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ORIGINAL PAPER

Residential emissions predicted as a major source of fine particulate matter in winter over the Yangtze River Delta, China

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Abstract

Air pollution is an increasingly critical health issue responsible for numerous diseases and deaths worldwide. In China, to address severe air pollution in the Yangtze River Delta region, the local government has formulated Five-Year Plans to set the road map for air pollution control by phased targets in 2020, but the effectiveness of these policies is still uncertain. There is therefore a need for accurate prediction of control strategies. Here we present a computational evaluation of the predicted effectiveness of four emission control strategies: normal or enhanced emission reduction for industry and power plants, and normal or enhanced emission reduction for industry, power plants and transportation, designed on the basis of policies of the 13th Five-Year Plans. Effectiveness was tested on concentrations of $PM_{2.5}$, e.g., particulate matter with aerodynamic diameter less than 2.5 µm, using the two-way coupled Weather Research and Forecasting—Community Multiscale Air Quality (WRF-CMAQ) model. Results show that by implementing the four emission control strategies, only Hangzhou with the strictest emission controls in four main cities (Hangzhou, Hefei, Nanjing and Shanghai) can meet the 20% reduction goals of $PM_{2.5}$ concentrations in the 13th Five-Year Plan, indicating that current policies are not sufficient to control the severe air pollution in the Yangtze River Delta region. Sensitivity tests show that residential emissions have the highest contributions to the $PM_{2.5}$ concentrations in January in the four main cities of Hangzhou, Hefei, Nanjing and Shanghai, followed by agriculture, industry, transportation and power plants. Predicted annual mean reduction percentages for $PM_{2.5}$ are the highest in Hangzhou, from − 9.7 to − 20.1%, followed by Nanjing, from − 8.2 to − 18.7%, Shanghai, from − 7.4 to − 15.8%, and Hefei, from − 6.1 to − 13.8%. This finding highlights the predominance of residential emissions, which should be better controlled, notably coal burning. By comparison, predicted annual contributions of regional transport and natural sources to mean $PM_{2.5}$ concentrations in four cities range from 29.2 to 36.6%. Overall, a major finding is that residential sources are of comparable importance to industrial, power plant and transportation sources to $PM_{2.5}$ concentrations, especially for winter. This information will help governments of other regions of China, as well as other developing countries, to formulate more appropriate emission control strategies where coal is used for heating and cooking purposes in the developing countries.

Keywords Emission reductions · WRF-CMAQ · Scenario analysis · Yangtze River Delta

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Introduction

Following rapid urbanization and industrialization over the past few decades, severe haze episodes occur in the economically developed, highly industrialized and densely populated areas in China, such as Beijing–Tianjin–Hebei, Yangtze River Delta and Pearl River Delta (Zhao et al. 2013; Yan et al. 2015; Lu et al. 2016; Huang et al. 2016; Hong et al. 2016; Yu et al. 2014a, b; Mehmood et al. 2018). Elevated concentrations of $PM_{2.5}$–particles with aerodynamic diameter less than 2.5 µm—contribute to a "haze day", which is defined as one with visibility lower than 10 km under conditions of 80% relative humidity (Huang et al. 2012; Lu et al. 2016). The Yangtze River Delta is one of the largest city clusters in China, including Shanghai municipality, and Jiangsu, Anhui and Zhejiang provinces, accounting for 24% of China's Gross Domestic Product while occupying only 4% of the land area (National Bureau of Statistics of China 2014). The measured 2013 annual mean $PM_{2.5}$ concentration in the Yangtze River Delta was 67 µg m^{-3} according to the Ministry of Environmental Protection of China (2014). This level is almost twice the secondary standard in China's National Ambient Air Quality Standard, of 35 µg m^{-3} for $PM_{2.5}$ (NAAQS, standard GB3095-2012). To address this serious $PM_{2.5}$ pollution problem, the Chinese government issued the Action Plan on Prevention and Control of Air Pollution in September of 2013, with the goal of reducing $PM_{2.5}$ concentrations by 20% over the Yangtze River Delta by 2017 relative to the 2012 levels (State Council of China 2013). Evaluating the effectiveness of the emission control strategies is thus essential.

To design effective emission control strategies, critical information is needed about the contributions of different sources and regions. Investigation of the impacts of emission control policies on air quality using computational atmospheric models is essential (Xing et al. 2011; Gao and Zhang et al. 2012; Sun et al. 2016; Liu et al. 2016; Li et al. 2017). For instance, the Community Multiscale Air Quality model system (Eder and Yu 2006) has been widely applied to predict the effects of emission control measures on air quality in China, especially for several large international events such as the 2008 Beijing Olympics (Xing et al. 2011; Gao and Zhang et al. 2012), the 2014 Beijing Asia–Pacific Economic Cooperation Summit, (Sun et al. 2016; Liu et al. 2016), and the 2016 G20 Hangzhou Summit (Li et al. 2017).

Wang et al. (2014a) studied the contributions of different source sectors to $PM_{2.5}$ in southern Hebei during the 2013 severe episode by the Community Multiscale Air Quality model with a brute-force method (BFM), in which a series of sensitivity simulations were performed, each with one emission sector eliminated and the differences between the results from the sensitivity and baseline simulations being attributed to the emission eliminated (Burr et al. 2011). It was concluded that industrial and domestic sources contributed almost equally, 28 and 27%, respectively, to total $PM_{2.5}$ concentrations in Hebei Province.

