Numerical Simulation Analysis on the Sources of Haze over Zhongshan City in southern China Using WRF-CMAQ Model

Jianhua Mai[1,2], Tao Deng[2], Lingling Yu[3], Xuejiao Deng[2], Haobo Tan[2], Xiantong Liu[2]
1 Zhongshan Meteorological Service, Zhongshan 528400, China
2 Institute of Tropical and Marine Meteorology/Guangdong Provincial Key Laboratory of Regional Numerical Weather Prediction, Guangzhou, China Meteorological Administration, Guangzhou 510080, China
3 Guangdong Meteorological Observatory, Guangzhou 510080, China

Corresponding Author: Tao Deng
Institution: Institute of Tropical and Marine Meteorology/Guangdong Provincial Key Laboratory of Regional Numerical Weather Prediction, Guangzhou, China
Address: 6th of Meihua Road, Yuexiu District, Guangzhou, China

E-mail: tdeng@grmc.gov.cn

Abstract: Until recently few study about sources of haze and evaluation of emission control measures over Pearl River Delta (PRD) in China could be found but that is very important to the control of haze. So the WRF-CMAQ model was adopted to simulate the haze pollution of Zhongshan in January 2014 to study the haze sources of Zhongshan. The model could simulate the variation of $PM_{2.5}$ concentration and visibility in Zhongshan well. When there was no cold front, weak cold front or strong cold front affected, the contribution ratios of local sources in Zhongshan were 58%, 34% ,28%; sources of other cities in PRD were 27%, 42%, 23%; and 13%, 21%, 45% for the sources outside Guangdong Province, respectively. Among the major components of aerosol, local emission sources in Zhongshan contributed most to the nitrates, whereas emission sources outside Guangdong contributed most to other substances. According to the emission control tests, $PM_{2.5}$ concentration was reduced by 47%, 52% and 58%, respectively, on the first, second and third day after turning off the emission sources of the whole Zhongshan region under stable weather conditions, and reduced by 24%, 22% and 15%, respectively, in the high pollution peak by turning off industrial, residential and transportation sources.

Keywords: WRF-CMAQ, Haze, Contribution ratio, Emission control tests

1. Introduction

Haze is a common atmospheric turbidity phenomenon known as “Dust-haze” [1], in which a large amount of ultrafine dry dust particles are uniformly suspended in the air, causing horizontal visibility less than 10km. Along with the rapid development of the economy and massive emissions of atmospheric pollutants, haze is tending to become more and more intense in urban agglomerations throughout China[2], and its hazard to manufacture, transportation and human health has already aroused extensive attention[3-4]. Haze is principally caused by the powder and the sulfur dioxide emitted by the combustion of fossil fuels, as well as the huge amount of nitric oxides and volatile organic compounds emitted by automobile; all these substances, through chemical reactions, generate the secondary aerosols that lead to atmospheric turbidity[5]. The study of haze can be further summarized as the study of atmospheric aerosols to a great extent.

As one of the three principal urban agglomerations in China, the PRD (Pearl River Delta) agglomeration is inevitably accompanied by increasingly deteriorating air quality as its economy rapidly develops. Research on haze phenomena have already aroused the highest level of attention of the Chinese government and masses[6-10]. Lots of scholars, based on huge observational data, focused on studying the composition of the haze, its weather characteristics and meteorological effects[11-14]. As computer technology has developed, numerical model has become an important tool[15] of conducting atmospheric environmental research. For example, the Hysplit trajectory model is mostly used to study the trajectory of airflow affect certain areas[16-17]. And the CMAQ model, well known as a numerical tool for environment researching, is often adopted to study pollution events happened over China[18-19]. Currently, quite a number of studies on the basis of numerical models are focused on the occurrence, evolution and influence of individual instances in the atmospheric pollution process[20-22]; however, few focus on the sources of atmospheric aerosols in the PRD region and the contribution ratio of emission sources in various regions under different weather conditions. Nevertheless, researches on these aspects are significant to the control of haze, and must be urgently undertaken.

Zhongshan City, as an important part of PRD, has also been affected by the frequent haze pollution in recent years. In 2014, the Zhongshan government invested in 10 projects to improve people's living; one project of which, known as haze forecast, prevention and control, aimed to greatly improve the local air quality. Accordingly, the study on the characteristics of haze in Zhongshan is especially important. A hazy day is defined as a day in which the average visibility is less than 10km and the average relative humidity is 90% or lower[23]. In January 2014, there were 16 hazy days and the haze pollution in this month was the most significant of the year. Currently there are a few of studies on the haze pollution happened in China in January 2014[24-25], but this paper mainly focused on PRD. By taking advantage of the observational meteorological and environmental data, and then integrate the data with the WRF-CMAQ model, a simulation analysis was performed about haze pollution in January 2014 over Zhongshan so as to study the contribution of emission sources from different areas to the $PM_{2.5}$ concentration and evaluate the emission control measures of local Zhongshan. The study results were aimed to be references for haze control for the government.

2. Research methods

2.1 Brief introduction and setting of the numerical model

This paper adopted the WRF-CMAQ model, which was used in a lot of numerical studies on air pollution events[26-27], to perform numerical simulation (using WRFv3.3 and CMAQv5.0) and the NCEP fnl reanalysis data as the initial field in the WRF model, which provided detailed meteorological data for CMAQ. Simulation tests adopted a two-way nested method. As was shown in Table 1 and Figure 1, the exterior grids of the atmospheric chemistry transport model covered most regions of China, whereas the interior grids covered the whole of Guangdong Province. The location of cities in PRD was showed in Figure 1 too. In this study, the simulation period was from 08:00 LST (local standard time) 25 Dec. 2013 to 08:00 LST 01 Feb. 2014, and the days before 08:00 LST 01 Jan. 2014 were for model spin-up.

Table 1. Setting of the simulation test zone.

Regions under simulation	Number of grid points	Grid space(km)
WRF Model Domain_1	283×184	27
CMAQ Model Domain_1	182×138	27
WRF Model Domain_2	233×163	9
CMAQ Model Domain_2	98×74	9

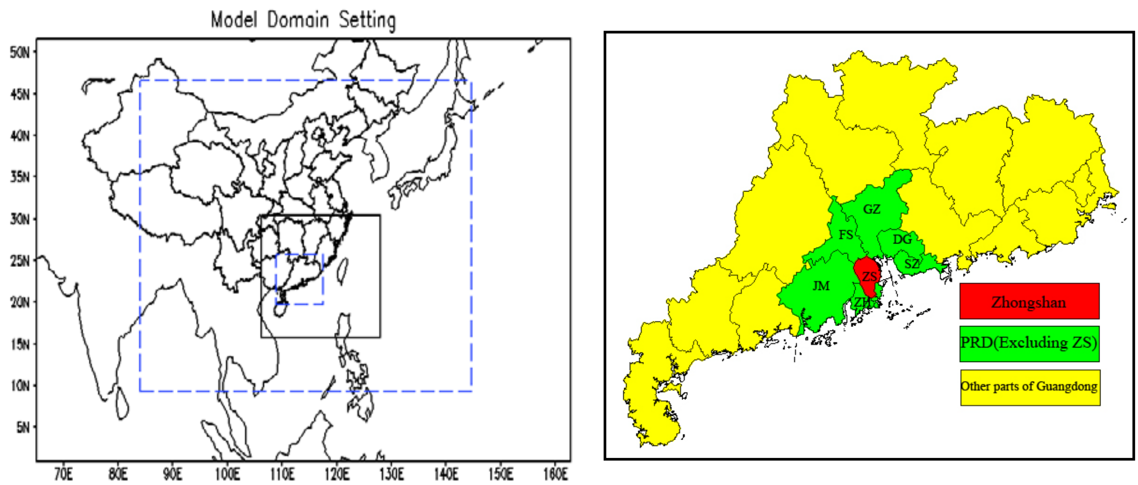


Figure 1. Model's domain setting and different parts of emission sources (in the left picture, the black solid line represented the WRF model; the dotted blue line represented the CMAQ model; the right picture showed different parts of emission

sources in sensitivity tests and details would be given in Section2.2). (Cities of PRD in the right picture: Zhongshan(ZS), Guangzhou(GZ), Foshan(FS), Jiangmen(JM), Zhuhai(ZH), Dongguan(DG), Shenzhen(SZ)).

