 was taken from the National Observation Network of Atmospheric Pollutants established by China National Environmental Monitoring Centre. Figure 2 is the time series of simulated and observed PM2.5 concentrations in different cities. In general, the NAPQMS reasonably reproduced the spatial and temporal variation of PM2.5 daily average concentration during January 2013 at most stations. In Northeast China (Shenyang), observed daily average concentrations of $PM_{2.5}$ were 115 µg m⁻³ or more (the Air Quality Standards for Grade III, moderate pollution) sustained from 3rd till 24th January. However, high concentrations of observed PM_{2.5} (above 200 µg m⁻³) in northern part of the North China Plain (e.g., Tianjin, Beijing) were influenced mainly by pollution events lasting from 2 to 3 days. PM_{2.5} concentrations in southern part of the North China Plain (e.g., Shijiazhuang, Cangzhou and Jinan) were much higher than the northern part, which were all above 200 μ g m⁻³ in most of time in January 2013. This observed spatial and temporal distribution of PM2.5 was reasonably reproduced



Figure 1 Two nested modeling domains and primary PM_{2.5} emission rates in this study. Observation sites for model evaluation are also shown.



Figure 2 Observed (black) and simulated (red) daily average concentrations of $PM_{2.5}$ at sites in China shown in Figure 1 in January 2013. Gray shadows stand for the standard deviation of observation concentration.

by the model. Note that both observations and simulation showed that $PM_{2.5}$ of the west parts to the Taihang Mountain was less than the east parts. Take Taiyuan for example, periods with high $PM_{2.5}$ daily average concentrations (>200 µg m⁻³) were only one weeklong (17th–23rd). Compared with those in the North China Plain and Northeast China, $PM_{2.5}$ concentrations in Yangtze River Delta (Jiangsu, Shanghai, and Zhejiang) were lower but still above 75 µg m⁻³ (Grade II, slight pollution).

The modeling results of Shijiazhuang (200–400 μ g m⁻³) were lower than the observation results (300–800 μ g m⁻³) during 10th–13th January. This is likely caused by the coarse resolution, uncertainty of emissions, and two-way feedback between PM_{2.5} and boundary-layer evolution. Shijiazhuang city is located at the foot of the Taihang Mountain, the local circulation (e.g. mountain-valley breeze) and the impact of PM_{2.5} loading on boundary layer by absorbing and scattering solar radiation were hard to be reproduced by the off-line chemistry transport model with 20 km simulation resolution. In the section 2.2, the impact of feedback mechanism between PM_{2.5} and boundary-layer on modeling results will be analyzed specifically.

2 Results and discussion

2.1 The regional transport during the haze period in HBT

2.1.1 The regional distribution of PM_{2.5} and its transport characteristics

(i) The regional distribution of surface PM_{2.5}. Figure 3 shows that the distribution of PM_{2.5} average concentration in East China in January 2013. PM_{2.5} concentrations in most provinces of mid-East China extending from Hunan to Northeast China have surpassed Grade II (75 μ g m⁻³) during the modeling period. The high concentration appeared mainly in southern part of North China (southern Hebei province and Shandong), central China (Henan province and Hubei province), and Anhui province, which was beyond 150 μ g m⁻³ (Grade IV, very unhealthy). The PM_{2.5} concentrations in south part of Hebei and north part of Henan even reached the hazardous level (>250 μ g m⁻³).

This regional severe haze was closely related to the abnormal weather conditions occurred in 2013. Compared with the weather conditions and meteorological elements of January in the past 30 years, weather conditions with high humidity and low visibility during this haze episode are barely seen in history. It was only observed once in 1989, 1992, 2001 and 2006, respectively (Huang et al., 2008). In particular, a Stratospheric Sudden Warming (SSW) in high latitudes was responsible for this extreme stable weather in 2013. SSW caused a long-term stagnation of North-Asia polar vortex in the Kamchatka coast and a superimposition in phase between northern and southern branch of ridge



Figure 3 Simulated monthly average concentrations of $PM_{2.5}$ and surface wind fields (m s⁻¹) in East China in January 2013.

from Xinjiang to south Asia subcontinent. As a result, the small trough in the north of Caspian Sea was cut off, which forced stable weather to last over the entire North China in January 2013 (Li et al., 2010a). And it is favorable for the formation of regional severe pollution.

