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A numerical study of contributions to air pollution in Beijing during CAREBeijing-2006

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Abstract. An online air pollutant tagged module has been developed in the Nested Air Quality Prediction Model System (NAQPMS) to investigate the impact of local and regional sources on the air pollutants in Beijing during the Campaign of Air Quality Research in Beijing 2006 (CAREBeijing-2006). The NAQPMS model shows high performance in simulating sulfur dioxide (SO_2), particulate matter (PM_{10}), nitrogen dioxide (NO_2), and ozone (O_3) with overall better agreements with the observations at urban sites than rural areas. With the tagged module, the air pollutant contributions from local and regional sources to the surface layer (about 30 m) and the upper layer (about 1.1 km) in Beijing are differentiated and estimated. The air pollutants at the surface layer in Beijing are dominated by the contributions from local sources, accounting for 65 % of SO_2, 75 % of PM_{10} and nearly 90 % of NO_2, respectively, comparatively, the 1.1 km layer has large source contributions from the surrounding regions (e.g., southern Beijing), accounting for more than 50 % of the SO_2 and PM_{10} concentrations. County scale analysis is also performed and the results suggest that Tianjin is the dominant source of SO_2 in Pinggu County, and Langfang, Hebei is the most important regional contributor to PM_{10} in Beijing. Moreover, the surrounding regions show larger impact on SO_2, PM_{10} and NO_2 in the eastern counties of Beijing (e.g., Pinggu, Tongzhou and Daxing) than those in western Beijing, which is likely due to the Beijing's semi-basin topography and the summer monsoon. Our results indicate that the efforts to control the air pollutants in Beijing should focus on controlling both local and regional emissions.

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1 Introduction

In order to improve the air quality in Beijing, the Campaign of Air Quality Research in Beijing (CAREBeijing), which was initiated by the Beijing Municipal Environmental Protection Bureau (Beijing EPB) and Peking University (PKU) in conjunction with other eighteen institutes and research centers, was conducted in August from 2006 to 2008. The present numerical study is to provide an in depth quantitative analysis on the contributions from local and regional sources to the air pollution in Beijing.

Air pollution in Beijing is regional in nature and not attributable only to local emissions (Streets et al., 2007), that is due to its semi-basin geophysical feature and unique weather pattern (Ren et al., 2003). Making insights into the main pathways of the surrounding air pollutants contribution to Beijing is therefore vital for the establishment of effective control strategies on major pollutants reduction in the megacity. Xu et al. (2005) investigated the transport pathways and found a convergence zone of air pollutants along the Yanshan-Taihang Mountains. However, accurate identification of the major pollutants sources and transport processes to thoroughly understand the mechanisms behind air pollution over Beijing remains a challenging task so far.

Recently, An et al. (2007) applied the CMAQ model to study a heavy particulate and sulfur episode observed in spring in Beijing by using emission switch on/off method, in his results, the contribution to the fine particles in Beijing was about 39 %∼53 % from the northwest (notably from Zhangjiakou), and 15 % from the southwest (notably from Baoding). Similarly, Streets et al. (2007) reported that $PM_{2.5}$ concentrations from Shandong Province might reach a maximum contribution of 63 % to Beijing under certain

meteorological conditions, Tianjin also exerted a significant influence on Beijing $PM_{2.5}$ concentration with the maximum contribution of 42 % and the average contribution of 13 %. Comparatively, the influences from regional sources on ozone were less pronounced, with maximum contributions of 28 % from Hebei, 27 % from Tianjin, and 24 % from Shandong province, but not at the same time. According to Chen et al. (2007) of the MM5-CMAQ modeling with five emission scenarios in the months of January, April, July and October in 2002, much efforts should be placed on demanding more pollution reduction and better emission control from surrounding provinces, while Beijing needed to take positive steps to reduce itself own emissions. These methods commonly used to estimate the contribution of targeted emissions, highlight significant bias in non-linear source-receptor relationships analysis. And the potential contributions at the different layers were not described.

This study, through simulations carried on the NAQPMS model, adopts air pollutant tagged module to estimate the contributions of local and regional emissions to the air pollutants in Beijing at the different layers. More detailed scales (by dissociating the whole Beijing and counties) analysis on air pollutants transport from regional emissions to Beijing, is also presented. That remains an extremely important issue for air quality management and emissions control efficiency. Section 2 describes the model and evaluation, Sect. 3 presents results analysis and Sect. 4 brings out conclusions and environmental implications.