Community Multi-scale Air Quality (CMAQ), representing the latest research results for the current mainstream atmospheric environment and transmission, diffusion and sedimentation of pollutants, is a multi-scale Euler air quality model. It can simulate the physical and chemical process of multiple pollutants in the atmosphere, including ozone, aerosol and various pollutant gases. The CMAQ model must be driven by the results of meteorological models and the emission source data varying with time and space. It comprehensively considers the meteorological and chemical process of the gaseous and liquid phases, heterogeneous chemical processes, the aerosol process and the dry-wet deposition process, and thence can better simulate the contribution of various aerosols to light extinction and atmospheric visibility, as well as the occurrence, duration and disappearance of haze phenomena.

As an important, integral part of the air quality model, emission sources affect simulation accuracy. Many scholars contributed to the development of the emission inventory [28-29]. The emission source data adopted by this paper came from the 2010 MEIC (Multi-resolution Emission Inventory for China) developed by Tsinghua University, where the spatial resolution was 0.25°×0.25°. The MEIC divided emission sources into five categories, *i.e.*: power plants, industry, agriculture, transportation and resident, covering the monthly emissions[30] of pollutant gases such as SO_2, NO_X, CO and NH_3, aerosol such as $PM_{2.5}$ and PM_{10}, as well as black carbon, organic carbon and other substances. Figure 2 showed the annual emission of NO2, NO, SO2 and CO of Guangdong province from the MEIC dataset. It could find that the PRD region, especially Guangzhou, Foshan and Shenzhen are regions with the most concentrated pollutant emissions, discharging more than 500×10^6mol of CO、NO, and 100×10^6mol of SO_2、NO_2 per year. Also the temporal variations of the MEIC dataset were set by referring to the study of Zheng[31]. By processing the emission source data and entering the results into the atmospheric chemistry transport model, emission source data with time-varying and identical spatial resolution could be provided for the CMAQ model.

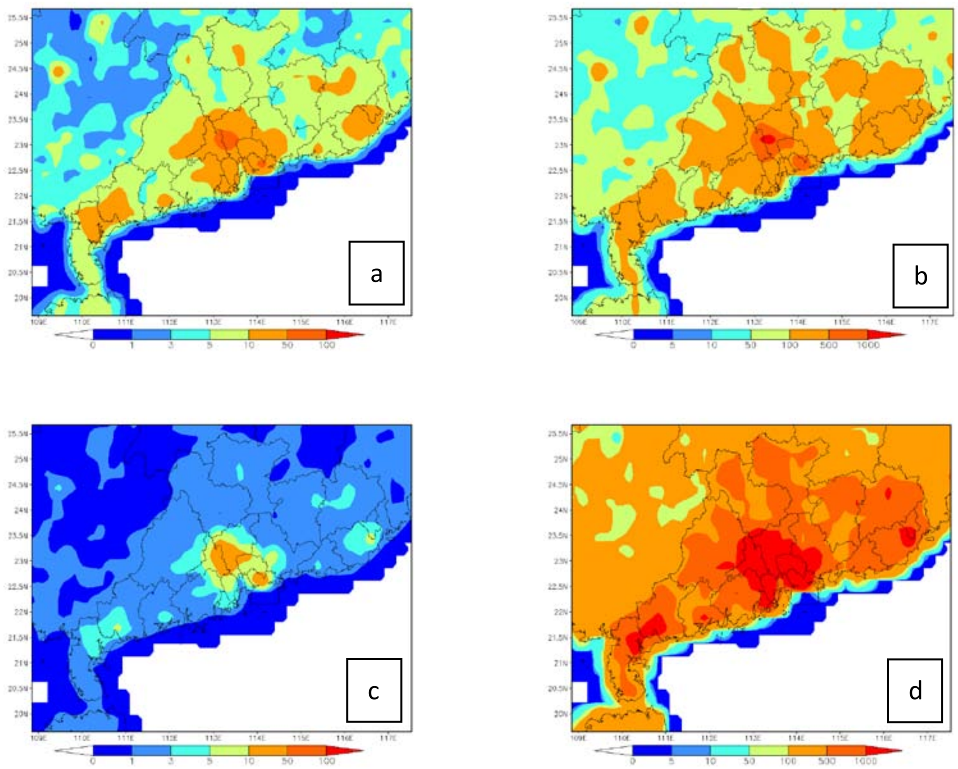


Figure 2. Annual emission of NO_2 (a), NO (b), SO_2 (c) and CO (d) of Guangdong Province from MEIC dataset (Unit:$\times 10^6$mol/year).

2.2 Sensitivity tests and contribution ratio calculation

In order to survey the influence of emission sources inside and outside Zhongshan to the local haze pollution, the paper used the WRF-CMAQ model to perform sensitivity tests and study the contribution of emission sources from different regions to the $PM_{2.5}$ concentration in Zhongshan. The principal test consideration was as follows: divide emission sources into different parts, turn off the emission sources from a certain region, and then compare the simulation results before and after emission shielding, at last calculate the contribution ratio of the emission sources of this region to the local $PM_{2.5}$ in Zhongshan. The research method was proved to be effective in some studies on the air quality during 2008 Beijing Olympic Game[32-33]. The specific test schemes and different parts of emission sources were shown in Figure 1 and Table 2(emission sources outside Guangdong Province were not showed here). Among the tests, Test_ctr, as the control test, represented the benchmark conditions of emission sources; in Test_zs the local emission sources of Zhongshan were turned off; in Test_prd the emission sources of other cities in the PRD region were turned off so as to evaluate the influence of emission sources from surrounding regions on Zhongshan; in Test_gd the emission sources outside Guangdong Province were turned off. By excluding the influences from local Zhongshan, the PRD and those from outside Guangdong, the remaining influences were from other parts of Guangdong (see Figure 1). The contribution ratio of emission sources of various regions could be calculated by the following formula:

$$C_x = C_{ctr} - C_{x,0} \qquad (1)$$

$$P_x = C_x / C_{ctr} \qquad (2)$$

where C_x and P_x represented the concentration change before and after emission sources were turned off in the region x and the contribution ratio of emission sources of region x to the local atmospheric pollution of Zhongshan, respectively; C_{ctr} and $C_{x,0}$ represented the $PM_{2.5}$ concentration of Zhongshan under the benchmark condition and the condition that the emissions from the region x were set to zero, respectively.

