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Probe into gaseous pollution and assessment of air quality benefit under sector dependent emission control strategies over megacities in Yangtze River Delta, China

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HIGHLIGHTS

- China's new air quality standards is employed for the very first time to examine air quality status over YRD.
- Primary gaseous pollutants are identified for different cities.
- Pollutants responses and air quality benefits to precursors emissions change are assessed under applicable scenarios.

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ABSTRACT

On February 29th 2012, China published its new National Ambient Air Quality Standard (CH-NAAQS) aiming at revising the standards and measurements for both gaseous pollutants including ozone (O_3), nitrogen dioxide (NO_2), and sulfur dioxide (SO_2), and also particle pollutants including PM_{10} and $PM_{2.5}$. In order to understand the air pollution status regarding this new standard, the integrated MM5/CMAQ modeling system was applied over Yangtze River Delta (YRD) within this study to examine the criteria gaseous pollutants listed in the new CH-NAAQS. Sensitivity simulations were also conducted to assess the responses of gaseous pollutants under 8 different sector-dependent emission reduction scenarios in order to evaluate the potential control strategies. 2006 was selected as the simulation year in order to review the air quality condition at the beginning of China's 11th Five-Year-Plan (FYP, from 2006 to 2010), and also compared with air quality status in 2010 as the end of 11th FYP to probe into the effectiveness of the national emission control efforts. Base case simulation showed distinct seasonal variation for gaseous pollutants: SO_2, and NO_2 were found to have higher surface concentrations in winter while O_3 was found to have higher concentrations in spring and summer than other seasons. According to the analyses focused on 3 megacities within YRD, Shanghai, Nanjing, and Hangzhou, we found different air quality conditions among the cities: NO_2 was the primary pollutant that having the largest number of days exceeding the CH-NAAQS daily standard (80 $\mu g\ m^{-3}$) in Shanghai (59 days) and Nanjing (27 days); SO_2 was the primary pollutant with maximum number of days exceeding daily air quality standard (150 $\mu g\ m^{-3}$) in Hangzhou (28 days), while O_3 exceeding the daily maximum 8-h standard (160 $\mu g\ m^{-3}$) for relatively fewer days in all the three cities (9 days in Shanghai, 14 days in Nanjing, and 11 days in Hangzhou). Simulation results from predefined potential applicable emission control scenarios suggested significant air quality improvements from emission reduction: 90% of SO_2 emission removed from power plant in YRD would be able to reduce more than 85% of SO_2 pollution, 85% NO_x emission reduction from power plant would reduce more than 60% of NO_2 pollution, in terms of reducing the number of days exceeding daily air quality standard. NO_x emission reduction from transportation and industry were also found to effectively reduce NO_2 pollution but less efficient than emission control from power plants. We

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also found that multi-pollutants emission control including both NO_x and VOC would be a better strategy than independent NO_x control over YRD which is China's 12th Five-Year-Plan (from 2011 to 2015), because O_3 pollution would be increased as a side effect of NO_x control and counteract NO_2 pollution reduction benefit.

1. Introduction

Yangtze River Delta (YRD) refers to the coastal area where Yangtze River drains into the East China Sea, including Shanghai, southern Jiangsu province and northern Zhejiang province of China. YRD is also called the Golden Triangle of the Yangtze. The rapid developing industries, urban buildup, and economics in this area have given rise to one of the largest megacity clusters in the world. Cities like Shanghai, Nanjing, and Hangzhou, each has a population of more than 8 millions. As home to over 80 million people with which 50 million are urban, YRD suffers from severe air pollution problems from the rapid progress of urbanization, industrialization, and expansion of automobile usage during the last decades, drawing attentions from both Chinese and national researchers. Anthropogenic emission from YRD has been documented in a few studies recently. For example, NASA's INTEX-B mission estimated anthropogenic emissions of SO_2, NO_x, CO, VOCs, PM_{10}, $PM_{2.5}$, BC, OC from Shanghai in 2006 were 618, 631, 1958, 594, 138, 91, 10, 8 Gg, respectively. Satellite observations of GOME and SCIAMACHY showed that the mean column concentrations of NO_2 increased about 50% during 1996–2004, and appeared to continually rise in the following years over YRD (Richter et al., 2005).

The intensive emissions within YRD cause serious air pollution problems. The severest haze episode in Yangtze River Delta occurred on Jan. 19, 2007. Hourly peak of 466 $\mu g\ m^{-3}$ for $PM_{2.5}$ which was resulted from stagnant dispersion condition due to local meteorology broke the record since the monitoring network of observation in this region was established (Fu et al., 2008). In addition, local pollutants could be transported to downstream areas such as Taiwan in the winter, and even could reach Hong Kong (Chuang et al., 2008). A few studies have been recently conducted to examine air pollution over YRD with both measurement and modeling methods. Geng et al. (2011) reported biogenic emission near Shanghai had important impacts on the ozone formation in the city based on observations; Tie et al. (2009) conducted modeling simulation for August 2007 in Shanghai and found that sub-tropical high pressure system in the city could buildup ozone concentration as high as 100–130 ppbv; Li et al. (2011b) applied $O_3/(NO_y{-}NO_x)$ and H_2O_2/HNO_3 indicators to analyze the ozone sensitivity in urban and rural areas of Shanghai in July 2007.

Although the above studies provided insightful information about particle and ozone pollution during certain time periods, very limited information is available regarding the overall conditions and seasonality of air quality over YRD. Also, these studies usually investigate one single air pollutant within one case rather than examining multi-pollutants simultaneously, and few studies focused on the pollutants responses to precursor emission changes from anthropogenic sources. Because air quality could be affected greatly by rapid change of meteorological conditions which usually happen over YRD especially in the monsoon season (Wang et al., 2006), steady state is possibly never reached due to the ultimate advection of air and sea breeze in and out of the YRD region. So instead of using chemical indicators such as $O_3/(NO_y{-}NO_x)$, we directly look into the sensitivity simulation results to quantify the ozone responses although more computation resource is consumed. Moreover, while China has recently released its new National Ambient Air Quality Standards in February 29, 2012 (http://kjs.mep.gov.cn/hjbhbz/bzwb/dqhjbh/dqhjzlbz/201203/t20120302_224165.htm), it is still unclear what is the attainment or non-attainment status over YRD if we examine it according to this new standard. Uncertainties also remain in the question that in case YRD is non-attainment area, what emission control actions should be employed in order to lower the air pollutants concentrations and meet the standard. In this study, we conducted model simulations over YRD for the whole year in 2006 to improve the understanding of the gaseous pollutants and the air quality conditions in terms of exceeding the new standards. We also conducted sensitivity analysis with emission reduction of NO_x, SO_2, and VOC from different emission sectors including traffic, industry, and power plant. Air quality benefits were assessed for potential emission control scenarios in order to provide scientific basis and suggestions for the effective emission control strategies design in YRD. Model description and simulation scenarios are described in Section 2. Section 3 examines the characteristics of air pollution in terms of gaseous pollutants, and also discusses the response of air pollutants (i.e. NO_2, SO_2, and O_3) under predefined emission reduction scenarios. Section 4 draws conclusions and provides implications for practical emission control strategies in YRD comparing the effects between before (2006) and after (2010) 11th Five-Year-Plan whether it could be transformed to 12th Five-Year-Plan.

2. Methodology

2.1. Model description

Community Multiscale Air Quality (CMAQ) model version 4.6 was applied over YRD for full year simulation in 2006. Although there has been updated versions of CMAQ released recently, the CB05 gas-phase chemistry scheme still remain the same among different versions. MM5 version 3.7 was applied in this study to provide meteorological field, with Four Dimensional Data Assimilation (FDDA) performed using National Centers for Environmental Prediction (NCEP) dataset ds351.0 and ds461.0. Detailed model configurations for MM5/CMAQ are shown in Table 1. CMAQ simulation was performed with 27 km for domain D1 and nested-down to 9 km domain D2, and finally to 3 km domain D3 as shown in Fig. 1. Initial and boundary conditions for D1 were downscaled from global model GEOS-Chem following the method described in Lam and Fu (2009). Our analysis will mainly focus on three most populated cities within YRD: Shanghai, Nanjing, and Hangzhou which have a population of 24, 8.1, and 8.7 million, respectively. Nanjing is the capital of Jiangsu province, and Hangzhou is the capital of Zhejiang province.

2.2. Emissions and simulation scenarios

In this study, the Intercontinental Chemical Transport Experiment-Phase B (INTEX-B) 2006 emission inventory (Zhang et al., 2009) was employed to provide the anthropogenic emissions for species SO_2, NO_x, CO, NMVOC, PM_{10}, $PM_{2.5}$, BC, OC from 4 different sectors: power plants, industry, residential, and transportation. The Model of Emissions of Gases and Aerosols from Nature (MEGAN) was used to provide biogenic emission inventory (http://bai.acd.ucar.edu/Megan/index.shtml). The 2D grid-cell-

Table 1
Model configurations for the MM5/CMAQ simulations.