Wang et al. (2014b) found that industrial and domestic sources contributed 58% and 16% of inorganic particulate matter in Xi'an in winter of 2013, respectively, using simulations from a source-oriented version of the Community Multiscale Air Quality model (CMAQ-PPM). Hu et al. (2015) found that residential emissions are the major contributor to primary particulate matter (30–70%) in winter/spring, and industrial emissions are more dominant in summer/fall with contributions of 40–60%, according to simulations with an updated version of the CMAQ model over China in a four-month study during 2012–2013. Hu et al. (2015) also found that residential emissions, industrial emissions and dust are three major source categories contributing to primary particulate matter in all seasons in Shanghai.

Here, we evaluate the impacts of different emission control scenario cases on haze formation over the Yangtze River Delta for the base year 2013. We use a two-way coupled Weather Research and Forecasting-Community Multiscale Air Quality (WRF-CMAQ) model to evaluate the impacts of different emission control strategies on haze formation in the Yangtze River Delta using 2013 as base year. The WRF-CMAQ model simulation periods, January, April, July and October, represent winter, spring, summer and autumn seasons, respectively. The performance of the WRF-CMAQ model is evaluated with available observations.

Experimental

Model descriptions and Observational data

The two-way coupled WRF-CMAQ modeling system (Wong et al. 2012; Yu et al. 2014a, b) is used to simulate the meteorological fields and air quality in the Yangtze River Delta (WRF, version 3.4, Skamarock et al. 2005; CMAQ, version 5.0.2, Eder and Yu 2006). Figure 1a shows the computational domain with a horizontal resolution of 12 km × 12 km covering most of China and a portion of East Asia. The model configurations used in this study are the same as those in Yu et al. (2014a, b) and are briefly described here. The physics of the WRF (ARW, e.g., Advanced Research WRF) includes the Morrison double-moment cloud microphysics scheme (Morrison et al. 2009), Rapid Radiative Transfer Model for General Circulation Models (RRTMG) shortwave and longwave radiation schemes, the Kain–Fritsch (KF2) cumulus cloud parameterization, the Pleim–Xiu (PX) land-surface scheme, and the asymmetric convective model (ACM2) for