(ii) The evidence of regional transport in stable weather. As shown in Figure 2, a regional haze episode occurred over HBT during 10th–13th January, in which the maximum of boundary layer height of Beijing was only 400–800 m (Aerosol lidar at IAP/CAS in Beijing, figure not shown). Figure 4 shows the time series of hourly PM_{2.5} concentration at different stations in HTB in this episode. It is clear that there was a time lag of PM_{2.5} concentration peak (>500 μ g m⁻³) from south to north along Baoding- Shijiazhuang-Tianjin-Beijing under the prevailing 3 m s⁻¹ southerly wind. And backward trajectory analysis indicated most of the pollutants in Beijing during this peak came from the regional transport from south.

Figures 5 and 6 show the daily average concentrations of $PM_{2.5}$ in East China and their transport fluxes during the studied case. Before this regional pollution over HTB (8th Januanry, 2013), as shown in Figure 5(a), the surface $PM_{2.5}$ concentration kept low in most regions of HBT under the prevailing northerly winds, with magnitudes of <75 and 100 µg m⁻³ in northern and southern HBT, respectively. The pollutants emitted in HBT were transported southwardly to Shandong, Henan, Anhui and north part of Jiangsu with transport fluxes of 800 µg m⁻² s or more which caused high pollutions there. During this episode (10th–13th January 2013), the prevailing southerly winds in the Huang River and Huai River Plain brought northwardly PM_{2.5} to HBT and Northeast China along the lines of Henan-Hebei-Beijing



Figure 4 Observed hourly average concentration of $PM_{2.5}$ at different sites during 9th–16th January 2013. (a) Sites in Shijiazhuang, Baoding and Tianjin; (b) sites in Yufa located in southern Beijing and IAP located in central Beijing.



Figure 5 The simulation results of daily average concentrations of PM_{2.5} and surface wind fields (m s⁻¹) in East China during 8th–12th January 2013.

1an 10 45°N 45°N 40°N 40° Flux 35°N 35° (µg m⁻² s⁻¹) 30°N-110°F 130°E 120°F 120°E 130 800 500 45°N 45°N 300 40°N 40°N 35° 35°ľ 30°**N⊣** 110°E 30° 130°E 110°E 130°E 120°E 120°E 500 500

Figure 6 The same as Figure 5, but for transport fluxes of PM_{2.5}.

and Shandong-Hebei-Bohai Bay-Liaoning, under the weak pressure system in the East China Sea. The fluxes even reached up to 800 μ g m⁻² s⁻¹ (Figure 6(b)–(d)). Consequently, daily average concentrations of PM_{2.5} of Tianjin and Beijing increased to 200–400 μ g m⁻³ (Figure 5(b)–(d)).

2.1.2 The impact of different area on $PM_{2.5}$ concentration of HTB

Table 1 lists the simulated impact of different area on monthly averaged $PM_{2.5}$ at stations in HTB (Beijing, Tianjin, Qinhuangdao and Cangzhou). On average, the self contribution (defined as the contribution of one source tagged region to itself) reached 46.8 µg m⁻³ (48.3%) and 43.7 µg m⁻³ (32.6%) at Beijing and Tianjin. In northern (Qinhuangdao) and southern Hebei province (Cangzhou), self contributions were 42.8 µg m⁻³ (54.1%) and 85.1 µg m⁻³ (47.1%), respectively. Regional transport contributed 46%–52% PM_{2.5} at

various stations in HBT and exhibited distinct features in different regions. In Beijing, source regions of transport were Hebei (26%) and Inner Mongolia (10.8%), while they were Hebei (35.4%), Shandong (9.9%) and Beijing (8.4%) in Tianjin, respectively. In northern Hebei (Qinhuangdao), regional transport from other regions (mainly referring to Liaoning) was second (17.1 μ g m⁻³, 21.6%) only to its self contribution. As for the southern Hebei (Cangzhou), contributions of Shandong and Tianjin were 31.5 μ g m⁻³ (17.5%) and 23.6 μ g m⁻³ (13.1%), while Henan, Anhui, and Jiangsu contributed more than 20.4 μ g m⁻³ (11.3%) PM_{2.5}.