2 Model description and validation

2.1 Model description

The Nested Air Quality Prediction Model System (NAQPMS) is a fully modularized, three-dimensional chemical transport model over regional and urban-scale air pollution (Wang et al., 2001, 2002, 2006). The chemical transport module reproduces the physical and chemical evolution of reactive pollutants by solving the mass balance equation in the terrain-following coordinates, and includes advection and diffusion processes, gas/aqueous chemistry and dry/wet deposition processes. More details are given elsewhere (Wang et al., 2002, 2006; Wu, 2010; Li et al., 2011), and its gas chemistry module is updated to CBM-Z (Zaveri and Peters, 1999) in Li et al. (2007). The air pollutant tagged module was developed and coupled into the NAQPMS model (Li et al., 2008) and described in Sect. 2.2. The model has been applied to study the issues related to the transport and chemical processes of air pollutants and the interaction between mineral aerosols and acid rain over east Asia in the regional scale (Huang et al., 1995; Wang et al., 2002, 2006; Li et al., 2007), and also the impact of mobile sources on air quality in Beijing in the urban scale (Wu et al., 2010).

The NCAR/PSU Fifth-Generation Mesoscale Model (MM5) (Grell et al., 1995) Version 3.6 is applied as the meteorological driver for the chemical transport module. The National Centers for Environmental Prediction (NCEP) global final analysis data (FNL), with $1^\circ \times 1^\circ$and four times a day, are used for the meteorological initial and boundary conditions. The select schemes in this study were the simple ice for explicit moisture, Grell cumulus, MRF for PBL and cloud scheme for atmospheric radiation according to the sensitivity numerical experiments in (Gao et al., 2007).

Three nested domains for horizontal resolution of 81 km, 27 km and 9 km with the center location at (35.0° N, 110° E) were used for both MM5 and NAQPMS model (Fig. 1). The side boundary condition for the D1 domain of the NAQPMS model was clean boundary condition. Vertically, the meteorological driver of the NAQPMS model had 20 pressure vertical layers extending from 1000 hPa to 100 hPa, and it was interpolated from 23 sigma layers of the MM5 model using INTERPB module.

The simulation was performed for the period of 20 July∼31 August 2006, and the first twelve days were used as spin-up time to reduce the influence of the initial concentration to the NAQPMS model. The influence of the initial conditions will be decreasing while the simulation of the air pollutant model goes further (Seinfeld and Pandis, 2006). In addition, according to Colle et al. (1999); Lo et al. (2007), the MM5 model might take at least 12 hours to spin up when it was initialized with a cold start. Thus, we finished the meteorological simulation with predicting cycle method: in each predicting cycle simulation, we doing 36 h simulation and taking the last 24 h simulation of the MM5 model results, and then linking them together as the meteorological driver of the NAQPMS transport module.

2.2 Air pollutant tagged method

As mentioned above, an online air pollutant tagged module was developed in the NAQPMS model and applied in this study. In this module, we assume that different tagged species (S^{T}) are properly integrated in each grid, each tagged species S^{T} shares the same loss coefficients with total species during outflow, chemical transformations and dry/wet removal processes, and the same as the total species (S) in the removal processes (Kondo et al., 2004; Davis et al., 2003). So all of the removal processes have no alteration on the fraction ($F_{S^{\mathrm{T}}}$) of the tagged species in the total species.

$$S = \sum_{\mathrm{T}=1}^{\mathrm{T}=\mathrm{ntagged}} S^{\mathrm{T}}, F_{S^{\mathrm{T}}} = S^{\mathrm{T}}/S$$

Thus, the tendency of $F_{S^{\mathrm{T}}}$ depends on the production processes and can be described as following:

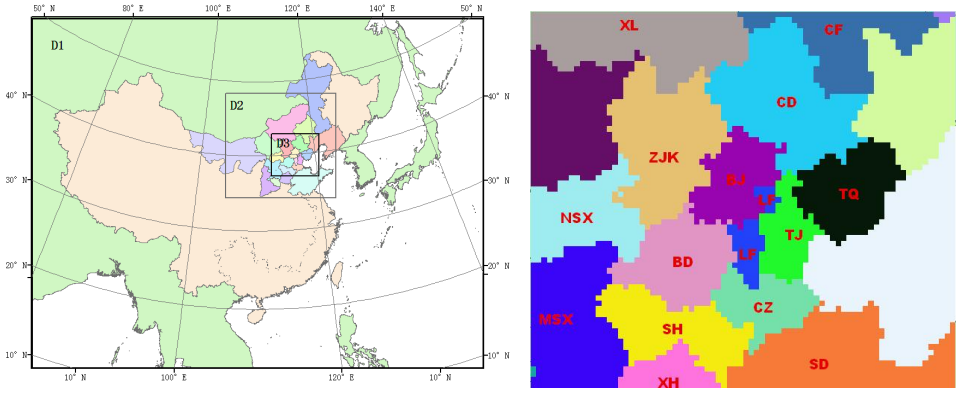

Fig. 1. The three nested domain used for simulation. Left: D1 covers East Asia with 83 × 65 grids; D2 with 61 × 58 grids includes North China; D3 with 79 × 70 grids consists of Beijing and its surrounding cities. Right: source tagged regions: Beijing (BJ); Tianjin (TJ); Tangshan and Qinhuangdao (TQ); Chengde (CD); Zhangjiakou (ZJK); Baoding (BD); Langfang (LF); Shijiazhuang and Hengshui (SH); Cangzhou (CZ); Xingtai and Handan (XH); Xilinguole (XL); Chifeng (CF); Shandong (SD); Middle Shanxi (MSX); Northern Shanxi (NSX).