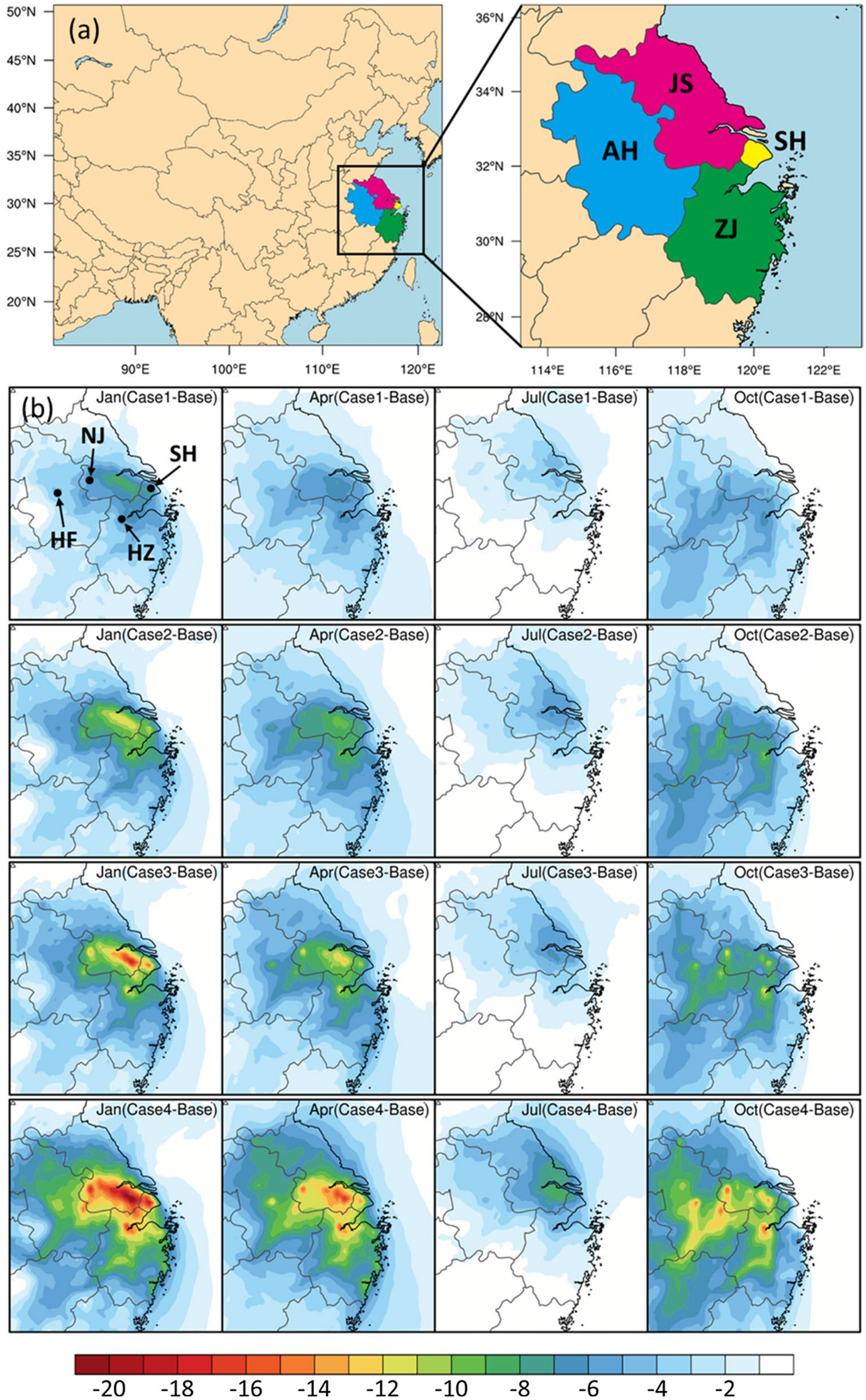

Fig. 1 **a** Map of Asia and China showing eastward the Yangtze River Delta region and districts Jiangsu (JS), Anhui (AH), Shanghai (SH) and Zhejiang (ZJ). **b** Spatial distributions of $PM_{2.5}$ concentration reduction amounts (μg m^{-3}) of four emission control scenarios relative to the baseline case over the Yangtze River Delta region in four months. Scenario Cases 1 and 2 are for general and enhanced emission reductions for both industrial and power plant emissions, respectively; Cases 3 and 4 are for the general and enhanced emission reductions for industrial, power plant and transportation emissions, respectively. Distributions reveal that reduction levels are similar for the same season, and cases with enhanced emission controls exhibit further reductions, while they are very different for different seasons. The highest reductions are predicted to occur in winter (January), the lowest in summer (July), while spring (April) and autumn (October) have comparable reduction levels. Most of the reductions are predicted to occur in south Jiangsu, north Zhejiang, east Anhui and Shanghai. *NJ* Nanjing, *SH* Shanghai, *HF* Hefei, *HZ* Hangzhou, J*an* January, *Apr* April, *Jul* July, *Oct* October

a planetary boundary layer (PBL) scheme (Pleim 2007). The carbon-bond chemical mechanism (CB05) (Yarwood et al. 2005) and AERO6 are the gas-phase chemistry mechanism and aerosol module, respectively. Biogenic emissions are calculated by the Model of Emissions of Gases and Aerosols from Nature (MEGAN v2.1) (Guenther et al. 2012).

Anthropogenic emissions of SO_2, NO_x, CO, NMVOC, NH_3, PM_{10} and $PM_{2.5}$ over China were generated by the Multi-resolution Emission Inventory for China (MEIC) (http://www.meicmodel.org) developed by Tsinghua University for 2012, while those for the rest of the domain were generated on the basis of Emissions Database for Global Atmospheric Research (EDGAR):HTAP V2. On the basis of the detailed source classifications, the MEIC emission data are aggregated to five sectors: agriculture, industry, power plants, residential and transportation. Note that there are three subsectors for the residential sector in MEIC, i.e., residential heating, residential combustion and residential solvent use, and waste emissions, were also assigned to residential sector. In MEIC, the residential sources were treated as nonpoint (area) sources. The nonpoint sources were allocated to each grid based on spatial proxies, such as urban or rural extents and population. Meteorological initial and lateral boundary conditions were derived from the National Center for Environmental Prediction (NCEP) final analysis dataset with a spatial resolution of 1°×1° and a temporal resolution of 6 h. The default chemical boundary conditions (BCONs) in the Community Multiscale Air Quality model were used in the simulations.

Hourly observed concentrations of $PM_{2.5}$, PM_{10}, NO_2, CO, SO_2 and O_3, at four cities (at 10, 10, 9 and 10 monitoring stations in Hangzhou, Hefei, Nanjing and Shanghai, respectively) in the Yangtze River Delta, obtained from the national air quality monitoring network operated and maintained by the Ministry of Environmental Protection (MEP) in China (http://datacenter.mep.gov.cn/), were used for evaluating the two-way coupled WRF-CMAQ model. Meteorological data (temperature, humidity) at a temporal resolution of 3 h in these four cities used for model evaluation were obtained from https://www.wunderground.com.

Emission control scenario cases description

In October 2012, the State Council of China issued the 12th Five-Year Plan for Air Pollution Prevention and Control in Key Regions, which directed 117 Chinese cities to achieve air quality improvements by 2015. In this 12th Five-Year Plan, the Beijing–Tianjin–Hebei region, the Pearl River Delta and the Yangtze River Delta were identified to make the most ambitious improvements, including a 6% reduction in $PM_{2.5}$ concentrations, a 12% reduction in SO_2, a 13% reduction in NO_x and a 10% reduction in industrial soot (State Council of China 2012). In September 2013, the State Council of China issued the first "Action Plan for Air Pollution Prevention and Control," named the "Action Plan", which requires all cities at the prefecture level and above to phase out inefficient coal boilers and achieve a 10% reduction in average annual PM_{10} level by 2017. In this Action Plan, eastern metropolitan areas are directed to achieve more ambitious improvements for $PM_{2.5}$ levels with 25, 20 and 15% reductions in the Beijing–Tianjin–Hebei area, the Yangtze River Delta and the Pearl River Delta, respectively (State Council of China 2013). Coal use should peak in those three regions by 2017, and construction of most new coal-fired power plants will be banned from 2017 onward.

In order to prevent and control air pollution, protect and improve the atmospheric environment, ensure public health, carry forward the construction of ecological civilization and promote the sustainable development of economy and society, the governments of Zhejiang (http://www.zjdpc.gov.cn/art/2017/4/28/art_90_1726404.html), Jiangsu (http://www.jsrd.gov.cn/zyfb/dffg1/201502/t20150202_156701.html) and Shanghai (http://www.shanghai.gov.cn/nw2/nw2314/nw2319/nw12344/u26aw50076.html) in the Yangtze River Delta region formulated their 13th Five-Year Plans for Air Pollution Prevention and Control (covering 2016–2020) on the basis of the national "Action Plan" and 13th Five-Year Plan for ecosystem and environmental protection (http://www.gov.cn/zhengce/content/2016-12/05/content_5143290.htm).