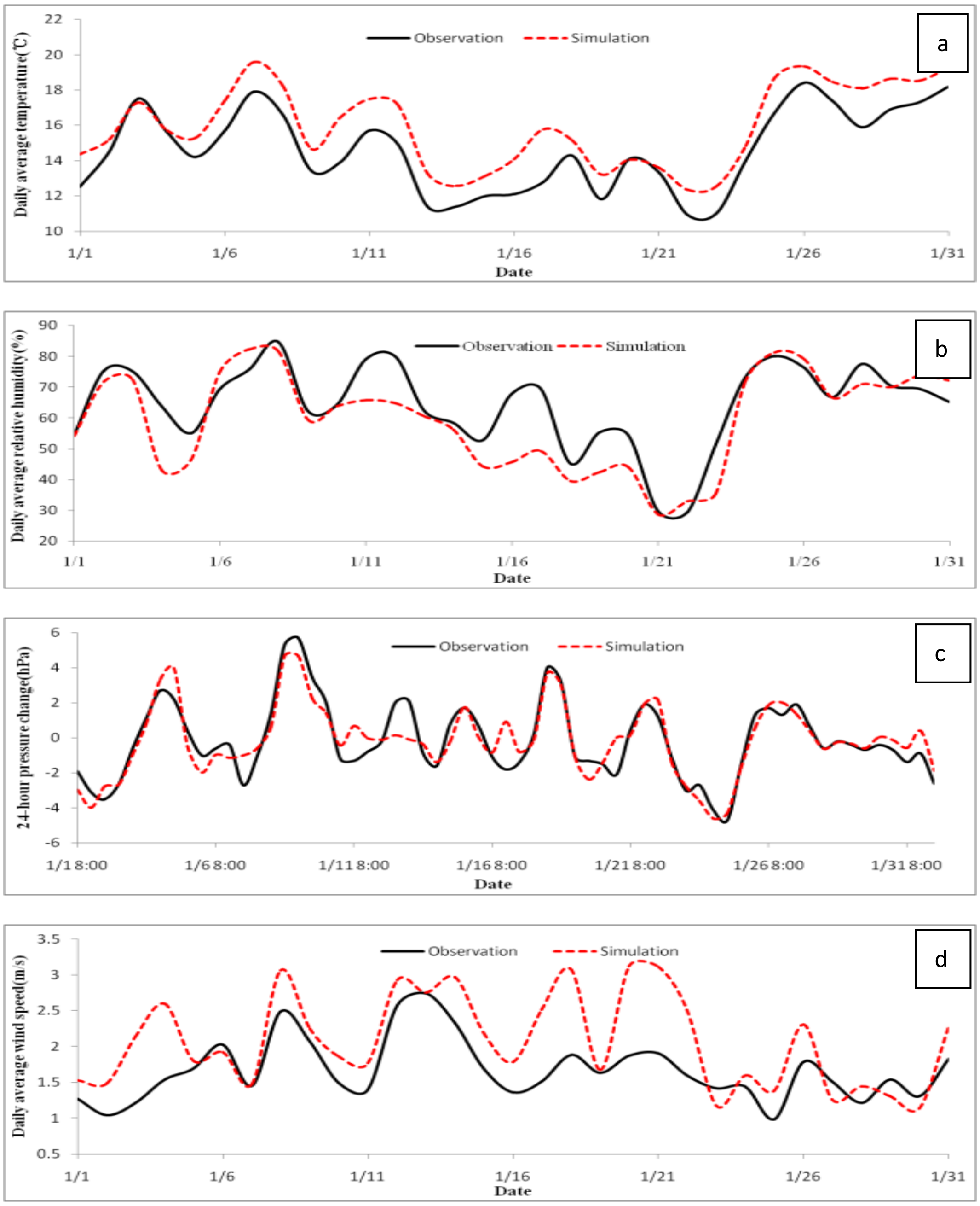
Table 2.Simulation scheme of sensitivity tests.

Test name	Emission source to be turned off	Test name	Emission source to be turned off
Test_ctr	None	Test_zs	Zhongshan
Test_prd	PRD (excluding Zhongshan)	Test_gd	Outside Guangdong

3. Control test results

3.1 Evaluation of meteorological simulation results

The simulation accuracy of the air quality model depends on the simulation results of the meteorological field to a large extent; therefore the WRF model simulation results should be first evaluated. The observational data used in this paper are from Zhongshan Meteorological Service(113.35° E，22.53° N). In January 2014, Zhongshan was frequently affected by cold front but there was no significant precipitation in the whole month. Among the 16 hazy days in January, 12 of them were caused by cold front and the remaining 4 hazy days were happened under stable weather condition with low wind speed and stable atmospheric stratification. Figure 3 showed the daily average 2m temperature, daily average relative humidity, 24-hour pressure variation, daily average wind speed and daily maximum frequency wind direction simulated by the WRF model, as well as the observational data. It was find that the simulation elements were close to the observations. The simulated 2m temperature was slightly higher than the actual temperature but the temporal variation fits the actual data well. The relative humidity was a little lower than the actual conditions, and the 24-hour pressure variation, which can represent the movement of the cold front, was basically equal to the actual conditions. From the variations of temperature and pressure we could know that there were six cooling periods in Zhongshan caused by cold front, namely 3-5, 8-9, 12-13, 18-19, 21-22 and 26-27 January. The simulated wind speed was stronger than the observational data, especially in the days affected by cold front (e.g. 8, 18 and 21 January), but the tendency of the daily variation also fit the actual data well. From the comparison of the wind direction we could found that in January 2014 Zhongshan was dominated by northerly, northeasterly and southeasterly wind, and the WRF model gave an excellent performance on wind direction in most of the time. Such results revealed that the WRF model obtained an accurate simulation of the weather conditions in January 2014. Therefore the simulation results of the meteorological model were reliable and could provide accurate and detailed meteorological field data for the CMAQ model.



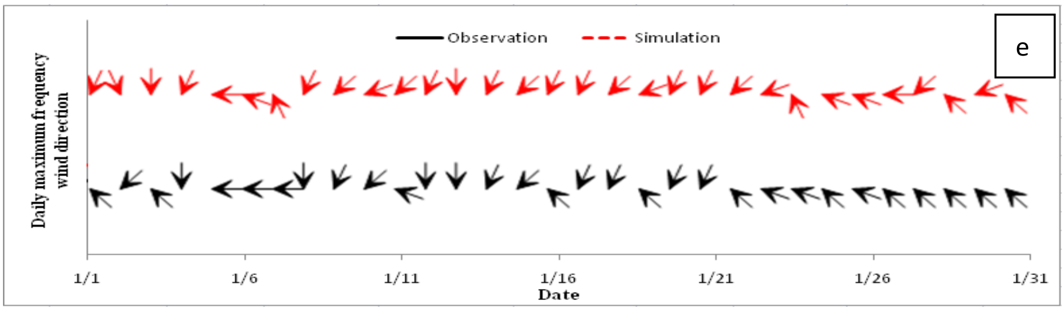


Figure 3 Comparison of simulated 2m temperature (a), relative humidity (b), 24-hour pressure variation(c), wind speed (d) and wind direction (e) with observational data.