$$\Delta F_{S^{\mathrm{T}}} = F_{\mathrm{t}} - F_{\mathrm{t}-1} = \frac{S^{\mathrm{T}} + \Delta S^{\mathrm{T}}}{S} - \frac{S^{\mathrm{T}}}{S} = \frac{\Delta S^{\mathrm{T}}}{S}$$

$$(\frac{dF_{S^{\mathrm{T}}}}{dt})_j = ((P_{\mathrm{T},j})_{\mathrm{chem+emis}^{\mathrm{T}}} + (M_{\mathrm{T},j})_{\mathrm{dif+adv+conv}})/S_j$$

where j represents the grid. $P_{\mathrm{T},j}$ is the production of the tagged species in the j^{th} grid, including the gross chemical production and emissions. When the j^{th} grid outside the tagged emission region, the emission is equal to zero. $(M_{\mathrm{T},j})_{\mathrm{dif+adv+conv}}$ is the S^{T} inflow from adjacent grid due to advection, diffusion and convection processes, and calculated by the flow fluxes and the S^{T} fraction in those adjacent grids. Compared with the sensitive analysis method of switching on/off emission, the online tagged method provides a different and more efficient measurement to evaluate the importance of various tagged regions. Similar analysis can also be found in previous works (Davis et al., 2003; Kondo et al., 2004; Sudo and Akimoto, 2007; Li et al., 2008). The Supplement provides more details on the tagged method, for example how the PM_{10} from Baoding and ozone from Tianjin has been tagged.

This study tagged 22 emissions regions in the model domain, according to the regional administrative system and the position relative to Beijing (shown in Table 1): defining Beijing and Tianjin Municipalities, Shandong and Liaoning Provinces as four tagged individual regions, Hebei Province divided into eight regions, Inner Mongolia into five regions and Shanxi Province in four regions, and the remaining neighboring areas are defined as one tagged emissions region and labeled into the NAQPMS model. The lateral, top boundary and initial pollutants are also tagged. The GIS software (Arc-GIS 8.3) was applied to properly provide readable regional labeled database for the model.

2.3 Emission inventory

The emissions from the Beijing and its surrounding areas are especially large (Matsui et al., 2009) and very important for modeling studies. Several anthropogenic emission inventories for this study were developed for the year 2006 with a horizontal resolution of 0.5 × 0.5° by revising those reported by Streets et al. (2003, 2007) using the method described by Zhang et al. (2009). Seasonal dependencies of emissions are not taken into account. Four emission inventories are considered in this study: the regional emissions with 10 km resolution update from TRACE_P emission inventory according to personal communications (Streets et al., 2003), including the large point source, industry, power plant, domestic bio/fossil fuel, transportation, livestock emissions and the biomass burning in the month of January/April/July, and the biogenic emissions, which can be found in the website: http://www.cgrer.uiowa.edu/EMISSION_DATA/; the power plant emission database of Beijing and its surrounding provinces including Tianjin, Hebei, Shanxi, Inner Mongolia and Shandong provinces from the Chinese Research Academy of Environmental Sciences (CRAES) was used to update the power plant emission in North China for the year 2006; the main industrial emissions in Tianjin and Hebei provinces from Beijing University of Technology (BUT) and CRAES; the detailed local emissions database of Beijing city from Beijing EPB, including the domestic/transportation/industry emission, was also used to update. The amount of emissions used is presented in Table 2. For all species listed in Table 2, the emissions used are greater than those reported by Streets et al. (2003) by a factor of 1.6 to 2.7 in the inner model domain (D3). These increases are mostly due to actual emissions growth since 2002, in addition to improvements in estimation methodology provided by Zhang et al. (2009).

Table 1. Tagged regions and its ID.