MM5 V3.7	
Horizontal resolution	27 km/9 km/3 km (one-way nested)
Number of vertical layers	34
Explicit precipitation scheme	Simple ice (Dudhia) scheme
Longwave radiation	RRTM
Shortwave radiation	Dudhia scheme
Land-surface	Noah
Advection	Global mass-conserving scheme
Planetary boundary layer(PBL) scheme	MRF
Cumulus option	27 km and 9 km domains: Grell 3 km domain: none
Horizontal resolution	27 km/9 km/3 km (one-way nested)
CMAQ 4.6	
Horizontal resolution	27 km/9 km/3 km
Vertical resolution	19 sigma-pressure levels (with the top pressure of 100 mb)
Projection	Lambert conformal conic
Advection	Piecewise parabolic scheme
Vertical diffusion	K-theory
Gas-phase chemistry	CB05 with Euler Backward Iterative (EBI) solver
Dry deposition	Wesely
Wet deposition	Henry's law
Aqueous chemistry	Walcek
Aerosol mechanism	AE4

based emissions for the 9 km and 3 km domains were obtained using top-down method (Du, 2008).

In order to better understand the contributions from each sector in our research domain, sectoral anthropogenic emission inventory (NO_x, VOC, SO_2, and CO) in YRD was shown and compared with China's national emission inventory as shown in Fig. 2. YRD was found to have a similar anthropogenic emission contribution pattern for all the four species listed above, indicating the consistency between emission inventories between nested domains (D1, D2, D3). This consistency also implies that the results concluded in this study based on this typical emission pattern of YRD might be also helpful as a reference for national level study. As shown in the figure, power plant sector is the largest contributor for both NO_x and SO_2, accounting 42.5% and 61.9% for the total anthropogenic emission, respectively. Transportation and industry contributed similar amount of NO_x and VOC emission, which are the two major ozone precursors.

Currently, well-developed technology regarding NO_x emission control, like Selective Catalytic Reduction (SCR) (Shelef, 1995) and Selective Non-Catalytic Reduction (SNCR) (Javed et al., 2007), are mainly applied to large utility, industrial, and municipal solid waste boilers with NO_x reduction efficiency of 70–95%. Although there are more recent applications of the NO_x control technologies for diesel engines, a stable reduction rate may still only possible for power plant sector, since transportation and industry is difficult to control uniformly for all individual vehicles and different type of industries. However, the application of SCR and SNCR technologies haven't been enacted widely in China. Within China's new "Twelfth Five - Year Plan" (12th FYP) guideline, NO_x emission reduction was set up at the very first time, targeting at 10% reduction by 2016 as compared to 2010 (http://english.mep.gov.cn/News_service/infocus/201202/t20120207_223194.htm).

For SO_2 emission control, flue-gas desulfurization (FGD) technology has been widely applied in US and Europe in typical coal-fired power stations and can remove 85–95% of the SO_2 emission (Hudson and Rochelle, 1982). However, there were very limited emission control efforts for SO_2 and NO_x in China before 2005 (http://www.mep.gov.cn/gkml/hbb/bgg/201104/t20110420_209449.htm). For instance, Lu et al. (2010) reported anthropogenic SO_2 emission increased by 53% from 2000 (21.7 Tg) to 2006 (33.2 Tg). Similarly, Zhang et al. (2009) reported anthropogenic SO_2 emission increased by 36% from 22.9 Tg to 31.0 during the same period. But after suffering acid rain and soil acidification for more than 28% of the country's territory (Zhao et al., 2009) in 2005, China has targeted reducing SO_2 emission by widely applying FGD to coal-fire power plants from 2006 to 2010 within the period of the "Eleventh Five Year Plan" (11th FYP). So in order to make reasonable emission control strategies and design applicable policies, we set up 8 emission control scenarios in this study as listed in Table 2. An emission reduction ratio of 20% for NO_x is designed for transportation (T-N20) and industry (I-N20), and the same for VOC (T-V20, I-V20). However, NO_x emission reduction aiming at reduce NO_2 pollution may cause the increase of O_3 through titration. To avoid this potential negative effect, another two emission control scenarios that consider simultaneous reduction of NO_x and VOC emission were designed (I-C20 and T-C20). As for the emission from power plants, an emission reduction ratio of 85% for NO_x (P-N85) and 90% for SO_2 (P-S90) was designed based on the efficiency of current control technologies.

The difference between base case and these control cases will be used to quantify the contributions of the potential feasible emission reduction strategies, and also provide an insightful path to understand the sensitivity response of gaseous pollutants for precursors emissions over YRD. We will also use air quality reports published by local Environmental Protection Bureau (EPB) for year 2010 to compare with these control scenarios to provide a brief assessment of the benefits from real control efforts during the 11th FYP.

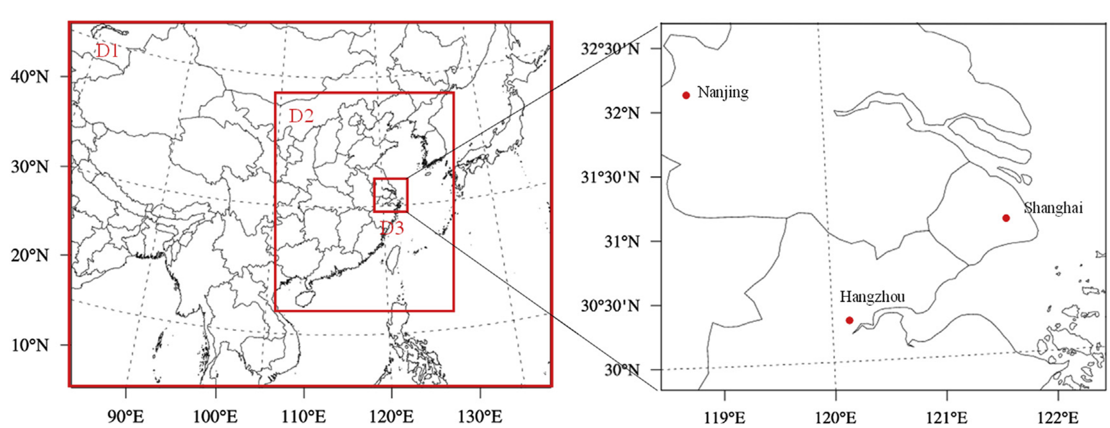

Fig. 1. Three nested modeling domains at 27-km (East Asia: D1), 9-km (East China: D2), and 3-km (Yangtze River Delta D3).

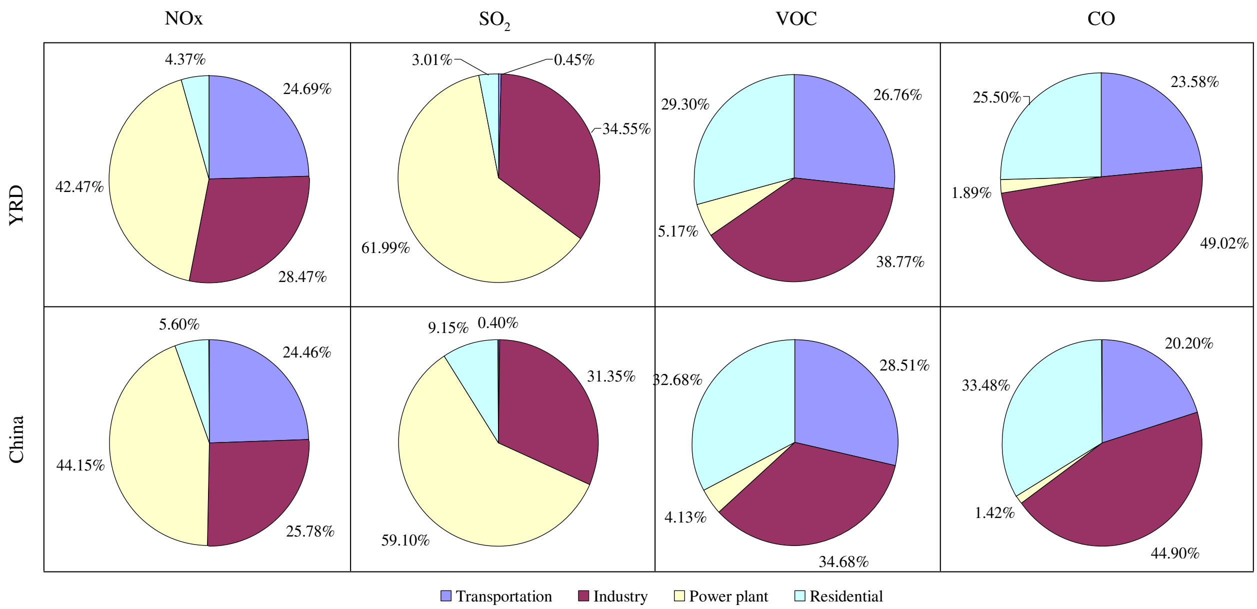

Fig. 2. Anthropogenic emission contributions of source categories in YRD (upper) and whole China (lower) for NO_x, SO_2, VOC, and CO.