3.2 Evaluation of CMAQ model results

Figure 4 is the comparison between the simulated $PM_{2.5}$ concentration and visibility in Zhongshan in January 2014 and the observational data. In general the model could accurately simulate the temporal variation of $PM_{2.5}$ concentration; however the emission sources in the model could not completely illustrate the actual discharge of air pollutants, besides simulation errors existed in the results of the WRF model, so during certain time periods the CMAQ simulation deviated. When there was no cold front affected, wind speed under the meteorological model simulation was generally stronger than observation, so the simulated $PM_{2.5}$ concentration was usually lower than the actual data, for instance on 5 and 23 January. When strong cold front moved southward, it transferred a large amount of air pollutants to the downwind areas. If the cold front is strong enough, the pollutants can reach a longer distance. So the weak simulation of cold front (simulated 24-hour pressure variation weaker than actual conditions) may result in the weak advection transfer of extraneous pollutants, and further reduce simulation concentration, for instance on 12 January. Due to the fact that it was approaching the lunar New Year (Chinese New Year) from 28 to the 31 January, pollutant emissions were obviously less than the usual. However, the emission sources adopted by the model did not consider such factors, causing an obvious difference between the actual pollutant emission and the model simulation. Besides stable weather conditions were available without cold front affected during these days, leading to a higher simulation concentration than the observation. But a PM2.5 concentration peak appeared at about 08:00 on 31 January for the burning of firecrackers, which is usual during the Chinese New Year, and the model failed to capture this variation. On the aspect of visibility simulation, the model could basically simulate the variation tendency. However, as was mentioned above, when no cold front affected, the $PM_{2.5}$ concentration from the model is usually lower than the observational data, causing higher simulated visibility than the actual one. But from 28 to 31 January the simulated visibility is lower than the observation because the simulated $PM_{2.5}$ concentration is overestimated. The difference was mostly obvious when the actual or the simulated visibility reached their peak values. Since the simulated $PM_{2.5}$ concentration was lower than the actual conditions in general, the simulated visibility was higher than the actual conditions most of the time.

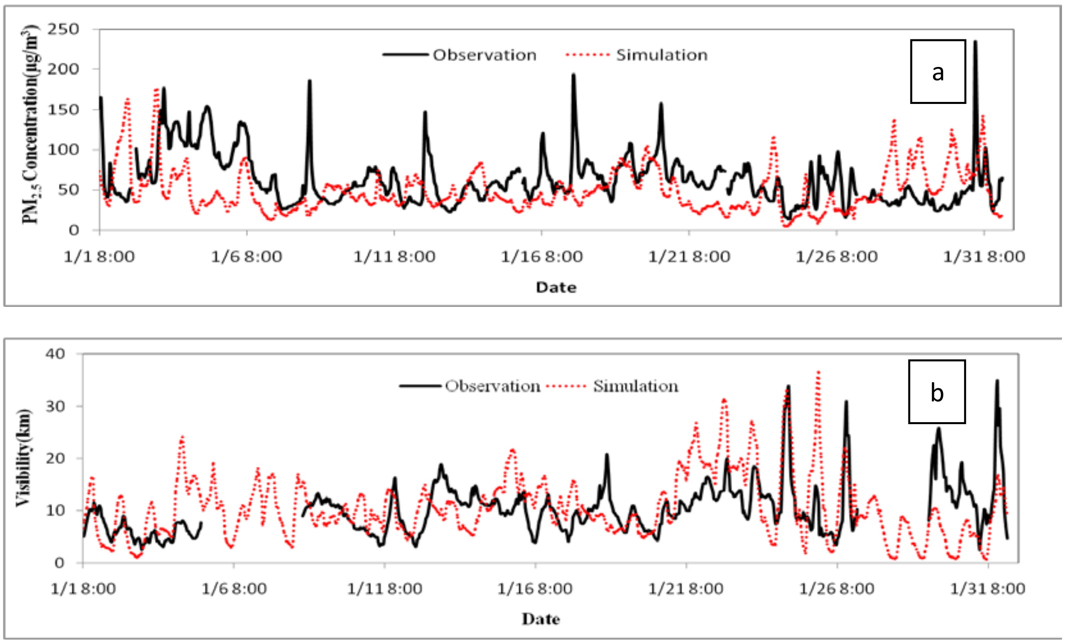


Figure 4. Comparison of simulated $PM_{2.5}$ concentration (a) and visibility (b) with observational data (the discontinuities showed missing values).

Table 3 showed the simulation error of $PM_{2.5}$ concentration and visibility from CMAQ model. Statistical metrics including average absolute bias (MB), average absolute error (ME), average root mean square error (RMSE), average normal deviation (MNB) and normal average deviation (NMB). The calculation methods were defined as follows:

$$MB = \frac{1}{n}\sum_{i=1}^{n}\left(Sim(i) - Obs(i)\right) \quad (1)$$

$$ME = \frac{1}{n}\sum_{i=1}^{n}\left|Sim(i) - Obs(i)\right| \quad (2)$$

$$RMSE = \left[\frac{1}{n}\sum_{i=1}^{n}(Sim(i) - Obs(i))^2\right]^{1/2} \quad (3)$$

$$MNB = \frac{1}{n}\sum_{i=1}^{n}\left(\frac{Sim\,(i) - Obs\,(i)}{Obs\,(i)}\right) \quad (4)$$

$$NMB = \frac{\sum_{i=1}^{n}(Sim\,(i) - Obs\,(i))}{\sum_{i=1}^{n} Obs\,(i)} \quad (5)$$

where Sim and Obs represented simulated and observed values, respectively. From the table it could see that the $PM_{2.5}$ concentration from the model is lower than the observation, while the simulated visibility is a little overestimated. But in general the differences between simulation and observation were not evident, which indicated that the simulation results were reliable.

Table 3. Simulation error of $PM_{2.5}$ concentration and visibility with CMAQ model.

	MB	ME	RMSE	MNB	NMB
$PM_{2.5}$ Concertration	-13	32	29	-4%	-21%
Visibility	0.8	4.9	5.6	8%	20%

4. Sensitivity analysis

4.1 Analysis of contribution ratio

Figure 5a showed the spatial distribution of average monthly $PM_{2.5}$ concentration simulated by CMAQ. A relatively high $PM_{2.5}$ polluted area was found in the middle of Foshan, where the average $PM_{2.5}$ concentration exceeded 60μg/m^3. $PM_{2.5}$ concentration in the northwest of Zhongshan was 50-55μg/m^3, while only 35-40μg/m^3 in the southern part, indicating that the $PM_{2.5}$ concentration from northwestern to southeastern of Zhongshan was gradually reduced. Figure 5b was the contribution ratio of emission sources of local Zhongshan. The contribution ratio reached 45% in the northwest of Zhongshan but reduced to 30-35% in the southern area. A part of areas of Jiangmen and Zhuhai, which located in the downwind direction of Zhongshan, were also affected by the emission sources of Zhongshan and the contribution ratio was 20-25%. Figure 5c gave the contribution ratio of emission sources of other cities in PRD. Evidently the contribution ratio in the middle and southeast of Foshan exceeded 60%. Contribution ratio in southern Guangzhou, western Dongguan and Shenzhen, and eastern Jiangmen also reached 50% or more. These parts of emission sources contributed 35-45% $PM_{2.5}$ in northern and 25-30% in southern Zhongshan. The emission sources outside Guangdong Province were very important to the entire areas of the PRD (Figure 5d). These parts of emission sources contributed relatively less nearby the southeast of Foshan, but the contribution ratio gradually increased in other parts of PRD. To Zhongshan, emission sources outside Guangdong contributed about 25% in the northern part but the contribution ratio increased to 35-40% in southern Zhongshan. From the discussion above it could find that emission sources of local Zhongshan, other cities in PRD and outside Guangdong Province were all very important to the $PM_{2.5}$ of Zhongshan.