These 13th Five-Year Plans set the future road map for air pollution control by phased targets in 2020. We designed the four emission control scenario Cases 1, 2, 3 and 4—listed in Table 1 to assess the impact of different emission control policies on $PM_{2.5}$ concentrations in the Yangtze River Delta under the 13th Five-Year Plan. Scenario Cases 1 and 2 are designed for general and enhanced emission reductions for both industrial and power plant emissions, respectively, while scenario Cases 3 and 4 are designed for the general and enhanced emission reductions for three emission sectors industry, power plants and transportation, respectively. The corresponding reduction percentages for each species for each emission sector in Table 1 are derived on the basis of targets for air quality improvement and key air pollutant emission reductions and the optimization of energy and industrial structures for 2020 in the 13th Five-Year Plan.

To investigate contributions of different emission sectors to $PM_{2.5}$ concentrations over the Yangtze River Delta region, the brute-force method (BFM) (Wang et al. 2018), as described above, was employed in this study (Burr et al. 2011). Six sensitivity simulation scenarios were designed as listed in Table 1: no agriculture (Case 5), no industry (Case 6), no power plants (Case 7), no residential (Case 8), no transportation (Case 9) and no anthropogenic (Case 10), in which emissions from agriculture, industry, power plants, residential, transportation and all five anthropogenic sources were eliminated, respectively.

Table 1 Emission control percentages for each species and emission sectors for four different emission control scenarios 1, 2, 3 and 4 and six sensitivity tests 5, 6, 7, 8, 9 and 10 of anthropogenic sectors

Scenario Cases	Description	Emission sectors	Emission control percentage (%)					
			SO_2	NO_x	CO	VOCs	$PM_{2.5}$	PM_{Coarse}
1	General control	Industry	29%	18%	0%	34%	26%	27%
		Power plants	53%	44%	0%	10%	73%	73%
2	Enhanced control	Industry	45%	29%	0%	54%	37%	36%
		Power plants	65%	50%	0%	20%	83%	83%
3	General control	Industry	29%	18%	0%	34%	26%	27%
		Power plants	53%	44%	0%	10%	73%	73%
		Transportation	75%	25%	31%	32%	73%	72%
4	Enhanced control	Industry	45%	29%	0%	54%	37%	36%
		Power plants	65%	50%	0%	20%	83%	83%
		Transportation	94%	52%	50%	65%	81%	80%
5	No agriculture	Agricultural sources in the Yangtze River Delta region completely removed						
6	No industry	Industrial sources in the Yangtze River Delta region completely removed						
7	No power plants	Power plant sources in the Yangtze River Delta region completely removed						
8	No residential	Residential sources in the Yangtze River Delta region completely removed						
9	No transportation	Transportation sources in the Yangtze River Delta region completely removed						
10	No anthropogenic	All five anthropogenic sources in the Yangtze River Delta region completely removed						

VOC volatile organic compounds

Results and discussion

Model performance evaluation

Predicted concentrations of CO, NO_2, SO_2, O_3, $PM_{2.5}$ and PM_{10} versus hourly observations at four months and four cities are summarized in Table 2. Normalized mean bias values for $PM_{2.5}$ are within ± 15% for all months and cities except that the normalized mean bias value in Hefei for October is − 27%, while the normalized mean bias values for PM_{10} are within ± 30% for all months and cities, except that the normalized mean bias values in Nanjing for April and October are − 46 and − 37%, respectively (Table 2). The large bias associated with PM_{10} is mainly attributed to errors in modeling dust emissions, which contribute to a large faction of coarse particles (Fu et al. 2014). Normalized mean bias values for CO are within ± 20% for all months and cities except that the normalized mean bias value in Hefei for October is 33%, while the normalized mean bias values for SO_2 and NO_2 for all months and cities are within ± 30% and ± 20%, respectively (Table 2). Model performance for O_3 is characterized by normalized mean bias values between − 16 and 36% for all months and cities, except that the normalized mean bias values in Nanjing for July and Hefei for April are 43–45%, respectively.

In summary, the results shown in Table 2 demonstrate a reasonable skill in reproducing concentrations of all species, CO, NO_2, SO_2, O_3, $PM_{2.5}$ and PM_{10}, for the four simulation months for the baseline emission scenario over the Yangtze River Delta. As summarized in Table 2, the Weather Research and Forecasting model used in the present study generally reproduced the temperatures in the four cities very well, with correlation coefficients between 0.71 and 0.90, and normalized mean bias values within ± 9% for July and October. The model consistently exhibited a cold bias for temperatures in January and April, with normalized mean bias values between − 11 and − 33% and between − 1 and − 16%, respectively. The model captured the observed relative humidity at all months and cities with normalized mean bias values within ± 30% except that normalized mean bias values in Hefei for April and in Shanghai for July are − 36 and 39%, respectively.

Predicted influence of emission control scenarios on $PM_{2.5}$ concentrations

We assessed the potential improvement of air quality in the Yangtze River Delta associated with emission control policies. For that we simulated monthly mean $PM_{2.5}$ ground-level concentrations in four cities, specifically mean concentrations at 10, 10, 9 and 10 monitoring stations in Hangzhou,

Table 2 Evaluation of model performance on meteorology variables, temperature (T), relative humidity (RH) and six pollutants, CO, NO_2, SO_2, O_3, $PM_{2.5}$ and PM_{10}, in four cities over the Yangtze River Delta region for four months