ID	Description
BJ	Beijing
TJ	Tianjin
TQ	Tangshan and QinHuangdao, Hebei
CD	Chengde, Hebei
ZJK	Zhangjiakou, Hebei
BD	Baoding, Hebei
LF	Langfang, Hebei
SH	Shijiazhuang and Hengshui, Hebei
CZ	Cangzhou, Hebei
XH	Xingtai and Handan, Hebei
CF	Chifeng, Inner Mongolia
XL	Xilinguole, Inner Mongolia
MIM	Middle Inner Mongolia
NIM	Northern Inner Mongolia
WIM	Western Inner Mongolia
NSX	Northern ShanXi
MSX	Middle ShanXi
WSX	Western ShanXi
Chz	Chingzhi, ShanXi
LN	Liaoning province
SD	Shandong province
Other	Other regions

Table 2. Emission of major anthropogenic species (Unit: 10^3 tons yr^{-1}). Beijing (BJ); Tianjin (TJ); Hebei (HB); Inner Mongolia (IM); Shanxi (SX); Shandong (SD).

Region	SO_2	NO_x	PM_{10}	$PM_{2.5}$	VOCs
BJ	172.5	236.2	162.5	67.9	289.2
TJ	340.6	387.0	113.7	90.9	204.1
HB	2112.0	1469.4	1508.7	1085.6	1567.6
IM	1491.1	1760.3	762.3	586.5	405.5
SX	2736.2	976.8	1093.3	791.7	336.7
SD	3319.3	1259.3	1619.2	1163.0	787.6

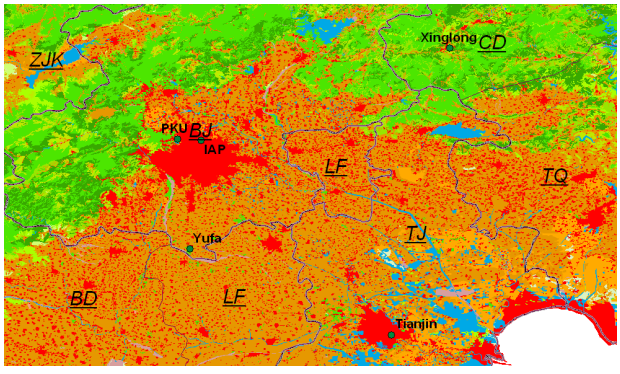

Fig. 2. The location of the observation stations (IAP, Yufa and Xinglong), red shaded indicates urban area, dark green indicates forest, light green indicates grass, orange indicates crop, blue indicates water.

2.4 Model evaluation

In order to estimate the model performances, the statistical parameters were introduced and defined in appendix. Sim(i) and Obs(i) represent the simulated and the corresponding observed variables respectively.

The observed air quality data were provided from the campaign observation sites and the National Standard Air Quality Observation Stations (NSAQ stations) in urban and outer Beijing during CAREBeijing-2006 (Garland et al., 2009). Details of the observation sites and measurements can be found in JGR special section paper about CAREBeijing-2006, such as (Garland et al., 2009; Takegawa et al., 2009). The model evaluation about SO_2, NO_2, PM_{10} and O_3 with the observed hourly data at IAP tower station (at 120 m and 280 m height), and SO_2, NO_2 and O_3 at Yufa and Xinglong stations are presented in this study. The IAP tower station locates at the north of the urban Beijing, and its meteorological tower has 325 m height, the air pollutant measurements has been setup in the height of 47 m, 120 m and 280 m, which marked as Layer-1(47 m), Layer-2(120 m) and Layer-3(280 m). We would refer the observation from the height of 120 m in the IAP tower station as the Layer-2, and the height of 280 m in the IAP tower station as the Layer-3 in the paper followed. The observation sites are shown in Fig. 2, of which the IAP tower station is located in the urban Beijing, and the Yufa station to the south of Beijing. Xinglong stations are located at the east of Beijing, where the air pollutants transport from the south and the east respectively were monitored.

Statistical parameters were used to evaluate the model results. Detailed comparisons between the hourly observed and simulated SO_2, PM_{10}, NO_2 and Ozone at the IAP tower station with the scatter plots are presented in (Fig. 3), that illustrate the model performance in the urban areas of Beijing. The scatter plot of the SO_2 concentration indicated that the NAQPMS model results overall agreed with the observation in urban Beijing with the correlation coefficients of 0.78 at Layer-2 and 0.73 at Layer-3, respectively. And the observed mean concentrations were 17.97 ppbv and 12.21 ppbv at the Layer-2 and Layer-3 respectively, which are close to the simulated values of 22.45 ppbv and 15.17 ppbv respectively. The vertical characteristics that the observed SO_2 concentration at the lower layer (Layer-2) was higher than the higher layer (Layer-3) was also simulated well. The model showed a high model performance on SO_2. At the Layer-2, the simulated NO_2 concentration (mean: 35.16 ppbv) was close to the observed (mean: 35.53 ppbv) according to Fig. 3, and the simulated PM_{10} and ozone concentration (shown in Fig. 3) were also highly correlated to the observations ($r = 0.86$, 0.88 and 0.85 respectively). However, the simulated PM_{10} averaged concentration was 97.61 $\mu g\,m^{-3}$, which was much higher than the observed of 68.49 $\mu g\,m^{-3}$, implying a higher