2.3. China's National Ambient Air Quality Standard

CH-NAAQS was employed in this study to examine the air pollutions over YRD and assess the benefit from potential emission control strategies. While the new CH-NAAQS is developed based on China's local air quality and have been barely mentioned in any few publications due to its recently release, it is necessary to briefly introduce it. US National Ambient Air Quality Standards (US-NAAQS) identifies two types of standards as the primary standards for public health protection, including the health of sensitive populations such as asthmatics, children, and the elderly, and the secondary standards for public welfare protection including protection against decreased visibility and damage to animals, crops, vegetation, and buildings. Similarly, CH-NAAQS identifies its two levels of standards based on land use types: the level 1 standard is applied to areas including national parks, scenic areas, and special protected zones; level 2 standard is less stringent, applied for all the rest areas including residential areas, mixed zones for commercial traffic and residents, cultural district, industrial zone, and also rural areas. Since YRD is mainly a mixed area of industrial, residential, commercial and agricultural zones, level 2 standard is considered as the benchmark standard in this study. Table 3 summarized level 2 standards from CH-NAAQS and the primary standards from US-NAAQS for criteria pollutants. Apparently, CH-NAAQS determined the similar thresholds for CO, NO_2, O_3 and PM_{10} as compared to US-NAAQS, but a less stringently thresholds for SO_2 and $PM_{2.5}$, which was likely due to the limited control efforts for these pollutants in China.

Table 2
Emission control scenarios.

Case no.	Emission reduction Category	Emission reduction ratio (%)		
		NO_x	VOC	SO_2
I-N20	Industry	20	–	–
I-V20	Industry	–	20	–
I-C20	Industry	20	20	–
T-N20	Transportation	20	–	–
T-V20	Transportation	–	20	–
T-C20	Transportation	20	20	–
P-N85	Power plant	85	–	–
P-S90	Power plant	–	–	90

2.4. Model evaluation

MM5 model performance was evaluated against surface dataset from the National Climatic Data Center (NCDC) following the evaluation protocol used in Fu et al. (2012). The four major meteorological variables evaluated included 2-m wind speed and wind direction, 2-m temperature, and 2-m relative humidity, which were the most important factors affecting air pollution. The domain-wide performance for all 12 months is summarized in Table 4. Temperature is reproduced well throughout the year. Its Mean Bias (MB), Gross Error (GE) and Index of Agreement (IOA) all meet the benchmarks for every month throughout the entire year; Underestimation of temperature occurred for most of the months except Feb., Apr., May., and Jun. For the simulated wind speed, overestimation dominated in most of the months except for Feb. However, the maximum positive bias which occurred in January was only 0.47 m s^{-1}, still less than the 0.5 m s^{-1} benchmark. Underestimation of relative humidity occurs from May to Oct., and other months are dominated by slightly overestimation. The statistics for our study almost meet the suggested benchmarks for every variable in every month, except for a slightly higher bias and gross error for wind direction in Jan., Oct., and Dec., indicating the meteorological field provided by MM5 in this study successfully replicated the meteorological conditions over YRD.

Model evaluation for CMAQ was performed following the evaluation protocol proposed by U.S. EPA (EPA, 2007) by using key statistical performance indicators including Normalized Mean Bias (NMB), Normalized Mean Error (NME), Mean Fractional Bias (MFB), and also Mean Fractional Bias (MFE) as shown in Fig. 3. Statistics of model performance are listed on the top of each plot. MFB and MFE benchmarks for ozone and $PM_{2.5}$ suggested by the U.S. EPA guidance were provided in red numbers in Fig. 3(a) and (d). Daily observations for SO_2, NO_2, and PM_{10} were derived from the Air Pollution Index (API) (http://datacenter.mep.gov.cn/report/air_daily/air_dairy_en.jsp), which is published by China Ministry of

Table 3
Comparison between CN-NNAQS and US-NAAQS.

Pollutant		Averaging time	US-NAAQS (primary level)	CN-NAAQS (level 2)	CN-NAAQS (level 2) under standard atmosphere[a]
Carbon Monoxide		24-h	–	4 mg m^{-3}	3.2 ppm
		8-h	9 ppm	–	
		1-h	35 ppm	10 mg m^{-3}	8 ppm
Lead		Rolling 3 month average	0.15 μg m^{-3}	–	–
Nitrogen Dioxide		1-h	100 ppb	200 μg m^{-3}	97.39 ppb
		24-h	–	80 μg m^{-3}	38.96 ppb
		Annual	53 ppb	40 μg m^{-3}	19.47 ppb
Particle pollution	$PM_{2.5}$	Annual	15 μg m^{-3}	35 μg m^{-3}	–
		24-h	35 μg m^{-3}	75 μg m^{-3}	–
	PM_{10}	Annual	–	70 μg m^{-3}	–
		24-h	150 μg m^{-3}	150 μg m^{-3}	–
Sulfur dioxide		1-h	75 ppb	500 μg m^{-3}	175.01 ppb
		24-h	–	150 μg m^{-3}	52.5 ppb
		Annual	–	60 μg m^{-3}	21.0 ppb
Ozone		8-h	75 ppb	160 μg m^{3}	74.67 ppb
		1-h	–	200 μg m^{-3}	93.34 ppb

[a] CN-NAAQS under standard atmosphere: Unit for gaseous pollutants employed in CN-NAAQS is μg m^{-3}, so the values for gaseous pollutants are converted to ppb unit under standard atmosphere (Seinfeld and Pandis, 1998) to make it comparable with US-NAAQS.

Environmental Protection (MEP) and is the only publicly available air quality data there. O_3 (June 11–17, July 11–17 and August 13–19 2006) and $PM_{2.5}$ (April, August, and November) observations are provided by Fudan University (Wang et al., 2006) in Shanghai. Simulated gas-phase species generally had better evaluation performance than particles. MFB and MFE for O_3 was −17.05% and 37.25%, respectively, which was slightly higher than the suggested benchmarks values as ±15% and 35%. MFB and MFE for SO_2 was 10.3% and 41.0% respectively, and the values for NO_2 was 18.1% and 34.4% respectively, indicating an overall mild overprediction for SO_2 and slight overprediction for NO_2. CMAQ underpredicted both $PM_{2.5}$ and PM_{10} as shown in Fig. 3(d) and (e), which may be mainly due to the underestimation of emissions. It was indicated in Zhang et al. (2009) that there were a lot of uncertainties in the emissions over China in 2006 for individual sites, which could affect the CMAQ simulation accuracy.

Monthly variations of CO, SO_2, NO_2 and O_3 were also investigated to examine the model performance and air quality status throughout the year. As shown in Fig. 4, the three megacities have similar monthly variation patterns for CO. It began with a high concentration in January, and started to decrease gradually until July when it reached the bottom values, and then bounced back gradually to high levels until December. Monthly variations of SO_2 and NO_2 were similar to CO, suggesting the overall air pollution was worse in winter than that in summer in terms of CO, SO_2, and NO_2 pollution. The low wind speed and Planet Boundary Layer (PBL) height resulted from the Siberian high lead to less convective dispersion and scavenging over YRD. Our findings were generally consistent with results reported from other studies and local observations: Li et al. (2011a) reported domain average NO_2 and SO_2 concentration over YRD was 43 μg m^{-3}, 45 μg m^{-3}, and 33 μg m^{-3}, 36μg m^{-3} in January and July 2004, respectively; Fu et al. (2008) reported the five-year (from 2002 to 2006) annual average NO_2 and SO_2 concentration was 59 μg/m^3 and 49 μg/m^3 respectively; Shanghai Municipal Environmental Protection Bureau reported the observed annual NO_2 and SO_2 concentration in 2006 was 56 μg/m^3 and 51 μg/m^3 respectively (http://www.envir.gov.cn/law/bulletin/b2007/en/en_0007.html). These values agree well with our CMAQ simulated results for SO_2 (52 μg m^{-3}) and NO_2 (61 μg m^{-3}). Nanjing Municipal Environmental Protection Bureau reported the downtown area observed annual NO_2 and SO_2 concentration in 2006 was 52 μg m^{-3} and 63 μg m^{-3} respectively, while our CMAQ prediction for the Nanjing was 47 μg m^{-3} and 56 μg m^{-3}, respectively. The INTEX-B emission inventory was found to have weak seasonal variations for NO_x and SO_2 emissions because these two species mainly come from industries, power stations, and transportation sector, which had insignificant seasonal cycles (Zhang et al., 2009). Thus the seasonal variations of these pollutants over YRD were mainly affected by regional meteorology and chemical process. YRD is located at the east coast of China with marine monsoon subtropical climate, and local weather is under the influence of Asian monsoon system. Summers are usually hot and humid, winters are cool and dry, and erratic spring and fall may switch between warm and cool. An increasing temperature from early spring until midsummer results in the increasing strength of both vertical mixing and horizontal advection processes, which lead to a favorable condition for air pollutants dilution, meanwhile the increasing solar radiation also boost the photochemical production of ozone. Along with peak precipitation in June and July, East Asia monsoon also brings clean air from East China Sea (part of west Pacific Ocean) and dilute the air pollutants in YRD starting from May and ends in October through both horizontal/vertical advections and wet deposition. This seasonality feature indicates that local emission reduction for these pollutants may also take into consideration of the meteorology impacts to select the major controlling episodes in order to have the most effective results.