		Hangzhou				Hefei				Nanjing				Shanghai			
		Jan	Apr	Jul	Oct	Jan	Apr	Jul	Oct	Jan	Apr	Jul	Oct	Jan	Apr	Jul	Oct
T (°C)	OBS	6.0	11.5	32.3	19.3	4.2	17.3	30.4	18.5	4.3	16.1	30.4	18.4	5.7	15.4	31.9	19.9
	PRE	4.3	10.1	31.0	18.8	3.7	17.1	32.1	18.5	2.9	15.4	30.8	18.0	4.4	13.0	28.9	20.0
	NMB	− 0.28	− 0.12	− 0.04	− 0.03	− 0.11	− 0.01	0.06	0.00	− 0.33	− 0.04	0.01	− 0.02	− 0.22	− 0.16	− 0.09	0.01
	R	0.80	0.83	0.85	0.87	0.79	0.86	0.72	0.90	0.84	0.87	0.71	0.90	0.81	0.84	0.82	0.89
RH (%)	OBS	73.9	82.0	50.5	72.5	81.8	61.0	78.1	70.7	74.7	50.8	67.0	67.4	70.1	56.2	57.6	71.1
	PRE	65.0	71.9	66.1	69.2	61.9	39.0	61.9	50.2	65.0	44.0	68.9	58.5	73.0	64.8	80.0	70.0
	NMB	− 0.12	− 0.12	0.31	− 0.05	− 0.24	− 0.36	− 0.21	− 0.29	− 0.13	− 0.13	0.03	− 0.13	0.04	0.15	0.39	− 0.02
	R	0.64	0.72	0.72	0.71	0.67	0.66	0.55	0.72	0.75	0.70	0.51	0.75	0.66	0.68	0.73	0.79
CO (mg/m^3)	OBS	1.67	0.94	0.59	0.80	1.41	0.73	0.46	0.88	2.11	0.72	0.69	0.91	1.42	0.95	0.66	0.66
	PRE	1.58	0.84	0.47	0.95	1.51	0.82	0.38	1.16	1.76	0.82	0.55	1.04	1.50	0.99	0.54	0.64
	NMB	− 0.05	− 0.10	− 0.20	0.19	0.07	0.12	− 0.16	0.33	− 0.17	0.14	− 0.20	0.14	0.06	0.04	− 0.18	− 0.03
	N	424	648	672	672	424	648	672	672	424	648	672	672	424	648	672	672
	R	0.33	0.55	0.47	0.42	0.06	0.66	0.38	0.30	0.38	0.35	0.15	0.37	0.47	0.46	0.43	0.35
SO_2 ($\mu g/m^3$)	OBS	44.8	29.4	16.8	24.5	38.3	20.1	10.8	28.4	54.5	38.2	26.1	27.5	35.5	26.9	16.2	17.4
	PRE	49.1	26.2	13.2	24.2	48.3	22.0	11.6	25.0	49.2	27.3	22.1	27.9	37.2	31.3	17.6	20.0
	NMB	0.10	− 0.11	− 0.22	− 0.01	0.26	0.09	0.07	− 0.12	− 0.10	− 0.29	− 0.15	0.01	0.05	0.16	0.09	0.15
	N	424	648	672	672	424	648	672	672	424	648	672	672	424	648	672	672
	R	0.25	0.18	0.12	0.13	0.11	0.37	0.39	0.19	0.43	0.1	0.12	0.28	0.36	0.43	0.49	0.13
NO_2 ($\mu g/m^3$)	OBS	78.2	51.5	31.1	49.0	66.5	35.8	17.8	40.7	76.6	55.2	33.1	62.7	69.1	53.6	33.7	37.1
	PRE	71.8	51.9	29.0	52.5	67.1	39.2	14.2	48.2	75.2	51.1	35.7	58.8	72.2	57.5	29.9	33.2
	NMB	− 0.08	0.01	− 0.07	0.07	0.01	0.09	− 0.20	0.19	− 0.02	− 0.08	0.08	− 0.06	0.05	0.07	− 0.11	− 0.11
	N	424	648	672	672	424	648	672	672	4242	648	672	672	424	648	672	672
	R	0.49	0.45	0.46	0.41	0.24	0.49	0.44	0.56	0.5	0.46	0.35	0.39	0.67	0.52	0.42	0.46
O_3 ($\mu g/m^3$)	OBS	–	67.4	71.6	52.3	–	59.1	77.4	44.9	–	87.7	62.2	45.4	–	87.8	90.5	75.2
	PRE	–	79.7	96.2	54.7	–	86.0	104.7	61.3	–	74.0	88.9	51.1	–	74.3	115.5	84.5
	NMB	–	0.18	0.34	0.05	–	0.45	0.35	0.36	–	-0.16	0.43	0.12	–	− 0.15	0.28	0.12
	N	–	648	672	672	–	648	672	672	–	648	672	672	–	648	672	672
	R	–	0.46	0.71	0.54	–	0.39	0.53	0.51	–	0.49	0.56	0.46	–	0.49	0.8	0.38
$PM_{2.5}$ ($\mu g/m^3$)	OBS	127.1	63.5	31.2	55.8	137.9	57.7	36.2	108.1	135.7	61.9	37.1	67.1	111.3	67.7	41.7	32.6
	PRE	120.3	59.6	27.2	56.6	154.5	66.6	34.1	78.4	142.0	62.0	37.7	64.2	109.0	59.4	36.0	33.9
	NMB	− 0.05	− 0.06	− 0.13	0.02	0.12	0.15	− 0.06	− 0.27	0.05	0.00	0.02	− 0.04	− 0.02	− 0.12	− 0.14	0.04
	N	424	648	672	672	424	648	672	672	424	648	672	672	424	648	672	672
	R	0.15	0.27	0.48	0.42	0.03	0.57	0.15	0.18	0.39	0.51	0.14	0.35	0.32	0.49	0.64	0.39

Table 2 (continued)