As shown in Table 1, transport from other city clusters outside HBT also played an important role. The contribution of trans-city cluster transport to Beijing, Tianjin, Cangzhou and Qinhuangdao reached 23.4%, 23.6%, 34.7% and 34.0%, respectively. This suggested that pollution control measures should not be limited in certain single city cluster, and the

 $\label{eq:Table 1} \mbox{ Contributions } (\mu g \ m^{-3}) \ of \ different \ source \ areas \ to \ monthly \ averaged \ PM_{2.5} \ at \ typical \ sites \ in \ HBT$

Sites ^{a)}	Simulated PM _{2.5}	Beijing	Tianjin	Hebei	Shandong	Shanxi	Inner Mogolia	Henan, Anhui Jiangsu	Others
Beijing	96.7	46.8	1.8	25.5	0.8	8.8	10.5	0.6	1.9
Tianjin	133.9	11.2	43.7	47.4	13.3	3.3	5.0	5.2	4.8
Qinhuangdao	79.1	6.0	3.7	42.8	1.4	1.8	5.4	0.9	17.1
Cangzhou	180.5	9.2	23.6	85.1	31.5	3.0	4.2	20.4	3.5

a) Locations of sites are shown in Figure 1

other clusters have to be considered as well.

Figure 7 shows the contribution of HTB and its surrounding areas to $PM_{2.5}$ regional distribution during 10th–13th January. Compared with the monthly average concentration, trans-city cluster transport played a more important role in $PM_{2.5}$ distribution. For example, the self contributions of $PM_{2.5}$ produced were only around 30%, 30%, and 30%– 50% in Beijing, Tianjin, and Hebei, respectively. The contribution of Shandong to southeastern Hebei and Tianjin reached 20–100 and 10–50 μ g m⁻³, respectively. This took 10%–30% of total PM_{2.5} at these two regions. Regional transport from Henan, Anhui, and Jiangsu affected southern and central Hebei and even Tianjin, with a contribution of 10–75 μ g m⁻³ (10%–30%). Due to the block of Taihang Mountain, Shanxi only affected western Hebei and southwest Beijing with a contribution of 20–50 μ g m⁻³ (10%–



Figure 7 Distribution of PM_{2.5} produced in different source areas during 10th–13th January 2013 (contours: %).

30%). Within HBT city cluster, Hebei significantly affected Beijing and Tianjin by the transport, with a contribution of 30–50 μ g m⁻³ (30%–50%), while Beijing and Tianjin only affected their boundary regions with Hebei (10–20 μ g m⁻³, <10%)

2.2 The impact of two-way feedback between aerosol and boundary-layer on haze formation

During this heavy haze period, the high aerosol loading was greatly lowered the surface temperature by reducing surface insolation caused by scattering and absorption of sunlight in the atmosphere. Meanwhile, absorption of solar radiation by black carbon and other absorbing aerosols resulted in regional heating of the upper part of boundary layer. This change in the vertical air temperature led to the enhancement of atmosphere stability of surface layer which conduced to the further accumulation of pollutants. This positive feedback mechanism between atmospheric boundary layer and pollution variation may be a very important factor to the formation of severe pollution. In this study, an on-line air quality model (NAQPMS and WRFv3.5) was developed to investigate the impact of this feedback mechanism between boundary layer physics and aerosols on the meteorological factors and pollutants concentration during January 2013. Two experiments were conducted: one without (w/o feedback) and one (w feedback) with this feedback mechanism.

Figure 8 shows the observed and two simulated (w feedback and w/o feedback) surface shortwave radiations and temperatures at the meteorological tower in Beijing. In the simulation of w/o feedback, the modeling values by WRF were much higher than the observation values in both surface shortwave radiation and temperature, while their difference had been greatly diminished in the w feedback simulation. The result indicates aerosols significantly altered solar radiation, temperature, and boundary layer altitude during the haze period. Note that the overestimation of surface shortwave radiation and temperature in w feedback may be caused by the error of cloud simulation.

Two simulated monthly mean $PM_{2.5}$ in HBT and their difference were shown in Figure 9. In the w/o feedback simulation, a great underestimation was found in heavy polluted cities like Beijing, Shijiazhuang, Baoding, Hengshui, and Cangzhou (Figure 9(a)). $PM_{2.5}$ concentrations in w feedback simulation were much closer to observations. Compared with those in w/o feedback, monthly $PM_{2.5}$ in w feedback simulation increased by 10%–30%, which even reached 40 µg m⁻³ in Shijiazhuang, Baoding, and Beijing. Hourly $PM_{2.5}$ increased 200 µg m⁻³ when they exceeded $300 µg m^{-3}$ (Figure 10). This contrast between two experiments suggested the two-way feedback mechanism between aerosols and boundary layer was a non-ignorable cause to formation and evolution of severe pollution.