3. Results and discussion

3.1. Exceedance of CH-NAAQS

While China has just published the new CH-NAAQS recently, there has been very limited publication reviewing air pollution status in YRD regarding how pollutants concentrations exceed the new daily standard. In this section, the total numbers of days exceeding the new CH-NAAQS in 2006 for these criteria gaseous pollutants were evaluated for Shanghai, Nanjing, and Hangzhou, as shown in Fig. 5. Modeling results were extracted over urban areas of each city in order to focus on the most populated region within YRD.

There was no exceedance for CO at any of the cities, so numbers of days that pollutants concentrations exceed air quality standards were only summarized for daily maximum 8-h ozone ($MDA8O_3$), NO_2, and SO_2. According to our simulation results, in Shanghai, NO_2 had 59 days (16% of the year) exceeding the 80 μg m^{-3} daily average standard, followed by SO_2 which had 30 days (8% of the year) exceeding the 150 μg m^{-3} standard, and O_3 had 9 days (3% of the

Table 4
Model Evaluation Statistics of MM5 prediction at 3 km over YRD in 2006.[a]

Mon	Temperature (K)						Wind speed ($m\ s^{-1}$)						Wind dir. (deg)				Humidity ($g\ kg^{-1}$)					
	Mean_Obs	Mean_Sim	MB	GE	RMSE	IOA	Mean_Obs	Mean_Sim	MB	GE	RMSE	IOA	Mean_Obs	Mean_Sim	MB	GE	Mean_Obs	Mean_Sim	MB	GE	RMSE	IOA
Benchmarks	–	–	**<±0.5**	**<2**		**≥0.8**	**-**	**-**	**<±0.5**		**<2**	**≥0.6**	–	–	**<±10**	**<30**	–	–	**<±1**	**<2**		**≥0.6**
Jan	278.53	278.33	−0.20	1.25	1.56	0.87	3.36	3.82	0.47	1.27	1.58	0.74	159.48	140.83	11.40	33.04	4.41	4.47	0.06	0.42	0.51	0.73
Feb	278.50	278.54	0.03	1.04	1.28	0.89	3.77	3.74	−0.02	1.08	1.36	0.82	157.59	140.59	1.65	26.86	4.21	4.32	0.12	0.42	0.53	0.76
MAR	284.56	284.51	−0.05	1.10	1.39	0.93	3.38	3.55	0.18	1.12	1.44	0.81	183.27	158.63	2.76	32.24	5.52	5.83	0.31	0.72	0.89	0.71
Apr	289.92	289.86	0.03	1.15	1.44	0.93	3.61	3.84	0.19	1.23	1.55	0.78	190.81	193.28	2.47	30.77	8.11	8.55	0.46	1.17	1.44	0.66
May	293.82	293.85	0.04	1.07	1.35	0.93	3.52	3.79	0.27	1.11	1.40	0.78	170.80	165.33	−1.35	28.11	10.78	10.63	−0.16	1.22	1.54	0.63
Jun	298.69	298.77	0.11	1.16	1.49	0.91	3.11	3.53	0.41	1.20	1.50	0.73	166.53	157.13	−1.00	36.16	15.90	14.89	−1.01	1.61	2.05	0.48
Jul	302.09	302.00	−0.09	1.22	1.53	0.87	3.76	4.03	0.28	1.27	1.60	0.75	162.85	152.55	−0.24	33.72	19.75	18.74	−1.01	1.92	2.37	0.47
AUG	302.78	302.35	−0.43	1.21	1.51	0.91	3.25	3.58	0.33	1.14	1.44	0.69	153.31	141.27	−1.37	35.65	19.19	17.85	−1.34	2.26	2.81	0.45
Sep	296.20	295.94	−0.26	1.08	1.33	0.92	3.24	3.42	0.18	1.11	1.41	0.77	110.05	92.45	6.46	34.88	13.45	13.04	−0.42	1.31	1.66	0.54
Oct	294.37	294.14	−0.22	1.12	1.37	0.93	2.66	2.96	0.30	0.98	1.24	0.76	130.55	127.64	10.21	37.90	11.74	11.09	−0.65	1.35	1.69	0.58
Nov	287.72	287.58	−0.11	1.04	1.31	0.94	3.04	3.31	0.25	1.17	1.50	0.77	210.38	190.94	5.76	35.85	7.31	7.42	0.09	0.80	1.00	0.78
Dec	280.31	279.93	−0.39	1.33	1.68	0.91	2.75	2.85	0.10	0.99	1.30	0.78	240.13	215.48	10.44	39.93	4.52	4.53	0.01	0.54	0.67	0.80

$MB = 1/IJ\sum_{j=1}^{J}\sum_{i=1}^{I}(Sim_j^i - Obs_j^i)$, Sim_j^i is the individual simulated value at site j and time i, and Obs_j^i is the individual observed value at site j and time i, and the summations are overall sites (I) and over time periods (J).

$GE = 1/IJ\sum_{j=1}^{J}\sum_{i=1}^{I}\left|Sim_j^i - Obs_j^i\right|$.

$RMSE = [1/IJ\sum_{j=1}^{J}\sum_{i=1}^{I}(Sim_j^i - Obs_j^i)^2]^{1/2}$.

$IOA = 1 - [IJ \cdot RMSE^2 / \sum_{j=1}^{J}\sum_{i=1}^{I}(\left|Sim_j^i - \overline{Obs}\right| + |.Obs_j^i - \overline{Obs}|.)]$, where $\overline{Obs}$ is the average observed concentration and a value of 1 indicates perfect agreement between predicted and observed values.

[a] All 6 sites including Shanghai, Nanjing, Hangzhou, Shengsi, Lusi, Hongqiao from NCDC within domain D3 were used for MM5 evaluation.

year) exceeding the 160 $\mu g\ m^{-3}$ maximum-8-hour ozone standard. Shanghai Environment Bulletin (http://www.envir.gov.cn/law/bulletin/b2007/en/en_0005.html) reported there were 10 days in 2006 that NO_2 was monitored as the daily primary pollutant, and 17 days for SO_2 as the daily primary pollutant, both of them are lower than the model predictions. However, daily primary pollutant was the pollutant with highest index on that day so it was unnecessarily indicating that the pollutants concentrations may exceed air quality standards. So the API data for SO_2 and NO_2 provided by local agency in Shanghai were converted to ambient concentrations and examined to investigate the exceedance within the city. We found there were only 6 days (2% of the year) when daily SO_2 exceeded the standard, and there were 52 days (14% of the year) when daily NO_2 exceeded the standard. So model prediction of exceeding days from CMAQ agreed well with local observations for NO_2, but was inconsistent for SO_2. The large discrepancy for SO_2 may due to the uncertainty within deriving SO_2 concentration from API (Wang et al., 2006), and also the uncertainties within emission inventory may also lead to less accurate daily fluctuations from model. In addition, other studies using CMAQ also reported overestimation of SO_2 over YRD (Li et al., 2011b; Wang et al., 2012). Huang et al. (2011) developed YRD emission inventory for year 2007 and reported the annual emission for NO_x, SO_2, VOC, and $PM_{2.5}$ is 2293, 2392, 3767, and 1510 Gg respectively, while the values from INTEX-B for year 2006 is 3223, 3749, 3641, 1528 Gg respectively. Apparently INTEX-B has very close estimations as the local data for VOC and $PM_{2.5}$, but higher estimations for NO_x and SO_2. So more effort should be devoted to address the uncertainties within emission inventory over YRD and China. So actually instead of O_3 which draws most of the interest and focus in published studies regarding Shanghai, NO_2 was found to be also an important gaseous pollutant for the city because it had the maximum number of days exceeding the standard. While on the other hand, although there are only 9 days for $MDA8O_3$ exceedance, it may not necessarily indicate an optimistic ozone pollution potential in Shanghai because the high NO_2 concentration in urban area usually implies a strong titration effect, which may lead to more exceedance of ozone if control strategy is made simply to reduce NO_2 pollution. As compared to Shanghai, the magnitudes and exceedances of gaseous pollutants were different in Nanjing and Hangzhou. NO_2 was the most important pollutant in Nanjing, with 27 days of exceedance, followed by O_3 and SO_2 which had 14 and 10 days of exceedance, respectively. There was no daily data available for NO_2 and SO_2 at Nanjing and Hangzhou in 2006, but Nanjing Municipal Environmental Protection Bureau reported that in 2006 there were 30 days of NO_2 exceedance and 11 days of SO_2 exceedance (http://www.njhb.gov.cn/art/2007/6/4/art_28_12223.html), suggesting the number of exceeding days predicted from CMAQ in this study agreed well with the local data. In Hangzhou, SO_2 was the most important pollutant with 28 exceedances followed by NO_2 and O_3 with 20 and 11 exceedances, respectively.