		Hangzhou				Hefei				Nanjing				Shanghai			
		Jan	Apr	Jul	Oct	Jan	Apr	Jul	Oct	Jan	Apr	Jul	Oct	Jan	Apr	Jul	Oct
PM_{10} (μg/m^3)	OBS	160.8	107.5	60.3	98.2	175.9	112.4	66.8	145.2	203.2	143.2	62.5	136.7	142.2	100.6	68.6	59.7
	PRE	163.4	81.5	51.5	82.4	188.8	81.9	59.9	105.5	178.4	77.3	63.4	85.8	158.4	84.1	61.9	52.1
	NMB	0.02	− 0.24	− 0.15	− 0.16	0.07	− 0.27	− 0.10	− 0.27	− 0.12	− 0.46	0.02	− 0.37	0.11	− 0.16	− 0.10	− 0.13
	N	424	648	672	672	424	648	672	672	424	648	672	672	424	648	672	672
	R	0.12	0.17	0.52	0.35	0.03	0.36	0.19	0.15	0.41	0.23	0.14	0.30	0.27	0.40	0.54	0.18

Jan January, *Apr* April, *Jul* July, *Oct* October, in 2013, *OBS* mean observation, *PRE* mean prediction, *NMB* normalized mean bias, *N* data numbers, *R* correlation coefficient

Hefei, Nanjing and Shanghai, respectively. Table 3 shows the predicted $PM_{2.5}$ reduction percentages for different emission control scenarios for the four simulation months and entire year in the four cities.

Spatial distributions of the predicted reduction of $PM_{2.5}$ for the four months for the different emission control scenarios are shown in Fig. 1b. Absolute amounts and relative percentages of predicted reductions for $PM_{2.5}$ influenced by the four emission control policies, scenario Cases 1–4, vary for different months and locations as shown in Table 3. For example, three cities, Hangzhou, Hefei and Nanjing, are predicted to have the highest $PM_{2.5}$ reduction percentages in October, followed by April, July and January, while Shanghai has the highest reduction percentages in July, followed by April, January and October. Although predicted absolute $PM_{2.5}$ reductions in the four cities are relatively large in January, the reduction percentages are the smallest among these four months, because of the highest $PM_{2.5}$ base concentrations in the winter period.

On the basis of annual averages, Hangzhou is predicted to exhibit the highest percentage reductions for $PM_{2.5}$, ranging from − 9.7 to − 20.1%, followed by Nanjing, ranging from − 8.2 to − 18.7%, Shanghai, ranging from − 7.4 to − 15.8%, and Hefei, ranging from − 6.1 to − 13.8% (Table 3). In comparison with the general emission reduction strategies of scenario Cases 1 and 3, the corresponding scenario Cases 2 and 4 with enhanced emission reductions all predict to lead to further modest reductions of $PM_{2.5}$ in each city by 2.6 to 4.9% annually, depending on seasons and locations, as expected (Table 3). For example, scenario Cases 2 and 4 relative to the corresponding cases (Case1 and 3) lead to further reduction of $PM_{2.5}$ concentration in Hangzhou by 3.5 and 4.9%, Hefei by 2.6 and 3.9%, Nanjing by 2.8 and 4.9% and Shanghai by 2.6 and 3.8%.

Spatial distributions of the reduction of $PM_{2.5}$ in Fig. 1b reveal that reduction levels are similar for the same season, although cases with enhanced emission controls exhibit further reductions as expected, while they are very different for different seasons. For example, most of the $PM_{2.5}$ reductions in January for all four emission reduction strategies are predicted to occur in southeast Jiangsu, northeast Zhejiang and Shanghai, with slightly broader areas and higher reductions for the enhanced emission control scenarios (Fig. 1b), being consistent with the emission control strengths (Table 1). Predictions of spatial patterns of $PM_{2.5}$ reductions in April (Fig. 1b) are similar to those in January, but with expected smaller reduction amounts, owing to the lower base $PM_{2.5}$ concentrations in spring months. On the other hand, most of the predicted reductions of $PM_{2.5}$ in July are located in east Jiangsu, southeast Jiangsu and north Shanghai, with the lowest reduction amounts occurring in summer months because of the lowest base $PM_{2.5}$ concentrations. Moreover, spatial distributions of predicted reductions of $PM_{2.5}$ in October

Table 3 Predicted monthly and annual mean reduction (%) and amounts in parentheses (μg m^{-3}) of $PM_{2.5}$ concentrations relative to the baseline case in four cities: Hangzhou, Hefei, Nanjing and Shanghai