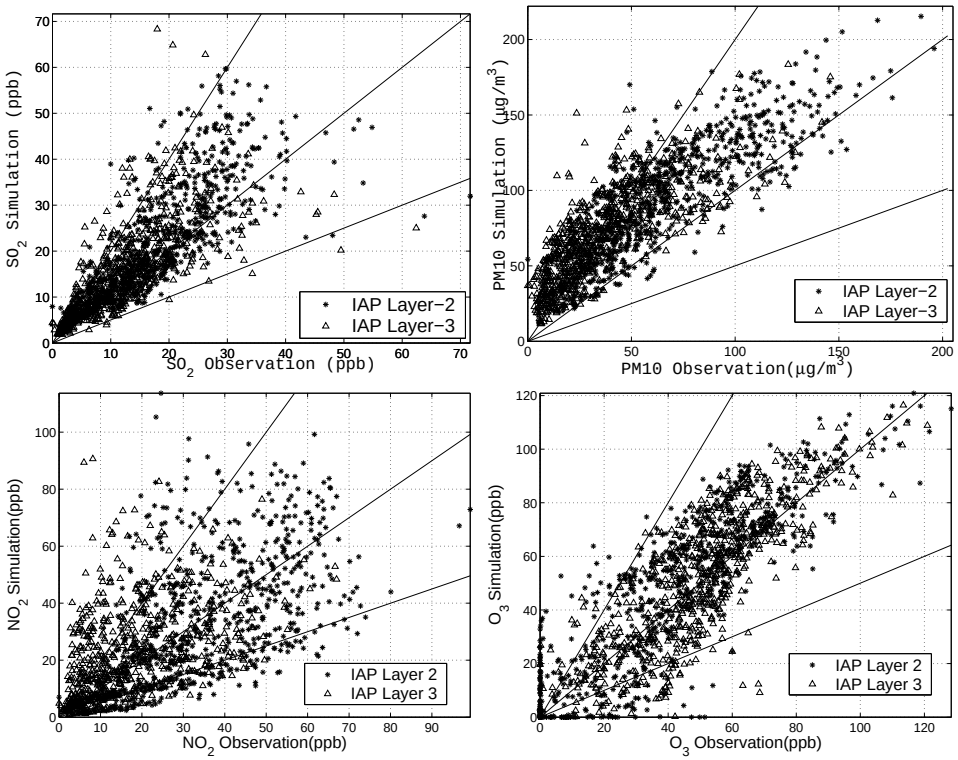

Fig. 3. The scatter plots of SO_2, PM_{10}, NO_2 and O_3 at IAP tower station: three lines indicate $y = 0.5x$, $y = x$ and $y = 2x$. The meteorological tower in IAP tower station has 325m height, the "Layer-2" indicates the air pollutant measurements was setup in 120 m height, while the "Layer-3" in 280 m height.

RMSE of about 35.89 μg m^{-3}. This controversy might be due to some environmental measures implemented in Beijing for reduction of PM_{10} emissions in the urban Beijing (Hao et al., 2008) were not considered in the emissions inventory. Compared with the PM_{10}, the average concentration of simulated ozone seems to be closer to the observation at the Layer-3. The mean bias and normal mean bias of ozone concentration at the Layer-2 are 6.42 ppbv and 0.27 respectively, in comparison to 1.72 ppbv and 0.06 at the Layer-3, suggested that the model had a better model performance at the Layer-2.

As shown in the Table 3 and Table 4, the simulation in this study appears to be more accurate than many previous numerical studies in Beijing, such as (An et al., 2007) and (Matsui et al., 2009). The updated emission data significantly reduced the model biases and errors, and the model results in urban Beijing were reasonable for the evaluation of the air pollutant contribution from different sources.

However, the model results showed worse performance at other sites (Yufa and Xinglong). Despite the high correlation coefficients of NO_2 (0.58) and O_3 (0.62) at Yufa station, most statistical parameters described in Table 5 for the model performance at Yufa and Xinglong stations were weaker than those at the IAP tower station, e.g. the normal mean bias at

Table 3. Model evaluation at the Layer-2 of IAP tower (SO_2, NO_2 and O_3 unit: ppbv; PM_{10} unit: μg m^{-3}).

	SO_2	PM_{10}	NO_2	O_3
Obs mean	17.97	68.49	35.53	47.47
Sim mean	22.45	97.61	35.16	53.88
MB	4.47	29.12	0.38	6.42
MAGE	6.34	30.35	14.93	13.16
RMSE	9.41	35.89	19.50	16.54
NMB	0.35	0.57	0.01	0.27
NME	0.43	0.59	0.48	0.46
FB	0.12	0.19	0.08	0.05
R	0.78	0.85	0.56	0.85

Xinglong site was greater than that at the IAP tower station. That means the model has the different performance in the urban and rural sites, the IAP tower station was a urban sites, the Yufa and Xinglong were the rural sites. That might due to the urban area has more detailed emission data than the rural.