In section 2.4, Fig. 4 showed that Shanghai had the highest monthly average NO_2 and lowest O_3, which was consistent with findings in this section that Shanghai has the largest number of NO_2 exceeding days and smallest number of O_3 exceeding days. While there was no public available monitored or reported SO_2 and NO_2 data for Hangzhou, this overall consistency provided important information to review the gaseous pollution in other areas within YRD.

While in Section 2.4, seasonality analysis suggested the most polluted episode for each pollutant in the cities, such as O_3 reached its peak in spring (March) and early summer (June), NO_2, and SO_2 generally spiked in winter (January). However, a higher monthly concentration may not necessarily indicate the higher frequency of violating air quality standards because larger diurnal variation could

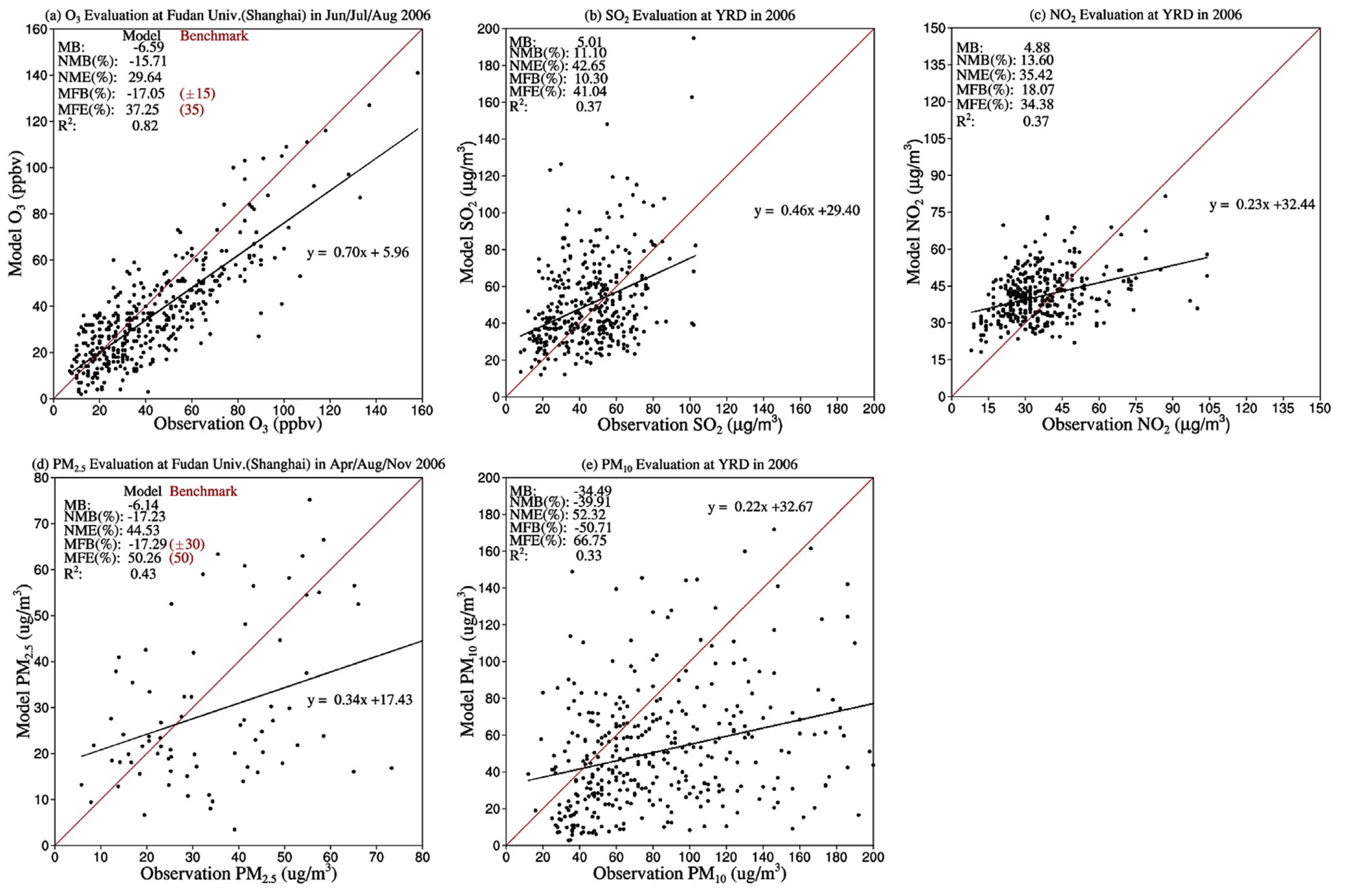

Fig. 3. Statistical evaluations of (a) O_3, (b) SO_2, (c) NO_2, (d) $PM_{2.5}$, and (e) PM_{10}. Normalized Mean Bias (NMB) $= \sum_1^N (\text{Model} - \text{Obs})/\sum_1^N \text{Obs} \times 100\%$, where Model is the CMAQ prediction value of a data pair, and Obs is the observation value of the same data pair, and summations are overall data pairs (N). Normalized Mean Error (NME) $= \sum_1^N |.\text{Model} - \text{Obs}|./\sum_1^N \text{Obs} \times 100\%$; Mean Fractional Bias (MFB) $= \frac{2}{N}\sum_1^N \text{Model} - \text{Obs}/\text{Model} + \text{Obs} \times 100\%$; Mean Fractional Bias (MFE) $= \frac{2}{N}\sum_1^N |\text{Model} - \text{Obs}|/\text{Model} + \text{Obs} \times 100\%$; Correlation Coefficient (R^2) $= \sum_1^N(\text{Model} - \overline{\text{Model}}) \times (\text{Obs} - \overline{\text{Obs}})/\sqrt{\sum_1^N (\text{Model} - \overline{\text{Model}})^2 \times \sum_1^N (\text{Obs} - \overline{\text{Obs}})^2}$, where $\overline{\text{Model}}$ is the averaged model prediction, and $\overline{\text{Obs}}$ is the averaged observation.

lead to frequent exceedance and lower monthly averages at the same time. Other than annual exceeding days that showing overall pollution status, monthly exceeding days of CH-NAAQS summarized in Fig. 6 provide more details about gaseous pollution in each season. As shown in Fig. 6(a), Shanghai had exceedance of MDA8O$_3$ standards only in May, June, July, and August, and the number of days exceeding MDA8O$_3$ standards in August was 5, which was 56% of the total annual 9 days of exceedance. Ozone exceedance reached