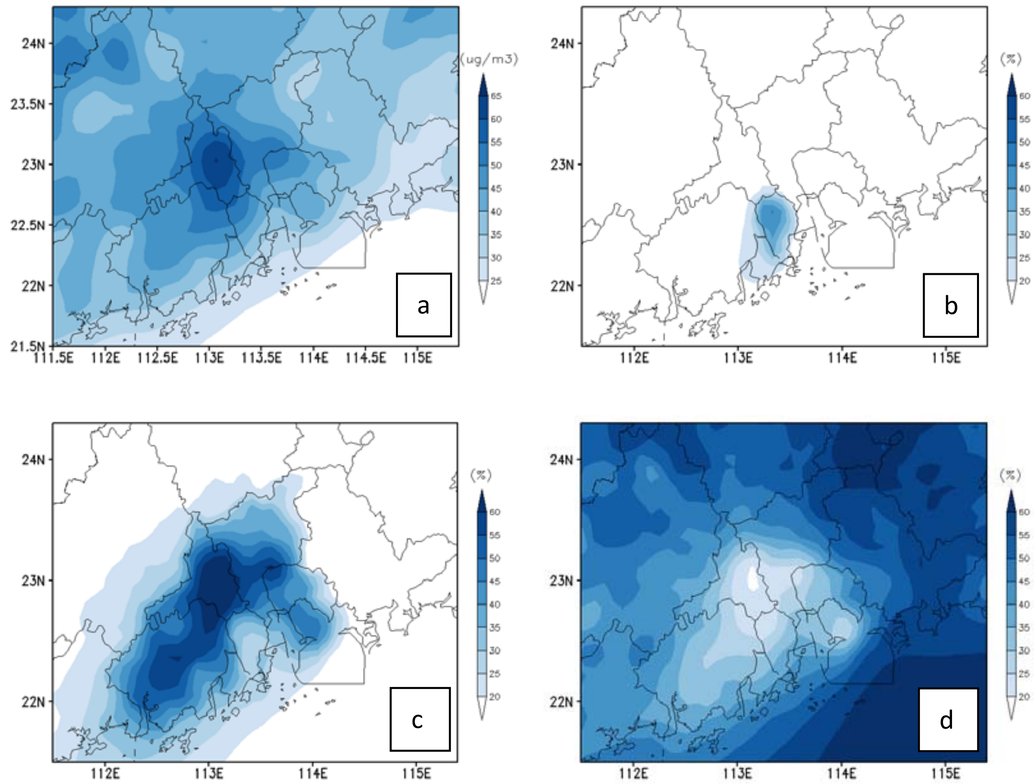


Figure 5. Space distribution of simulated monthly average $PM_{2.5}$ concentration (a) and the contribution ratio of emission sources of Zhongshan (b), PRD (c) and emission sources outside Guangdong (d).

By summarizing test results and selecting haze pollution processes generated under different weather conditions, the contribution ratio of every region to the $PM_{2.5}$ concentration of Zhongshan under different weather situations could be obtained (see Figure 6). When there was no cold front affected, haze pollution was mostly caused by low wind speed and stable atmospheric stratification. Such weather condition was not beneficial for the long-distance transfer of air pollutants; so the contribution ratio of local emission sources of Zhongshan was the highest (58%), and contribution ratio of emission sources of other nearby cities in the PRD region and emission outside Guangdong were 27% and 13%, respectively. When strong cold front went southwards and affected Zhongshan, a large amount of non-local pollutants from regions beyond Guangdong Province were transferred to the PRD region along with the cold front, causing the contribution ratio of emission sources outside Guangdong Province to hugely increase (45% in this case), following by that of local emission sources of Zhongshan (28%) and emission sources of other cities in PRD (23%). When weak cold front was available, it could enhance the horizontal transfer of pollutants to a certain extent, but such transfer was mainly dominated by the short-distance transfer so the contribution ratio of emission sources of other cities in

PRD to Zhongshan was highest (42%), following by that of local emission sources of Zhongshan (34%), and the contribution ratio of emission sources outside Guangdong Province was also increased when compared to no cold front case, 21% at this time.

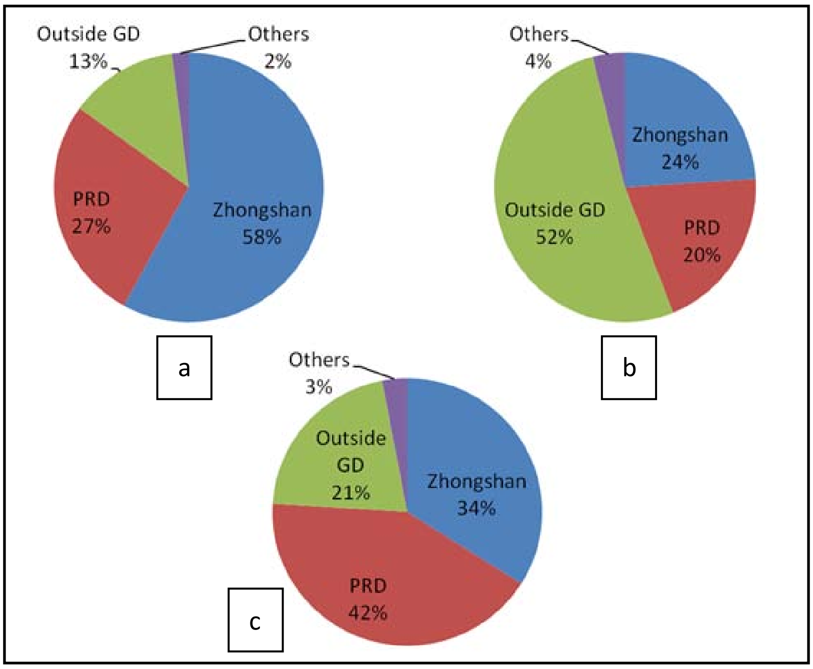


Figure 6. Contribution ratio of emission sources of various regions under different weather conditions to $PM_{2.5}$ concentration of Zhongshan (a: no cold front; b: strong cold front; c: weak cold front).

From the results above it could be found that under different weather conditions, emission sources in every region contributed differently to the haze pollution of Zhongshan; emission sources of local Zhongshan and surrounding cities dominated when there was no cold front and only weak cold front available; however emission sources outside Guangdong Province contributed maximally when there was strong cold front affected. Additionally, the simulated wind speed was higher than actual conditions, especially when cold front affected, which might overestimate the cross-region transfer of air pollutants to some extent, and might weaken the pollution generated in the low wind speed cases. Therefore, the model results might overestimate the contribution ratio of non-local emission sources, but underestimate the local one.

As was showed in Table 4, it could be seen that the local $PM_{2.5}$ concentration of Zhongshan decreased and the visibility increased after turning off emission sources of certain regions. When no cold front was available and the local emission sources of Zhongshan were turned off, the visibility was most significantly improved by up to 90%; when weak cold front was available and the emission sources of other cities in PRD were turned off, the visibility was improved by 76%; when strong cold front was affecting and the emission sources outside Guangdong Province were turned off, the visibility was improved by 84%. The visibility did not increase in a linear way as the $PM_{2.5}$ concentration decreased; the percentage of increment of visibility might be equal to the decrement of $PM_{2.5}$ concentration when $PM_{2.5}$ decreased in a low level, but might be far higher than the decrement of $PM_{2.5}$ concentration when $PM_{2.5}$ decreased greatly.

Table 4. Variation of $PM_{2.5}$ concentration and visibility in sensitivity tests.