2.3 The impact of regional transport on haze prevention and control in HTB

As shown in Figure 6 and Table 1, regional transport played an important role in $PM_{2.5}$ concentration variation in HTB. Therefore, it is necessary to consider the impact of regional transport for optimized reduction of emission gross of atmospheric pollutants. In this study, an optimization model is operated based on the following formula:

$$C_{i} = \sum_{j=1}^{N} (1 - A_{j}) Q_{ij} , \qquad (1)$$

$$C_i \leq C_{\text{target}}$$
, (2)



Figure 8 Observed (green lines) and simulated hourly surface shortwave radiation (a) and 2 m temperature (b) at the meteorological tower of IAP. Red and blue represent the simulated results in w/o and w feedback experiment, respectively.



Figure 9 The simulated monthly average concentrations of $PM_{2.5}$ in HBT. (a) w feedback experiment; (b) w/o two-way feedback experiment; (c) difference between w and w/o feedback experiments; (d) similar as (c), but in relative values. The solid cycles and their color refer to location of observation sites and observed values.



Figure 10 The scatter plot of simulated $PM_{2.5}$ hourly concentration (μg m⁻³) in w and w/o feedback experiments at different sites. BJ: Beijing; SJZ: Zhijiazhuang; BD: Baoding; CZ: Cangzhou; HS: Hengshui.

where *i* and *j* refer to the *i*th observation site and the *j*th PM_{2.5} geographical source region and the *j*th grid cell observation, respectively. C_i and Q_{ij} are mass concentration of

PM_{2.5} and contribution of the *j*th PM_{2.5} production region at the *i*th station. Here A_j is reducing coefficient for emissions of the *j*th PM_{2.5} production region, which ranges from 0 (no reduction) to 100% (all emissions are cut off); C_{target} stands for target concentration and it is 75 µg m⁻³ (Grade II) in this study. This model assumed that the reducing percentage of pollution source equals to the reducing percentage of its contribution to other areas, which might bring errors. Note that, though eq. (2) is a restraint, it allows the existence of multiple solutions to A_j in eq. (1). Hence, more than one solution can be found to meet the Air Quality Standard. This model is designed to analyze the feasible variation range of A_i (based on all the reducing plans).

Table 2 shows all the possible ranges of $PM_{2.5}$ reducing coefficient at Beijing, Tianjin and Hebei for meeting the Air Quality Standard for Grade II in various scenarios. Clearly, the $PM_{2.5}$ emissions would have to be reduced by above 90%, 90% and 60% in Hebei, Tianjin and Beijing respectively, when HBT surrounding provinces (Henan, Shandong, Anhui and Inner Mongolia) did not take any control measures. When the surrounding reduced 20% emissions, the reducing coefficient would be above 75% for Hebei, 65% for Tianjin, and 5% for Beijing. And when the surrounding reducing coefficient is above 50%, 55% reduction in Hebei is already enough to meet the standard.

 Table 2
 The reducing coefficients ranges of emissions in different areas

 for meeting Air Quality Standard for Grade II in different scenarios

Surrounding areas ^{a)}	Hebei	Tianjin ^{b)}	Beijing ^{b)}	
	20%	-	_	
0	55%	-	_	
0	75%	-	-	
	90%	90%-100%	60%-100%	
	20%	_	-	
20	55%	_	-	
20	75%	90%-100%	70%-100%	
	90%	65%-100%	5%-100%	
	20%	_	-	
50	55%	85%-100%	65%-100%	
50	75%	15%-100%	0-100%	
	90%	0-100%	0-100%	
	20%	_	-	
70	55%	35%-100%	0-100%	
/0	75%	0-100%	100%	
	90%	0-100%	0-100%	

a) Surrounding areas refer to Henan, Anhui, Jiangsu, Shandong, and Inner Mongolia. b) "–" means that $PM_{2.5}$ in HBT still exceeded Air Quality Standard for Grade II, even if its emissions were completely cut off.

3 Conclusions

The stable boundary layer and weak cold air masses caused by the rare historical weather condition over the North China Plain in January 2013 prevented pollutants from being diffused into free atmosphere. This is favorable for the formation of regional severe pollution in January 2013.

Regional transport contributed a lot to monthly $PM_{2.5}$ concentration during the severe haze in January 2013 over HTB. At typical stations (Beijing, Tianjin, Qinhuangdao and Cangzhou), the contribution of trans-cluster transport to $PM_{2.5}$ concentrations ranged from 20% to 35%, and the contributions of transport inner HBT ranged from 26% to 35%. The total transport contribution (trans-cluster transport plus inner transport) was comparable to the self-contribution. Regional transport was more important in $PM_{2.5}$ concentration change.