City	Month	Case 1		Case 2		Case 3		Case 4		Case 5		Case 6		Case 7		Case 8		Case 9		Case 10		Transport + natural	
Hangzhou	Jan	− 5.5	(− 6.61)	− 7.9	(− 9.49)	− 9.2	(− 11.19)	− 12.6	(− 15.22)	19.8	(23.81)	17.7	(21.41)	1.2	(1.41)	30.3	(36.63)	3.4	(4.15)	73.1	(88.38)	26.9	(32.35)
	Apr	− 10.7	(− 6.53)	− 15.0	(− 9.10)	− 17.4	(− 10.60)	− 22.9	(− 13.95)	22.0	(13.33)	31.6	(19.26)	5.8	(3.50)	11.5	(7.00)	11.1	(6.78)	72.4	(44.05)	27.6	(16.77)
	Jul	− 9.9	(− 2.40)	− 13.2	(− 3.20)	− 14.0	(− 3.39)	− 18.8	(− 4.56)	12.6	(3.05)	24.4	(5.91)	9.1	(2.21)	6.7	(1.63)	10.3	(2.50)	49.0	(11.88)	51.0	(12.36)
	Oct	− 11.1	(− 6.36)	− 15.0	(− 8.58)	− 18.0	(− 10.35)	− 23.4	(− 13.46)	17.3	(9.86)	34.0	(19.50)	5.7	(3.29)	10.8	(6.25)	10.8	(6.23)	73.3	(42.06)	26.7	(15.24)
	Ann	− 9.7	(− 5.35)	− 13.2	(− 7.39)	− 15.2	(− 8.64)	− 20.1	(− 11.44)	19.0	(12.51)	25.1	(16.52)	4.0	(2.60)	19.6	(12.88)	7.5	(4.92)	70.8	(46.59)	29.2	(19.18)
Hefei	Jan	− 2.6	(− 4.18)	− 3.9	(− 6.15)	− 4.2	(− 6.58)	− 6.7	(− 10.54)	14.9	(23.49)	9.2	(14.58)	0.0	(0.03)	32.8	(51.83)	2.9	(4.62)	61.7	(97.57)	38.3	(60.59)
	Apr	− 7.2	(− 5.04)	− 10.1	(− 7.09)	− 12.0	(− 8.39)	− 16.1	(− 11.28)	14.1	(9.83)	22.6	(15.82)	1.6	(1.13)	25.1	(17.52)	6.4	(4.50)	67.5	(47.12)	32.5	(22.68)
	Jul	− 6.2	(− 1.89)	− 8.9	(− 2.72)	− 9.1	(− 2.80)	− 12.7	(− 3.89)	8.0	(2.44)	18.2	(5.59)	3.6	(1.09)	13.7	(4.21)	5.5	(1.68)	41.9	(12.85)	58.1	(17.77)
	Oct	− 7.3	(− 5.72)	− 10.3	(− 8.12)	− 12.5	(− 9.79)	− 17.4	(− 13.66)	13.2	(10.34)	23.5	(18.44)	2.2	(1.76)	28.0	(21.97)	8.0	(6.26)	71.7	(56.28)	28.3	(22.15)
	Ann	− 6.1	(− 4.20)	− 8.7	(− 6.00)	− 9.9	(− 6.91)	− 13.8	(− 9.76)	13.7	(11.52)	16.2	(13.61)	1.2	(0.99)	28.3	(23.88)	5.1	(4.27)	63.4	(53.46)	36.6	(30.80)
Nanjing	Jan	− 4.2	(− 6.08)	− 6.2	(− 8.96)	− 8.3	(− 11.88)	− 11.1	(− 15.99)	17.4	(25.04)	13.4	(19.23)	1.2	(1.67)	33.7	(48.52)	6.1	(8.85)	70.5	(101.48)	29.5	(42.35)
	Apr	− 8.8	(− 5.65)	− 12.1	(− 7.73)	− 16.3	(− 10.46)	− 21.4	(− 13.74)	18.0	(11.54)	24.6	(15.78)	4.1	(2.64)	14.7	(9.41)	11.0	(7.04)	67.8	(43.39)	32.2	(20.61)
	Jul	− 9.2	(− 3.01)	− 11.8	(− 3.85)	− 11.4	(− 3.73)	− 16.7	(− 5.45)	16.4	(5.36)	19.5	(6.36)	8.4	(2.75)	6.2	(2.01)	6.8	(2.23)	46.4	(15.14)	53.6	(17.48)
	Oct	− 9.0	(− 5.72)	− 12.2	(− 7.76)	− 17.4	(− 11.13)	− 23.0	(− 14.70)	15.7	(10.02)	25.1	(16.03)	4.0	(2.53)	14.8	(9.41)	13.5	(8.63)	69.6	(44.44)	30.4	(19.34)
	Ann	− 8.2	(− 5.02)	− 11.0	(− 6.88)	− 13.8	(− 9.03)	− 18.7	(− 12.11)	17.1	(12.99)	18.9	(14.35)	3.2	(2.40)	22.8	(17.34)	8.8	(6.69)	67.2	(51.11)	32.8	(24.95)
Shanghai	Jan	− 4.8	(− 5.29)	− 7.0	(− 7.70)	− 8.7	(− 9.69)	− 11.5	(− 12.81)	11.6	(12.43)	17.4	(19.20)	2.2	(2.19)	36.4	(40.78)	5.1	(5.62)	75.1	(83.48)	24.9	(26.96)
	Apr	− 8.2	(− 4.77)	− 11.4	(− 6.63)	− 14.1	(− 8.27)	− 18.3	(− 10.69)	17.1	(9.83)	27.6	(16.06)	5.4	(3.12)	14.6	(8.51)	10.5	(6.13)	70.0	(40.77)	30.0	(17.30)
	Jul	− 11.0	(− 3.35)	− 15.1	(− 4.61)	− 15.4	(− 4.70)	− 21.3	(− 6.47)	14.7	(4.43)	27.8	(8.48)	11.3	(3.44)	8.9	(2.71)	10.1	(3.06)	60.4	(18.39)	39.6	(12.02)
	Oct	− 4.8	(− 1.70)	− 5.5	(− 2.02)	− 8.5	(− 3.06)	− 10.8	(− 3.90)	6.1	(2.20)	22.2	(7.61)	4.5	(1.50)	10.0	(3.59)	6.2	(2.27)	49.1	(17.14)	50.9	(16.59)
	Ann	− 7.4	(− 3.62)	− 10.0	(− 4.98)	− 12.0	(− 6.09)	− 15.8	(− 8.02)	12.4	(7.22)	22.1	(12.84)	4.4	(2.56)	23.9	(13.90)	7.3	(4.27)	68.7	(39.94)	31.3	(18.22)

Scenario Cases 1 and 2 are, respectively, for general and enhanced emission reductions for both industrial and power plant emissions. Scenario Cases 3 and 4 are, respectively, for general and enhanced emission reductions for industrial, power plant and transportation emissions, respectively. Contributions of different anthropogenic sectors: Scenario Case 5 for agriculture, 6 for industry, 7 for power plants, 8 for residential, 9 for transportation and 10 for all five anthropogenic sectors. Note that the contributions of regional transport and natural sources (transport and natural) are estimated on the basis of Case 10

Jan January, *Apr* April, *Jul* July, *Oct* October, *Ann* Annual are more scattered spatially, which are mainly located in central Anhui, east Anhui, south Jiangsu, north Zhejiang and Shanghai, with comparable amounts to those in April.