Even so, from the time series plots of the observed/simulated SO_2, NO_2 and ozone concentration (Fig. 4), we could find the NAQPMS model actually

Table 4. Model evaluation at the Layer-3 of IAP tower (SO_2, NO_2 and O_3 unit: ppbv; PM_{10} unit: $\mu g\,m^{-3}$).

	SO_2	PM_{10}	NO_2	O_3
Obs mean	12.21	34.39	16.38	50.89
Sim mean	15.17	68.48	21.35	52.60
MB	2.95	34.10	4.97	1.72
MAGE	4.90	34.41	10.77	11.70
RMSE	7.68	39.36	15.66	14.75
NMB	0.42	1.70	0.76	0.06
NME	0.53	1.71	1.03	0.29
FB	0.12	0.38	0.11	0.1
R	0.73	0.78	0.49	0.81

simulated well the trends of the pollutants, especially ozone. The model results not only showed good agreements in trends, but also the level of ozone concentration at both Yufa and Xinglong station. The ozone trend depends more on the daily radiation, but the level of concentration is limited by the amount of its precursor concentrations. Meanwhile, the model performance of ozone concentration was better than that of SO_2 and NO_2. Combined with the trends in ozone concentrations, such characteristics indicated the complex photochemical relationships, particularly between NO_2 and ozone, which is supported by many previous studies about photochemical pollution (Streets et al., 2007; Su, 2005). Moreover, the simulated SO_2 at Xinglong station was much higher than the observed in the whole concentration distribution, indicating that the SO_2 emissions to the east of Beijing might be overestimated.

3 Results and discussion

To better understand the impact of regional transport to the air pollutants in Beijing, we identified the potential regional contribution at the different vertical layers as well as the principal direct pathways of non-Beijing pollutants in this study. The monthly mean contribution percentage was estimated at each layer, and the comparisons between different tendencies were presented. In addition, the counties scale analysis was performed to better characterize the regional contribution to the air pollution in Beijing.

3.1 The contribution of the SO_2 concentration to Beijing

In August 2006, four obvious pollution accumulation/extinction processes (3–10, 14–19, 22–26 and 27–29) can be separated through the daily variations of SO_2 contribution (Fig. 5). On 7, 15, 16 and 25 August, there were clear evidence showing significant SO_2 contribution from Tianjin (TJ) with the daily range of 12 %∼25 %. For example, the contribution from TJ under the southeasterly

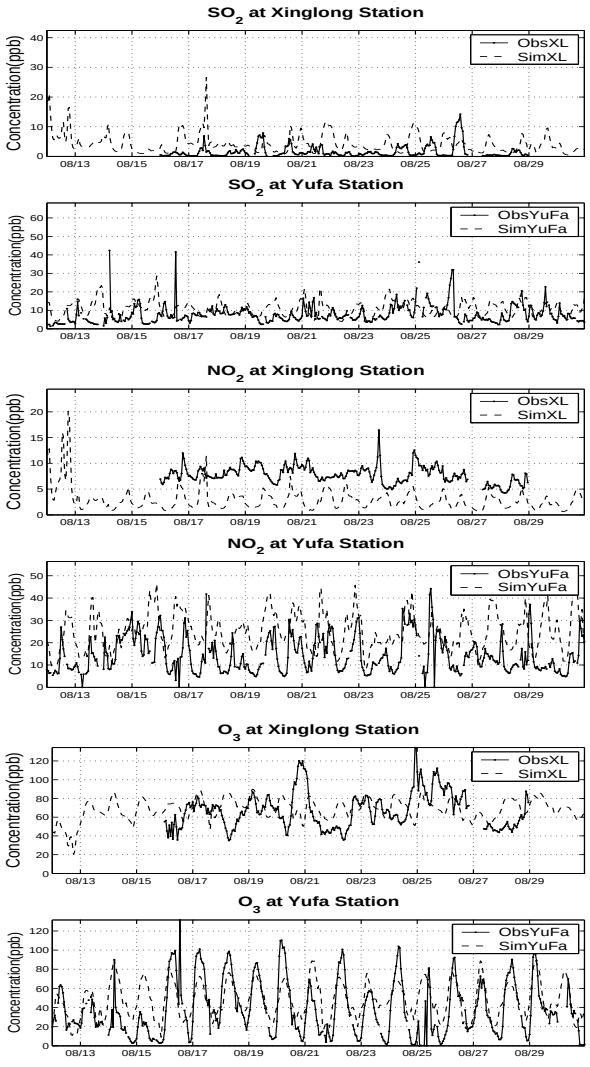

Fig. 4. Temporal variations of observed and simulated SO_2, NO_2 and Ozone (O_3) concentration at Xinglong and Yufa station in August 2006.