By the comparison of the two simulation experiments, aerosol-boundary layer feedback mechanism is found to significantly affect the accuracy of simulation on meteorological factors and $PM_{2.5}$ concentrations in HBT, particularly in severe pollution cases. For a better understanding of evolution of severe haze in the future, further analysis is needed on the physical and chemical characteristics of boundary layer and its feedback mechanism with pollutants.

For a better prevention and control of atmospheric pollution of HTB, more attention has to be paid to not only the inner region but also the city clusters outside. Without control actions to be taken to the surroundings, $PM_{2.5}$ over HTB region can meet the requirement of national Grade II on $PM_{2.5}$ only if the reducing coefficient reaches 90% for Hebei and Tianjin and 60% for Beijing.

This study was supported by the CAS Strategic Priority Research Program (Grant Nos. XDB05030200 and XDB05030101) and the National Natural Science Foundation of China (Grant No. 41278138). We also thank the observation network "CARE-China".

- Chang D, Song Y, Liu B. 2009. Visibility trends in six megacities in China 1973–2007. Atmos Res, 94: 161–167
- Donkelaar A, Martin R V, Brauer M, et al. 2010. Global estimates of ambient fine particulate matter concentrations from satellite-based aerosol optical depth: Development and application. Environ Health Perspect, 118: 847–855
- Grewe V. 2004. Technical Note: A diagnostic for ozone contributions of various NO_x emissions in multi-decadal chemistry-climate model simulations. Atmos Chem Phys, 4: 729–736
- Guenther A, Hewitt N, Erickson D, et al. 1995. A global model of natural volatile organic compound emissions. J Geophys Res, 100: 8873–8892
- Huang J, Wu D, Huang J H, et al. 2008. Visibility variations in the Pearl River Delta of China during the period of 1954–2004 (in Chinese). J Appl Meteoro Sci, 19: 61–70
- Kurokawa J, Ohara T, Morikawa T, et al. 2013. Emissions of air pollutants and greenhouse gases over Asian regions during 2000–2008: Regional Emission inventory in ASia (REAS) version 2. Atmos Chem Phys Discuss, 13: 10049–10123
- Li J, Wang Z F, Wang X Q, et al. 2011. Impacts of aerosols on summertime tropospheric photolysis frequencies and photochemistry over Central Eastern China. Atmos Environ, 45: 1817–1829
- Li J, Wang Z F, Zhuang G, et al. 2012. Mixing of Asian mineral dust with anthropogenic pollutants over East Asia: A model cast study of a super-duststorm in March 2010. Atmos Chem Phys, 12: 7591–7607
- Li J, Wang Z, Wu Q. 2010a. A Study of the quantitative diagnosis for the regional transport of tropospheric O₃ concentrations(in Chinese). Clim Environ Res, 15: 529–540
- Li L, Li C Y, Tan Y K, et al. 2010b. Stratospheric sudden warming impacts on the weather/climate in China and its role in the influences of ENSO (in Chinese). Chin J Geophys, 53: 1529–1542
- Li J, Wang Z, Akimoto H, et al. 2008. Near-ground ozone source attributions and outflow in central eastern China during MTX2006. Atmos Chem Phys, 8: 7335–7351
- Shao M, Tang X Y. 2006. City clusters in China: Air and surface water pollution. Front Ecol Environ, 4: 353–361
- Wang Z, Maeda T, Hayashi M, et al. 2001. A nested air quality prediction modeling system for urban and regional scales: Application for highozone episode in Taiwan. Water Air Soil Pollut, 130: 391–396
- Wang X, Yang T, Wang Z. 2011. Impact of dust haze episode from one air pollution control region to the other-one case study (in Chinese). Clim Environ Res, 16: 690–696
- Wu D. 2012. Hazy weather research in China in the last decade: A review (in Chinese). J Environ Sci, 32: 257–269
- Wu Q Z, Wang Z F, Gbaguidi A, et al. 2011. Numerical study of contributions to air pollution in Bei jing during CAREBeijing-2006. Atmos Chem Phys, 11: 5997–6011
- Wu Q Z, Wang Z F, Xu W S, et al. 2010. Multi-model simulation of PM10 during the 2008 Beijing Olympic Games: Effectiveness of emission restrictions (in Chinese). J Environ Sci, 30: 1739–1748
- Zhang X Y, Sun J Y, Wang Y Q, et al. 2013. Factors contributing to haze and fog in China. Chin Sci Bull, 58: 1178–1187
- Zhang J K, Sun Y, Liu Z R, et al. 2013. Characterization of submicron aerosols during a serious pollution month in Beijing (2013) using an aerodyne high-resolution aerosol mass spectrometer. Atmos Chem Phys Discuss, 13: 19009–19049