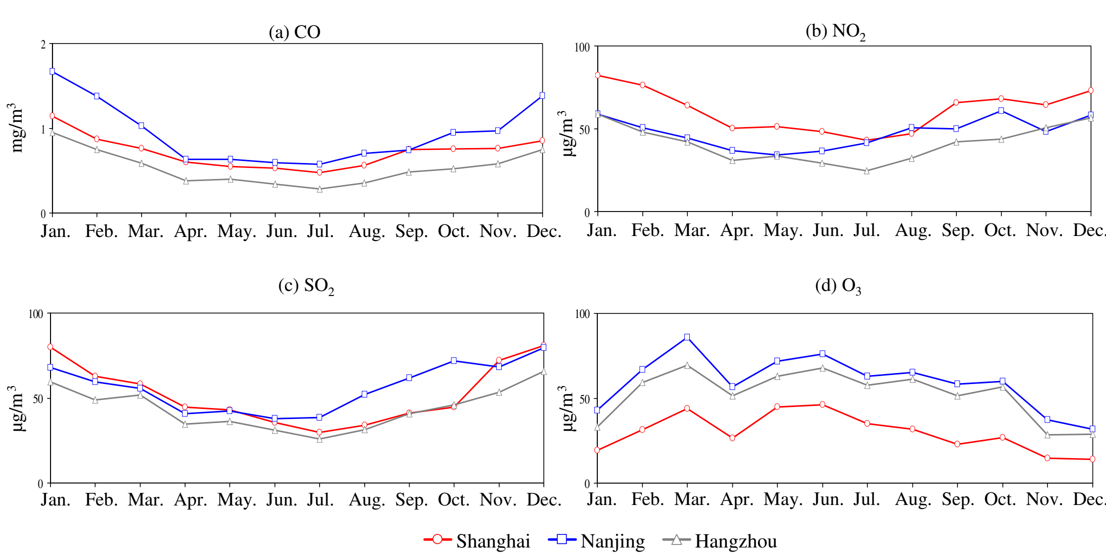

Fig. 4. Monthly average values of CO, NO_2, SO_2, and O_3 in Shanghai, Nanjing, and Hangzhou.

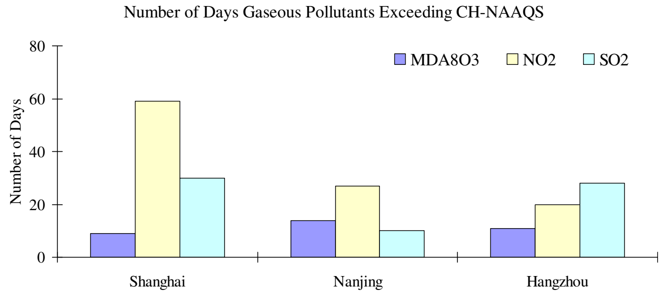

Fig. 5. Annual exceedance of MDA8O$_3$, NO$_2$, and SO$_2$ in Shanghai, Nanjing, and Hangzhou.

to its peak values in March for Nanjing and in June for Hangzhou. In Nanjing, March had the highest monthly average O$_3$ of 85.9 μg m^{-3}, and it also had the largest exceedance as 5 days. Exceedance of O$_3$ in Hangzhou followed a similar pattern but for different specific months. Observation studies have documented the generally high O$_3$ in late spring over YRD: Wang et al. (2005) reported observed daytime average O$_3$ could reach up to 65 ppb in April 2000, while the highest value was only 49 ppb in July; Cheung and Wang (2001) reported there were 10 and 3 days in May and July 2000, respectively, that MDA8O3 exceed 80 ppb. However, in Shanghai, as shown in Fig. 4(e) and Fig. 6(a), monthly averaged O$_3$ peaked in June as 46.2 μg m^{-3}, but the maximum exceedance happened in August when monthly averaged O$_3$ was 31.8 μg m^{-3}. The inconsistency between the monthly averages and the monthly number of days exceeding air quality standards is due to the fact that highest monthly average daytime O$_3$ was in August as 98 μg m^{-3}, also with the largest difference with night time O3 (43 μg m^{-3}), indicating the intensive NO$_x$ emission in Shanghai may lead to both rapid photochemical production and night time titration of O$_3$.

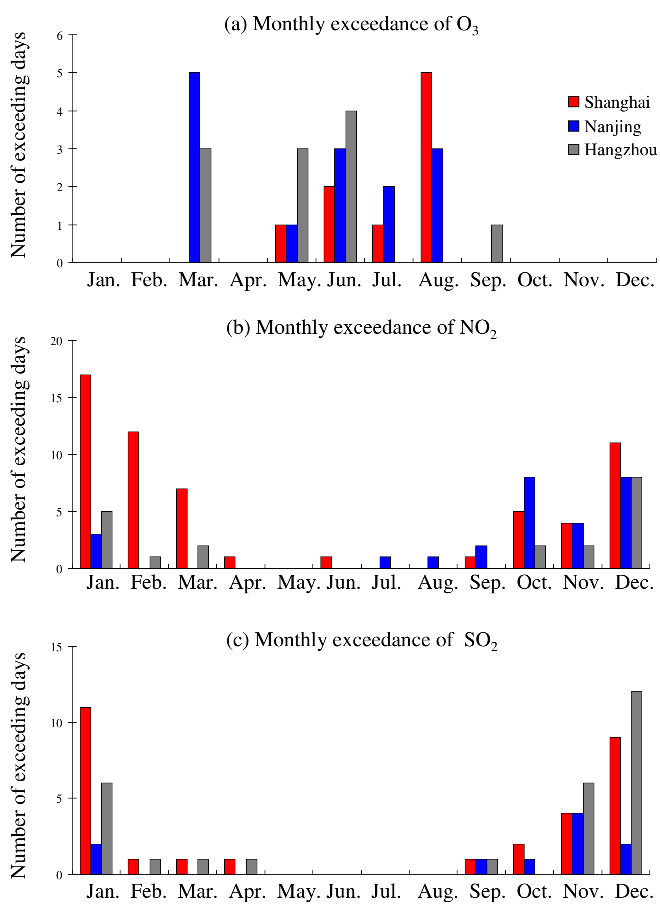

Fig. 6. Monthly exceedance of O$_3$, NO$_2$ and SO$_2$ in Shanghai, Nanjing, and Hangzhou.

Monthly exceedance of SO$_2$ and NO$_2$ were generally consistent with their seasonality in all the three cities. In Shanghai, SO$_2$ and NO$_2$ had more exceedance in winter than summer, and there were exceedance for 9 out of 12 months, indicating pollution for these species was an important issue for Shanghai throughout the year. NO$_2$ exceeded standard in January (3 days), and also from June (1 day) to December (8 days) at Nanjing, but most of the exceedance mainly happened from October to January, indicating late fall and winter were the major NO$_2$ pollution seasons. Hangzhou had NO$_2$ exceedance from January (5 days) to March (2 days) and from October (2 days) to December (8 days), indicating winter and spring were the NO$_2$ pollution seasons. SO$_2$ exceeding days were distributed to more months for in Hangzhou, indicating a longer SO$_2$ pollution season, which lasted from September (1 day) to December (12 days) and January (6 days) to April (1 day). Nanjing was found to have a shorter SO$_2$ pollution season which lasted from September (1 day) to December (2 days) and also January (2 days).

3.2. Air quality benefit assessment of control scenarios

As demonstrated in Section 3.1, SO$_2$, NO$_2$ and O$_3$ were important gaseous pollutants due to their frequent days violating air quality standards in YRD, so the predefined emission control scenarios discussed in Section 2.2 are analyzed in this section to assess the air quality benefits in terms of the reduction of exceeding days for these gaseous pollutants.

3.2.1. SO$_2$ benefit from power plant SO$_2$ emission control

Power plant was the primary emission source for SO$_2$ in YRD, which contributed 61.9% of the total anthropogenic emissions. P-S90 scenario (described in Section 2.2 and Table 2) which removed 90% of SO$_2$ emission from power station sector over YRD is designed to assess its air quality benefit in terms of reducing ambient SO$_2$ concentration in YRD, as shown in Fig. 7. The total annual exceedance of SO$_2$ in Shanghai decreased from 30 days in the base case to only 5 days in this control case. The number of exceedance in Nanjing decreased from 10 to 0, and the number of Hangzhou decreased from 28 to 4. Clearly, SO$_2$ emission control from power plant sector would greatly benefit the air quality in YRD regarding reducing the ambient SO$_2$ concentration and meet the new CH-NAAQS requirement. CMAQ predictions for control scenario P-S90 generally reproduce the SO$_2$ benefits well: According to reported data published by local EPBs, total anthropogenic SO$_2$ emission was reduced by 49.7%, 5.8%, and 7% for Shanghai (from 613 to 358 Gg), Nanjing (from 146 to 116 Gg), and Hangzhou (from 121 to 85 Gg) from 2006 to 2010, respectively. It was also reported that applying FGD to coal-fire power plants was the predominant control solution during the 11th 5 YP. According to local environmental bulletins in 2010, there were no exceeding days for SO$_2$ in these cities in 2010. Apparently, the real control efforts during 11th 5 YP showed the expected air quality benefits by reducing the number of exceeding days greatly for SO$_2$ pollution.