winds in the relative high pollution event on August 25 was about 25 % (2/3 of the total surrounding areas contribution). The increase of SO_2 concentration on 22–23 August was associated with the contribution (30 %∼40 %) from Hebei Province (essentially from BD and LF); the non-Beijing contribution in the last sequence (on 27–29 August) seemed to be also dominated by the contribution from TJ, but with less (about 15 %). Such important direct contribution from closer neighboring areas illustrated the important roles surrounding regions played in affecting the air pollution in Beijing. Particularly, the emission controls on industrial and vehicles sources in these surrounding regions are not as stringent as those in Beijing, and the emissions of pollutants are often high.

Table 5. Model evaluation at Xinglong and Yufa station. SN: number of sample, SO_2, NO_2 and O_3 unit: ppbv; PM_{10} unit: $\mu g\,m^{-3}$.

Components	Station	SN	ObsMean	SimMean	RMSE	MB	NMB (%)	*R*
SO_2	Xinglong	290	1.57	4.26	4.67	2.69	17.2	0.40
	Yufa	473	7.70	10.66	7.98	3.00	37.7	0.56
NO_2	Xinglong	300	7.80	2.85	5.43	−4.95	23.4	0.45
	Yufa	474	14.5	24.6	13.8	9.97	66.5	0.62
O_3	Xinglong	300	67.4	70.3	22.3	2.89	4.29	0.46
	Yufa	467	38.5	47.4	22.7	9.62	24.1	0.58

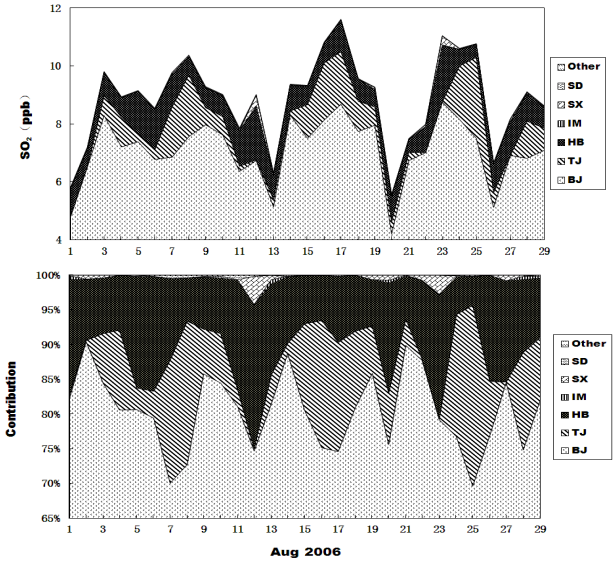

Fig. 5. Time series of the contributions to the daily mean SO_2 concentration (top) over Beijing at the surface layer and its percentage (bottom). Beijing (BJ); Tianjin (TJ); Hebei (HB); Inner Mongolia (IM); Shanxi (SX); Shandong (SD).

Analysis of the monthly mean SO_2 concentrations and its contribution over Beijing is presented in Figs. 6 and 7. Results suggested a significant regional contribution of SO_2 from surrounding areas to Beijing at the 1.1 km layer, mostly from Shanxi, Shandong and Inner Mongolia. This indicated that there might have large SO_2 emissions in these regions. The local contribution to Beijing was more significant at the surface layer. The monthly mean concentration from local sources was about 5.53 ppbv at the surface layer, accounting for more than 65 % of the total. In comparison, the contribution from local sources only accounted for 14 % of the total at the 1.1 km layer. However, the surrounding areas which are closer to Beijing (especially TJ, BD, LF, ZJK and CD) also showed considerable contributions to the surface layer, with the total monthly contribution about 28 %.

At the 1.1 km layer, the most significant contribution to the monthly mean SO_2 concentration in Beijing was from the southern regions (about 50 % of total contribution), of which TJ accounted for 19 %, SD for 11 %, and XH for 10 % respectively. The total contribution from surrounding areas at this layer was estimated to be about 86 %. Consistent with the previous studies (An et al., 2007; Streets et al., 2007), the results in this study further confirm the long range transport of sulfur dioxide to Beijing, and the significant SO_2 emissions in those regions relatively far from Beijing. Our results also provided an estimation of the contributions from the surrounding areas to the different vertical layers, with a contribution of 86 % at the 1.1 km layer and 30 % at the surface layer.

These findings suggested the important roles that the SO_2 emissions in surrounding areas played in affecting the air pollution in Beijing. These regions are all industrialized, urbanized and coal burning areas within a few hundred kilometers of Beijing. Therefore, SO_2 directly emitted from power plant, industrial facilities, breweries etc., can be easily transported to Beijing under the favorable meteorological conditions.