Weather conditions	No cold front	Weak cold front	Strong cold front
Test content	Turn off emission sources of Zhongshan		
$PM_{2.5}$ decrement (%)	58	34	28
Visibility increment (%)	94	39	30
Test content	Turn off emission sources of other cities in PRD		
$PM_{2.5}$ decrement (%)	27	42	23
Visibility increment (%)	30	76	27
Test content	Turn off emission sources outside Guangdong		
$PM_{2.5}$ decrement (%)	13	21	45
Visibility increment (%)	15	23	84

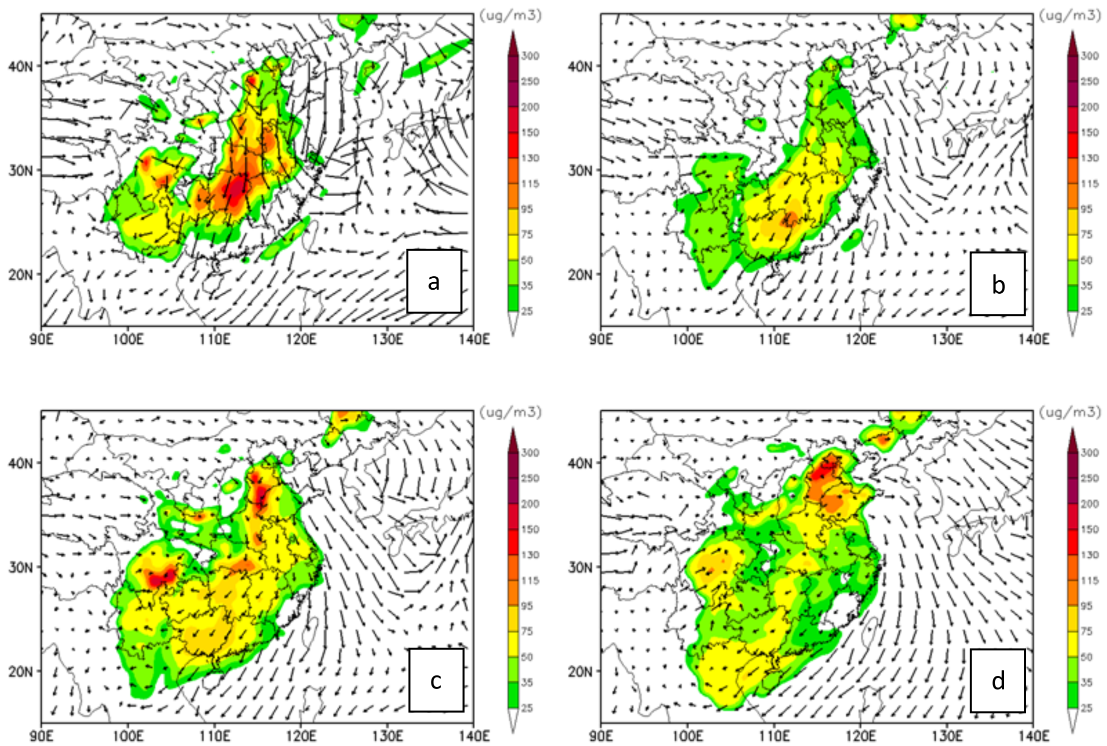


Figure 7. Schematic Diagram for transfer of $PM_{2.5}$ by cold front (vector for 10m wind field; shade for $PM_{2.5}$ concentration) (a: 01-08 08:00; b: 01-08 17:00; c: 01-08 23:00; d: 01-09 17:00).

According to the analysis above, it could be seen that emission sources outside

Guangdong Province contributed maximally to the local $PM_{2.5}$ concentration of Zhongshan when strong cold front affected. Figure 7 showed the transfer process of $PM_{2.5}$ occurring on 8-9 January. From the figure it could be found that the highest $PM_{2.5}$ concentration was located in Hunan Province on 8 January; as the cold front moved southwards, the aerosols were transferred southwards accordingly, and the $PM_{2.5}$ concentration in northern Guangdong increased significantly. On the night of 8 January, a large amount of aerosols were transferred to the PRD region and the whole western Guangdong by the cold front. By the afternoon of 9 January, the $PM_{2.5}$ concentration in the PRD region was significantly decreased. Meanwhile, due to the diffusion, sedimentation and aging, $PM_{2.5}$ concentration might decrease to some extent when pollutants were transferred southwards. In other cold front processes, similar transfer was also available; certainly and apparently, air pollutants outside Guangdong Province might cause a larger influence on the local haze weather over Zhongshan in winter with frequent cold front.

4.2 Analysis of the contribution ratio of emission sources in different regions to the primary composition of aerosols in Zhongshan

Sulfate, nitrate, ammonium salt, organic carbon and elemental carbon are the main components of atmospheric aerosols. Table 5 showed the contribution ratio of emission sources in different regions to the primary composition of atmospheric aerosols in Zhongshan in January 2014. The sulfate in the atmosphere of Zhongshan was mainly provided by emission sources outside Guangdong (56%), followed by that of emission sources in other PRD cities (25%) and local emission sources in Zhongshan (15%). Sulfate is mainly from various chemical reactions of SO_2 generated and discharged by large power plants and industries, however there is few large power plants or large industrial sources in Zhongshan, so non-local sources accounted for a high percentage. As for nitrate, NO_x discharged by motor vehicles and small and medium industries is the main precursor of nitrate, and nitrate easily volatilizes, making transfer difficult over long distances; therefore local emission sources of Zhongshan contributed the most (68%), and the contribution from emission sources in the other three regions was basically the same (10% approximately). Ammonium salt comes mainly from agricultural discharge. The emission sources inside Zhongshan and outside Guangdong contributed 36% and 39%, respectively, to ammonium salt, while the emission sources in other PRD cities contributed only 17%. As for organic carbon and elemental carbon, the emission sources outside Guangdong contributed the most (51% and 45%), followed by the local emission sources in Zhongshan (32% and 31%), and finally the emission sources in other PRD regions (11% and 19%). Obviously, the contribution of emission sources in various regions to the primary composition of aerosols is related to the industrial structure.

Table 5. Contribution ratio of emission sources in various regions to primary composition of atmospheric aerosol of Zhongshan.

Composition of aerosols	Zhongshan(%)	PRD (%)	Outside GD(%)	Others(%)
Sulfate	15	25	56	4
Nitrate	68	12	10	10
Ammonium salt	36	17	39	8
Organic carbon	32	11	51	6
Element carbon	31	19	45	5

4.3 Emission control analysis

This paper studied the impacts of various emission sources and emission control measures of Zhongshan on local $PM_{2.5}$ concentration and visibility. According to the analysis above, when strong cold front was affecting, the emission sources outside Guangdong contributed maximally to the local $PM_{2.5}$ concentration of Zhongshan. Under such weather conditions, emission control measures might not work perfectly. When there was no cold front, low wind speed and stable atmospheric stratification were available, the contribution of local emission sources in Zhongshan increased greatly; therefore the emission control analysis in this study was performed under such weather conditions. According to the specific emissions reduction plan, the emission sources of Zhongshan were firstly turned off so as to observe the change in $PM_{2.5}$ concentration and visibility within 1-3 days after emission control were taken; and next, industrial, residential and transportation sources within the emission sources were respectively turned off to evaluate the impacts of different emission sources on $PM_{2.5}$ concentration and visibility.

Table 6. Variation of $PM_{2.5}$ concentration and visibility after turning off local emission sources of Zhongshan.