Overall, the predicted reduction percentages in these four cities for all four emission control policies do not meet the 20% $PM_{2.5}$ reduction goals over the Yangtze River Delta region in the 13th Five-Year Plan in winter (Table 3). Only Hangzhou is predicted to meet these goals on the basis of the annual simulation results (Table 3). One of the reasons is that residential emissions make the highest contributions to the $PM_{2.5}$ concentrations in all four cities in winter when heavy haze pollution is at its highest.

Predicted contributions of agriculture, industry, power plants, residential, transportation, as well as all five anthropogenic emission sectors to mean $PM_{2.5}$ levels in four cities are summarized in Table 3. Note that the contributions of regional transport and natural sources are estimated on the basis of Case 10 (100%–Case 10). On the basis of annual averages, the predicted mean contributions of agriculture, industry, power plants, residential, transportation and all five anthropogenic emission sectors to average $PM_{2.5}$ concentrations are 19.0, 25.1, 4.0, 19.6, 7.5 and 70.8% in Hangzhou, 13.7, 16.2, 1.2, 28.3, 5.1 and 63.4% in Hefei, 17.1, 18.9, 3.2, 22.8, 8.8 and 67.2% in Nanjing, 12.4, 22.1, 4.4, 23.9, 7.3 and 68.7% in Shanghai.

The results in Table 3 also show that the other sources, including regional transport from outside of the Yangtze River Delta region and natural sources, are predicted to contribute 29.2, 36.6, 32.8 and 31.3% to annual mean $PM_{2.5}$ concentrations in Hangzhou, Hefei, Nanjing and Shanghai, respectively. On a seasonal basis, residential emissions are predicted to have the highest contributions to the $PM_{2.5}$ concentrations in January in all four cities, ranging from 30.3% in Hangzhou to 36.4% in Shanghai, followed by agriculture, industry, transportation and power plants. Industrial emissions have the highest contributions to $PM_{2.5}$ concentrations in April, ranging from 24.6% in Nanjing to 31.6% in Hangzhou and July (18.2% in Hefei) in all four cities. Agricultural emissions are also influential in the $PM_{2.5}$ level, with annual contributions ranging from 12.4% in Shanghai to 19.0% in Hangzhou. Overall, residential emissions are predicted to make the highest contributions to the $PM_{2.5}$ concentrations in the Yangtze River Delta in January, the most polluted period, and make the highest contributions in three cities, except Hangzhou on the basis of annual simulations.

Conclusion

The two-way coupled WRF-CMAQ model has been used to assess the potential benefits of different emission control strategies for $PM_{2.5}$ concentrations, in the Yangtze River Delta. Predicted annual mean reduction percentages for $PM_{2.5}$ are the highest in Hangzhou, ranging from − 9.7 to − 20.1%, followed by Nanjing, ranging from − 8.2 to − 18.7%, Shanghai, ranging from − 7.4 to − 15.8%, and Hefei, ranging from − 6.1 to − 13.8% for strategies 1: normal emission controls for industry and power plants, 2: enhanced emission controls for industry and power plants, 3: normal emission controls for industry, power plants and transportation, and 4: enhanced emission controls for industry, power plants and transportation.

As a consequence, among the four major cities in the Yangtze River Delta, only Hangzhou can meet the 20% $PM_{2.5}$ reduction goals. Sensitivity tests reveal that the predicted annual mean contributions of the agriculture, industry, power plants, residential, transportation and all five anthropogenic emission sectors to average $PM_{2.5}$ were estimated at 19.0, 25.1, 4.0, 19.6, 7.5 and 70.8% in Hangzhou, 13.7, 16.2, 1.2, 28.3, 5.1 and 63.4% in Hefei, 17.1, 18.9, 3.2, 22.8, 8.8 and 67.2% in Nanjing, 12.4, 22.1, 4.4, 23.9, 7.3 and 68.7% in Shanghai.

The residential emissions have the highest contributions to the $PM_{2.5}$ concentrations in January in all four cities, followed by agriculture, industry, transportation and power generation, while industrial emissions have the highest contributions to $PM_{2.5}$ concentrations in April and July in all four cities. Predicted annual contributions of regional transport from beyond the Yangtze River Delta region as well as natural sources to mean $PM_{2.5}$ concentrations in Hangzhou, Hefei, Nanjing, and Shanghai are 29.2, 36.6, 32.8 and 31.3%, respectively.

A major finding is that residential sources are of comparable importance to industrial, power plant and transportation sources to $PM_{2.5}$ concentrations over the Yangtze River Delta of China. While current policies have only considered the emission reductions of industry, power plants and transportation, emission control strategies of residential sources are not specifically proposed in the 13th Five-Year Plans. Furthermore, agricultural sources and regional transport of pollutants have important impact on the concentration of $PM_{2.5}$, 12.4–19.0% and 29.2–36.6%, respectively, which are needed to be considered in the policies, too. Overall, more comprehensive emission control policies are needed to be formulated by the local governments in the Yangtze River Delta of China to accomplish the reduction goals of $PM_{2.5}$ in the 13th Five-Year Plan. This information is important not only for the Yangtze River Delta but also for all other regions of China, as well as other developing countries, when they formulate and implement effective emission control policies.

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