3.2.2. NO$_2$ benefit from NO$_x$ emission control

NO$_2$ is a both primary and secondary pollutant and could be emitted from many different anthropogenic sources, such as vehicles and fossil-fuel combustion. As reactive nitrogen species in the troposphere and a precursor of NO$_2$, NO has a very short lifetime as only a few seconds, and NO$_2$ also has a short lifetime which is approximately 1 day with respect to conversion to nitric acid

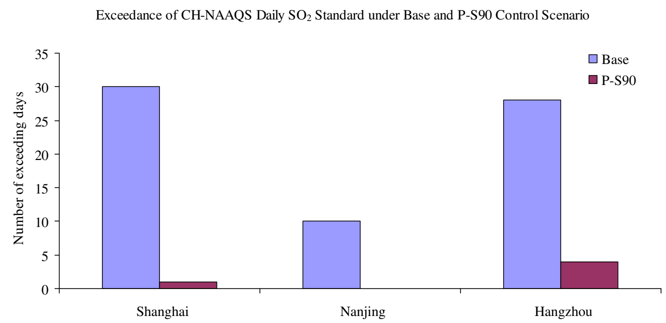

Fig. 7. SO_2 exceedance under base and P-S90 control scenario in Shanghai, Nanjing, and Hangzhou.

(Seinfeld and Pandis, 1998). So controlling NO_x emission was expected to reduce NO_2 pollution effectively. In this section, air quality benefits in terms of reducing the number of days exceeding NO_2 standard are analyzed for NO_x control scenarios T-N20, I-N20, and P-N85.

As shown in Fig. 8(a), while compared with base case, numbers of days exceeding CH-NAAQS daily NO_2 standard decreased under all the NO_x control scenarios, except for T-N20 scenario in Nanjing. Monthly average NO_2 concentration at Nanjing decreased by up to 3% from 16.8 ppbv under base case to 16.2 ppbv under T-N20 case, indicating that emission control might lead to lower concentration but not necessarily result in less exceedance. In order to better understand the impact of each control scenario, the percentage decrease of NO_2 exceeding days as compared with base case was also calculated and shown in Fig. 8(b). NO_x emission control from power plant was found to have most significant impact, which reduced the number of NO_2 exceeding days by 66%, 74%, and 90% for Shanghai, Nanjing, and Hangzhou, respectively. One of the reasons for different impacts of the control scenarios was likely due to the different amount of emission from each sector. As shown in Fig. 2, power plant contribute to 42.3% of anthropogenic NO_x emission in YRD, while industry and transportation contribute to 28.5% and 24.7% respectively, so P-N85 caused largest changes of NO_2 mainly because it reduced the largest amount of total NO_x, followed by industry and then transportation. Another reason was the location of the emission sources over YRD. Power plants were point sources which located mainly in some certain grids close to the city areas, industry emission was also usually restricted to limited areas, while transportation emission spread all over the YRD domain (Du, 2008). Thus, removing an intensive point source would be likely more effective than removing a well distributed area or line source in terms of reducing the number of exceeding days.

3.2.3. *Ozone benefit from NO_x/VOC emission control*

O_3 response under emission control could be complicated to predict due to its non-linearity (Seinfeld and Pandis, 1998; Fu et al., 2012). As mentioned in Section 2.2, NO_x emission reduction aiming at reducing NO_2 pollution may have side-effect by increasing O_3 because of the titration effect. Therefore in this section, all NO_x and

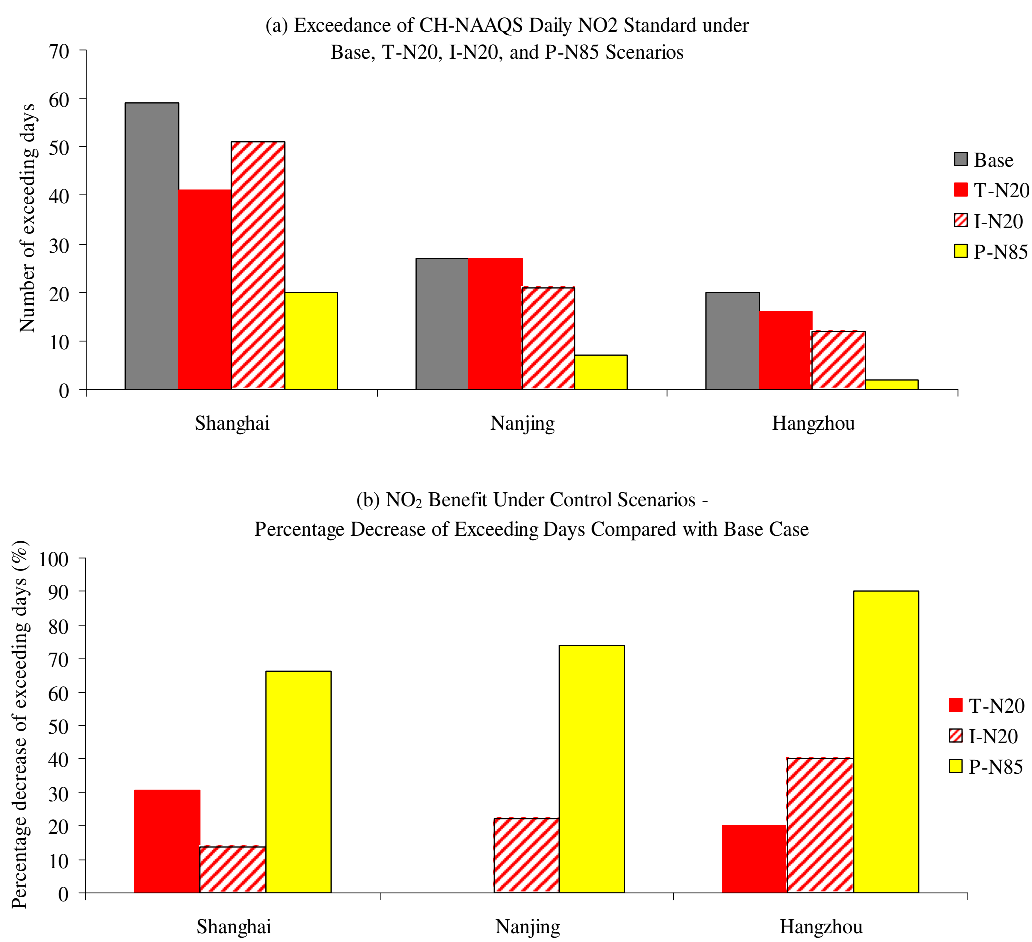

Fig. 8. NO_2 benefit under T-N20, I-N20, and P-N85 scenarios in Shanghai, Nanjing, and Hangzhou.

VOC involved scenarios (i.e. I-N20, I-V20, I-C20, T-N20, T-V20, T-C20, and P-N85) are analyzed to assess the O_3 response in terms of number of days exceeding CH-NAAQS MDA8O_3 standard, as shown in Fig. 9.

As shown in Fig. 9(a), controlling NO_x emission independently (T-N20, I-N20, P-N85) will lead to more exceeding days of O_3 pollution in all the three cities. Fig. 9(b) demonstrated the percentage difference of exceeding days between base case and control scenarios. In Shanghai, number of O_3 exceedance would be increased by 11%, 33%, and 78% under T-N20, I-N20, P-N85 scenarios, respectively. Meanwhile, independent VOC emission removal scenarios were found to be able to reduce the O_3 pollution efficiently. O_3 exceedance could be reduced by 33%, 56% under T-V20 and I-V20 scenarios, respectively. So in Shanghai, a more possibly applicable and effective control strategy might be reducing emission of NO_x and VOC simultaneously to meet both O_3 and NO_2 standards. Under scenarios T-C20 and I-C20 which both NO_x and VOC emission were reduced by 20% from transportation and industry, respectively, the overall O_3 exceedance was decreased by 11% and 44% respectively, indicating that the O_3 reduction effect from VOC control was counteracted by the side-effect of NO_x control. O_3 response in Nanjing was different from that in Shanghai, emission control scenarios defined in this study were found to have very limited impacts in reducing the number of days exceeding the new air quality standards. As shown in Fig. 4(a), Nanjing had the highest monthly average O_3 among all three cities, which was consistent with the results shown in Fig. 5, suggesting Nanjing had the largest number of days exceeding MDA8O_3 standard. While as shown in Fig. 9, independent NO_x emission control (T-N20, I-N20, P-N85) would increase the number of exceeding days for MDA8O_3, while independent VOC emission control (T-V20, I-V20) could not reduce the exceeding days, indicating the peak concentrations of daily MDA8O_3 under the control scenarios are still higher than the 160 $\mu g\ m^{-3}$ standard. The only effective scenario over Nanjing was T-C20 which reduced the number of exceeding days by 7% (1 day), indicating that simultaneous control for NO_x and VOC from transportation emission could be applied in Nanjing to reduce the local O_3 pollution. O_3 response under control scenarios in Hangzhou was similar to that in Shanghai, where independent NO_x emission control would result in more exceeding days, and independent VOC emission control would reduce the number of exceeding days. The only difference happened for control scenario T-C20 which removed 20% of NO_x and VOC emission simultaneously from transportation sector. Number of exceeding days was reduced by 11% (1 day) under T-C20 scenario in Shanghai, but not changed in Hangzhou, indicating the VOC control effect was totally counteracted by the NO_x emission control. It is also important to notice that although O_3 response might be similar in three cities under same scenario, changes of exceeding days may vary since same emission reduction rate may indicated large difference of reduction amount. As Shanghai has highest power plant emission, removing 85% NO_x under P-N85 scenario lead to more exceeding days in Shanghai than other two cities.