	$PM_{2.5}$ concentration decrement (%)	Visibility increment (%)
1st day	47	57
2nd day	52	63
3rd day	58	90

According to Table 6, when under stable weather conditions, $PM_{2.5}$ concentration decreased by 47%, 52% and 58% on the first, second and third day respectively after turning off the local emission sources of Zhongshan; emission control efficiency apparently increased with the lapse of time. Corresponding to the increasing reduction of $PM_{2.5}$ concentration, local visibility improved by 57%, 63% and 90% respectively on the first, second and third day. By the way, all results shown in Table 6 were obtained in ideal conditions of turning off emission sources for the whole Zhongshan region. However, it is impossible to create such conditions when making actual emission control measures, but emission control can only be made for partial emission sources; therefore emission control was suggested to be implemented 2-3 days before hazy days to reduce $PM_{2.5}$ by 50%.

Industrial, residential and transportation sources contributed significantly to local $PM_{2.5}$ concentration; so those three parts of emission sources in Zhongshan were turned off so as to observe the variation of $PM_{2.5}$ concentration and visibility during high pollution periods (average value of 5 hours before and after $PM_{2.5}$ concentration reached peak value was used here); such high pollution periods occurred on the third day after emission control started. It could be seen from Table 7 that the emission sources which contributed the most to $PM_{2.5}$ concentration during the time period were industrial sources, and then residential and transportation sources. By turning off these three parts of emission sources, $PM_{2.5}$ concentration was reduced by 24%, 22% and 15%, and the visibility increased by 26%, 21% and 16%, respectively. Therefore, in conditions without an influence from a significant weather system, the government could consider emission control measures from the perspectives of industrial, residential and transportation sources, for instance, restricting industrial production and reducing emissions of high pollution enterprises, adopting odd-and-even license plate regulations for motor vehicles, promoting a green lifestyle and reducing energy consumption so as to obtain an ideal effect.

Table 7. Variation of $PM_{2.5}$ concentration and visibility during high pollution periods after turning off industrial, residential and transportation sources of Zhongshan.

Sources to be turned off	$PM_{2.5}$ decrement (%)	Visibility increment (%)
Industrial sources	24	26
Residential sources	22	21
Transportation sources	15	16

5. Conclusions

Research about the sources of haze pollution and evaluation on the emission control measures are important to the control of haze, but few study about that over PRD can be found until recently. So the haze pollution over Zhongshan in January 2014 was simulated using the WRF-CMAQ model, and the main conclusions reached, which could be used as a reference for haze prevention and control for government, were as follows:

The simulation results based on CMAQ model basically reflected the variation tendencies of the $PM_{2.5}$ concentration and visibility in Zhongshan. $PM_{2.5}$

concentration of Zhongshan gradually reduced from northwestern to southeastern, with 50-55μg/m^3 in the northern part and 35-40μg/m^3 in the southern part. The contribution ratio of emission sources in local Zhongshan reached 45% in the northwest of Zhongshan but reduced to 30-35% in the southern area. Emission sources of other cities in RPD contributed 35-45% $PM_{2.5}$ in northern and 25-30% in southern Zhongshan. The emission sources outside Guangdong Province were very important to Zhongshan, which contributed about 25% in the northern part but the contribution ratio increased to 35-40% in southern Zhongshan.

The simulation tests showed that emission sources in Zhongshan, other PRD cities and outside Guangdong respectively contributed 58%, 27% and 13% when there was no cold front affected. However when strong cold front was available, a large amount of pollutants outside Guangdong Province were transferred to the PRD region via the advection airflow, making the contribution ratio of emission sources outside Guangdong increased to 45%, while the emission sources in Zhongshan and other PRD cities contributed respectively 28% and 23%; when weak cold front affected, emission sources of other PRD cities contributed 42%, and emission sources in Zhongshan and outside Guangdong respectively contributed 34% and 21%.

Among the primary compositions of atmospheric aerosols of Zhongshan, Sulfate came mainly from emission sources outside Guangdong, which accounted for 56%. As for nitrate, local Zhongshan emission sources contributed the most at 68%. Ammonium salt came from emission sources inside Zhongshan and outside Guangdong (36% and 39%, respectively). As for organic carbon and elemental carbon, emission sources outside Guangdong contributed the most (51% and 45%), followed by the contribution of local emission sources of Zhongshan (32% and 31%).

After turning off the local emission sources of Zhongshan, the $PM_{2.5}$ concentration decreased by 47%, 52% and 58% on the first, second and third day, so emission control was suggested to be implemented 2-3 days before hazy days to reduce $PM_{2.5}$ by 50%. According to the emission control simulation analysis, the contribution ratio of emission sources in Zhongshan mostly came from industrial sources, followed by residential and transportation sources. After turning off industrial, residential and transportation sources, $PM_{2.5}$ concentration decreased by 24%, 22% and 15%, respectively. So the conclusions came that in conditions without a significant weather system, the government could consider emission control measures from the perspectives of industrial, residential and transportation sources so as to obtain an ideal effect.

Acknowledgments

This study was supported by the Key Projects in the National Science & Technology Pilar Program(2014BAC16B06), Project supported by the National

Natural Science Foundation of China(41205123), Science & Technology Research Project of Guangdong Meteorological Service, China(2014B32). Also, the emission source inventory used in this paper is supplied by Center For Earth System Science from Tsinghua University, China. The authors would like to thank all people for their great work which helped to improve this manuscript.

Conflict of Interest

The authors declare that there is no conflict of interest regarding the publication of this article.

References

[1]Weather forecast manual of Guangdong, China Meteorological Press, Beijing, China, 2009.

[2]D. Wu, "Hazy weather research in China in the last decade: A review", Acta Scientiae Circumstantiae, vol. 32, no. 2, pp. 257-269, 2012.

[3]X. X. Tie, D. Wu, and B. Guy, "Lung cancer mortality and exposure to atmospheric aerosol particles in Guangzhou, China", Atmospheric Environment, vol. 43, no. 14, pp. 2375-2377, 2009.

[4]J. E. Penner, X. Q. Dong, and Y. Chen, "Observational evidence of a change in radiative forcing due to the indirect aerosol effect", Nature, vol. 427, pp. 231-234, 2004.

[5]H. H. Chen, D. Wu, H. B. Tan, F. Li, and S. J. Fan, "Study on the character of haze weather process from the year 2001 to 2008 over the Pearl River Delta", Journal of Tropical Meteorology, vol. 26, no. 2, pp. 147-155, 2010.

[6]Z. Wei, L. T. Wang, S. M. Ma, F. F. Zhang, and J. Yang, "Source contributions of $PM_{2.5}$ in the severe haze episode in Hebei cities", The Scientific World Journal, vol. 2015, Article ID 480542, 11 pages, 2015.

[7]R. Zhang, G. Sarwar, J. C. H. Fung, A. K. H. Lau, and Y. H. Zhang, "Examining the impact of nitrous acid chemistry on ozone and PM over the Pearl River Delta Region", Advances in Meteorology, vol. 2012, Article ID 140932, 18 pages, 2012.

[8]Y. C. Li, J. Z. Yu, S. H. H. Steven, Z. B. Yuan, A. K. H. Lau, and X. F. Huang, "Chemical characteristics of $PM_{2.5}$ and organic aerosol source analysis during cold front episodes in Hong Kong, China", Atmospheric Research, vol. 118, pp.41-51, 2012.

[9]P. F. Liu, C. S. Zhao, T. Gobel, E Hallbauer, N. Andreas, and L. Ran, "Hygroscopic properties of aerosol particles at high relative humidity and their diurnal variations in the North China Plain", Atmospheric Chemistry and Physics, vol. 11, pp. 3479-3494, 2011.

[10]D. Wu, X. X. Tie, and X. J. Deng, "Chemical characterizations of soluble aerosols in Southern China", Chemosphere, vol. 64, no. 5, pp. 749-757, 2006.

[11]J. H. Tan, S. J. Guo, Y. L. Ma, J. Duan, Y. Cheng, K. He, and F. Yang, "Characteristics of particulate PAHs during a typical haze episode in Guangzhou, China", Atmospheric Research, vol. 102, pp. 91-98, 2011.

[12]D. Wu, J. T. Mao, X. J. Deng, X. X. Tie, Y. H. Zhang, L. M. Zeng, F. Li, H. B. Tan, and X. Y. Bi, “Black carbon aerosols and their radiative properties in the Pearl River Delta region”, Science in China Series D: Earth Sciences, vol. 52, no. 8, pp. 1152-1163, 2009.

[13]H. J. Xu, X. M. Wang, U. Poschl, S. L. Feng, D. Wu, Y. Ling, S. X. Li, W. Song, G. Y. Sheng, and J. M. Fu, “Genotoxicity of total and fractionated extractable organic matters in fine air particulate matter ($PM_{2.5}$) in unban Guangzhou： comparison between haze and non-haze episode days”, Environmental Toxicology and Chemistry, vol. 27, no. 1, pp. 206-212, 2008.

[14]H. Yu, C. Wu, D. Wu, and J. Z. Yu, “Size distributions of elemental carbon and its contribution to light extinction in urban and rural locations in the Pearl River Delta, China”, Atmospheric Chemistry and Physics, vol. 10, no. 12, pp. 5107-5119, 2010.

[15]A. P. Kesarkar, M. Dalvi, A. Kaginalkar, and A. Ojha, “Coupling of the weather research and forecasting model with AERMOD for pollutant dispersion modeling: A case study for PM_{10} dispersion over Pune, India”, Atmospheric Environment, vol. 41,no. 9, pp. 1976-1988, 2007.

[16]P. Pongkiatkul, and N. T. K. Oanh, “Assessment of potential long-range transport of particulate air pollution using trajectory modeling and monitoring data”, Atmospheric Research, vol. 85, pp.3-17, 2007.

[17]W. P. Shan, Y. Q. Yin, H. X. Lu, and S. X. Liang, “A meteorological analysis of ozone episodes using HYSPLIT model and surface data”, Atmospheric Research, vol. 93, pp.767-776, 2009.

[18]Pu. Zhang, C. C. Meng, J. Su, Z. Wei, F. F. Zhang, W. Wei, and X. J. Zhao, “Quantifying the sources of the severe haze over the southern Hebei using the CMAQ model”, The Scientific World Journal, vol. 2013, Article ID 812469, 9 Pages, 2013.

[19]R. Zhang, S. Golam, C. H. F Jimmy and K. H. L. Alexis, “Role of photoexcited nitrogen dioxide chemistry on ozone formation and emission control strategy over the Pearl River Delta, China”, Atmospheric Research, vol. 132-133, pp. 332-344, 2013.

[20]Q. Z. Wu, Z. F. Wang, and A. Gbaguidi,, “Numerical study of contributions to air pollution in Beijing during CAREBeijing-2006”, Atmospheric Chemistry and Physics, vol. 41, pp. 5997-6011, 2011.

[21]J. S. Fu, D. G. Streets, C. J. Jang, H. Jiming, and K. B. He, “Modeling regional/urban ozone and particulate matter in Beijing, China”, Journal of the Air & Waste Management Association, vol. 59, pp. 37-44, 2009.

[22]X. S. Wang, Y. H. Zhang, Y. T. Hu, W. zhou, L. M. Zeng, M. Hu, D. S. Cohan, and A. G. Russell, “Decoupled direct sensitivity analysis of regional ozone pollution over the Pearl River Delta during the PRIDE-PRD 2004campaign”, Atmospheric Environment, vol. 45, no. 28, pp. 4941-4949, 2011.

[23]D. Wu, H. Z. Chen, M. Wu, B. T. Liao, and Y. C. Wang, “Comparison of three statistical methods on calculating haze days-taking areas around the capital for example”, China Environmental Science, vol. 34, no. 3, pp. 545-554, 2014.

[24]H. Geng, Y. Y. Xuan, X. T. Cai, Y. Zhang, H. Zhou, and Z. H. Zhang, 2015. “Mass

concentration variation and cluster analysis of urban air pollutants in Taiyuan, Shanxi Province during Chinese New Year of 2014", Acta Scientiae Circumstantiae, vol. 35, no. 4, pp. 965-974, 2015.

[25]Z. R. Liu, B. Hu, D. S. Ji, Y. H. Wang, M. X. Wang, and Y. S. Wang, "Diurnal and seasonal variation of the $PM_{2.5}$ apparent particle density in Beijing, China", Atmospheric Environment, vol.120, pp. 328-338, 2015.

[26]S. Yu, R. Mathur, J. Pleim, D. Wong, R. Gilliam, K. Alapaty, C Zhao, and X. Liu, "Aerosol indirect effect on the grid-scale clouds in the two-way coupled WRF-CMAQ: model description, development, evaluation and regional analysis", Atmospheric Chemistry and Physics, vol. 14, no. 20, pp. 11247-11285, 2014.

[27]M. T. Cabaraban, C. N. Kroll, S. Hirabayashi and D. J. Nowak, "Modeling of air pollutant removal by dry deposition to urban trees using a WRF/CMAQ/i-Tree Eco coupled system", Environmental Pollution, vol. 176c, no. 5, pp. 123-133, 2013.

[28]Q. Zhang, D. G. Streets, G. R, Carmichael, K. B. He, H. Huo, A. Kannari, Z. Klimont, I. S. Park, S. Reddy, J. S. Fu, D. Chen, L. Duan, Y. Lei, L. T. Wang, and Z. L. Yao, "Asian emissions in 2006 for NASA INTEX-B mission", Atmospheric Chemistry and Physics, vol. 9, 5131-5153, 2009.

[29]Q. Zhang, G. N. Geng, S. W. Wang, A. Richter, and K. B. He, "Satellite remote sensing of changes in NO(x) emissions over China during 1996-2010", Chinese Science Bulletin, vol. 57, pp. 2857-2864, 2012.

[30]K. B. He, "Multi-resolution Emission Inventory for China (MEIC): model framework and 1990-30 2010 anthropogenic emissions", International Global Atmospheric Chemistry Conference, 2012.

[31]J. Y. Zheng, Z. Y. Zheng, Z. L. Wang, L. J. Zhong, and D. Wu, "Biogenic VOCs emission inventory and its temporal and spatial characteristics in the Pearl River Delta area", China Environmental Science, vol.29, no. 4, pp. 345-350, 2009.

[32]Wang, L. T. Wang, J. M. Hao, He, K. B. He, S. X. Wang, J. H. Li, Q. Zhang, D. G. Streets, J. S. Fu, C. J. Jang, H. Takekawa, and S. Chatani , "A modeling study of coarse particulate matter pollution in Beijing: region source contributions and control implications for the 2008 Summer Olympics", Journal of Air and Waste Management Association, vol. 58, pp. 1057-1069, 2008.

[33]J. Xing, Y. Zhang, S. X. Wang, X. H. Liu, S. H. Cheng, Q. Zhang, Y. S. Chen, D. G. Streets, C. Jang, J. M. Hao, and W. X. Wang, "Modeling study on the air quality impacts from emission reductions and a typical meteorological conditions during the 2008 Beijing Olympics", Atmospheric Environment, vol. 45, pp. 1786-1798, 2011.