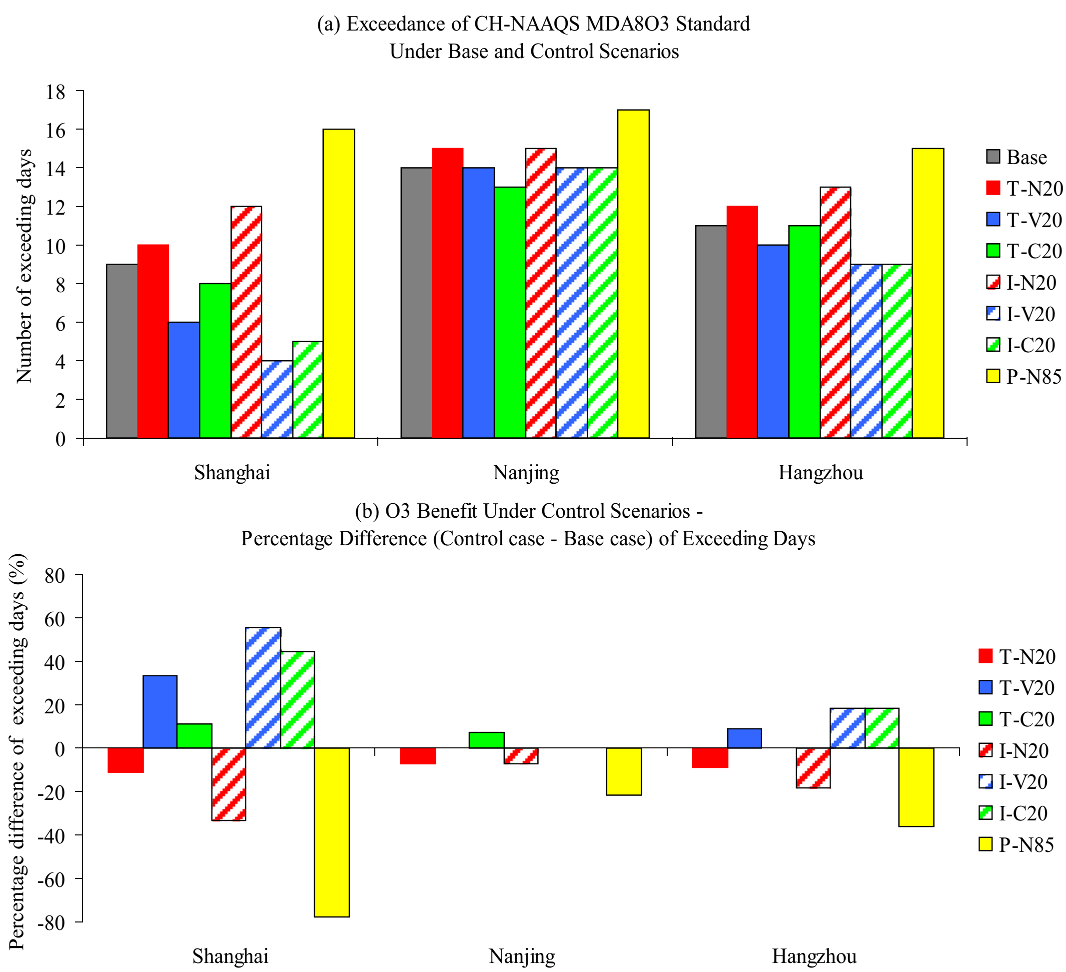

Fig. 9. O_3 benefit under I-N20, I-V02, I-C20, T-N20, I-N20, and P-N85 scenarios in Shanghai, Nanjing, and Hangzhou.

4. Conclusion

MM5/CMAQ modeling system was applied in this study and demonstrated to be able to reproduce both meteorology and air quality well for the year 2006 based on the statistically evaluated results in comparison to observations. Performance statistics showed that model simulations have an overall good agreement with both API value and hourly O_3 and daily $PM_{2.5}$ observations at Fudan University site. Monthly average concentrations of NO_2 and SO_2 showed higher values in winter, while O_3 was found to have higher values in spring and summer, suggesting the different polluted seasons for different pollutants. The major pollution episodes for SO_2 and NO_2 occurred in winter due to the weak dispersion, while the major pollution episode for O_3 was late spring in Nanjing and Hangzhou, and summer in Shanghai, due to intensive photochemical production.

Examination of air quality in Shanghai, Nanjing, and Hangzhou according to the new published CH-NAAQS provide important information for determining the attainment/non-attainment status over YRD: In Shanghai, NO_2 was found to have the largest number of days exceeding air quality standards (59 days), followed by SO_2 (30 days) and MDA8O_3 (9 days); In Nanjing, exceeding days of NO_2 is highest (27 days), but MDA8O_3 showed more exceeding days (14 days) than SO_2 (10 days); While in Hangzhou, SO_2 was the most important pollutant (28 exceeding days), followed by NO_2 (20 days) and MDA8O_3 (11 days). Reported exceeding days from local EPB bulletins were compared with predictions from CMAQ, and it was demonstrated that our findings agreed well with reported data for NO_2 exceedance in Shanghai, and for both NO_2 and SO_2 exceedance in Nanjing. The number of SO_2 exceeding days was overestimated by CMAQ, possibly due to uncertainties within temporal profile of emission inventory. The difference of primary gaseous pollutant may imply the diversity of emission pattern among the cities within YRD, but clearly suggest control efforts should be designed uniquely in order to target the most important one.

Results of gaseous pollutants responses under potential applicable emission control scenarios shed light on developing multi-pollutants emission control strategies in order to improve local air quality over YRD to meet the new CH-NAAQS. 90% SO_2 emission removed from power plant sector was found to be an effective solution to reduce SO_2 pollution over YRD: the number of SO_2 exceeding days in Shanghai was reduced from 30 to 1 under P-S90 scenario, from 10 to 0 in Nanjing, and from 28 to 4 in Hangzhou. This modeling result generally agreed well with the assessment of air quality in 2010 that there was no SO_2 exceedance due to control efforts during 11th FYP. NO_x emission control from power plant, transport, and/or industry sectors were also found to be effectively reduce the number of NO_2 exceeding days over YRD, with NO_x emission control from power plant was proved the most efficient strategy. While emission removal from transportation and industry showed similar effects under same emission reduction rate, although industry emission control was found slightly more efficient at Nanjing and Hangzhou due to higher amount of emission removed from this sector. However, independent NO_x emission control strategies were also found to strengthen O_3 pollution as a side-effect, and O_3 pollution was worse when more NO_x emission was controlled, indicating a strong titration effect over YRD. Since both NO_2 and O_3 are criteria pollutants listed in CH-NAAQS, multi-pollutants control is suggested as the best approach to improve local air quality in the cities. With 20% of NO_x and VOC emission control from transport or industry, the number of MDA8O3 exceeding days in Shanghai was reduced by 11% and 44%, respectively.

Although this study provided important information about gaseous pollutants over YRD regarding their seasonality and exceedance of air quality standards, the discrepancies between CMAQ prediction and monitored data indicated that continuous effort should be emphasized to address the uncertainties of emission inventory. Moreover, air quality benefit assessment in this study is performed mainly based on the simulation results, so uncertainties from modeling might also introduce inaccuracy to the predicted exceedance of air quality standards under both base and sensitivity scenarios. While model evaluation demonstrate the overall good accuracy of prediction, a small discrepancy might change a daily air quality from exceeding to non-exceeding status or vise versa, so a multiyear study is necessary in the future to provide a more comprehensive and stable assessment of air quality status and benefit from emission control over YRD. In addition, gaseous pollutants responses under pre-defined control scenarios shed light on potential control strategy design over YRD, but China's emission is always changing fast due to the rapid growth of industry and manufactory. So developing real air quality control policy for current and future years would require more modeling and observation effort for recent year(s).

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