Although Beijing local emissions were found to be dominant contributions at the surface layer, it is also important to know how the surrounding areas affect the air quality at the surface layer in Beijing. Sectoral analysis at the surface layer was then performed to gain insights into the direct impact of the surrounding regions on SO_2 pollution over certain counties in Beijing. Beijing administrative subdivisions were used (Fig. 7) to evaluate this impact. Consistent with the results above, the Beijing local contribution was dominant except in Pinggu County. However, TJ and ZJK were distinct from the surrounding areas with a significant direct contribution of SO_2 to its closer counties in Beijing, in particular, 86 % and 68 % of the surrounding contribution on average at Pinggu County and Yanqing County respectively (Table 6). The contribution from TJ SO_2 emissions to Pinggu County in Beijing was much higher than that from BJ local emissions. Overall, without the Beijing local contribution, the impact of surrounding regions (e.g., TJ, CD, ZJK, BD and LF) on the SO_2 pollution in Beijing are mainly via Pinggu, Miyun, Yanqing, Fangshan and Tongzhou counties respectively (Fig. 7, Table 6).

Basically, such characteristic of SO_2 contribution is closely associated with Beijing geographical position, highlighting the performance of the air pollutant tagged method

Table 6. The most important surrounding contribution to air pollution over Counties in Beijing at the surface layer. The symbol "TJ/27" means that the first important surrounding contribution to SO_2 over Huairou in Beijing is from TJ with 27 % of the surrounding SO_2 concentration contribution, and the others are the similar.

County	SO_2 (%)		NO_2 (%)		PM_{10} (%)	
Huairou	TJ/27	CD/24	ZJK/49	CD/32	CD/41	ZJK/41
Miyun	TJ/56	CD/22	TJ/49	CD/36	CD/40	LF/30
Yanqing	ZJK/68	–	ZJK/96	–	ZJK/91	–
Changping	ZJK/46	BD/20	ZJK/77	–	ZJK/84	–
Pinggu	TJ/86	–	TJ/84	LF/10	TJ/63	LF/25
Shunyi	TJ/58	LF/19	TJ/52	LF/41	LF/68	TJ/22
Mentougou	BD/48	ZJK/32	ZJK/58	BD/40	ZJK/62	BD/38
Urban Beijing	BD/35	TJ/27	LF/41	TJ/28	LF/44	BD/31
Tongzhou	TJ/48	LF/32	LF/49	TJ/48	LF/71	TJ/25
Fangshan	BD/76	ZJK/10	BD/88	–	BD/80	ZJK/16
Daxing	LF/41	BD/30	LF/68	TJ/18	LF/76	BD/16

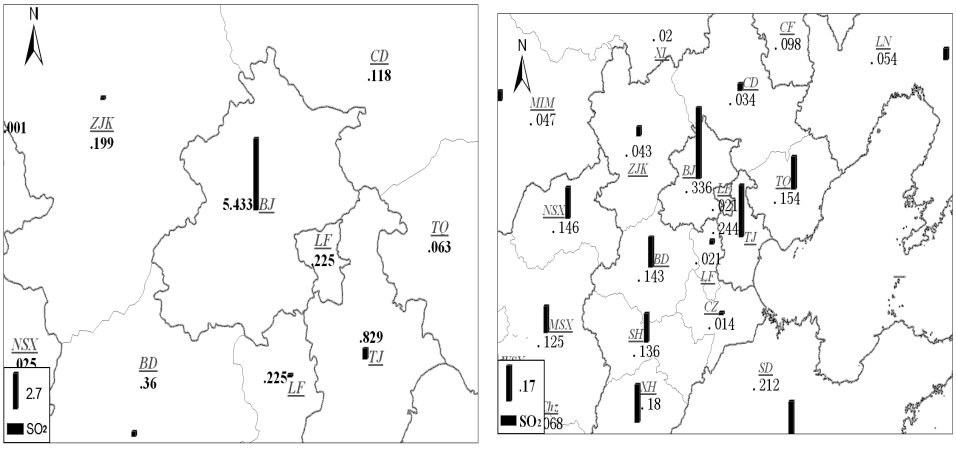

Fig. 6. The contribution to the SO_2 monthly mean concentration (ppbv) in Beijing at surface layer (left), and at the upper layer about 1.1 km height (right). The black bars and numbers in each county indicate the contribution of SO_2 in Beijing that comes from that county.

in estimating the contributions to air pollution over Beijing. The results specify the principal enter-ways of the typical influence of the key surrounding contributors on SO_2 pollution over Beijing. Undoubtedly, this results would be helpful for the further control strategy of the SO_2 pollution, especially for the County-by-County SO_2 emissions control program over Beijing.

3.2 The contribution of the PM_{10} concentration to Beijing

The PM_{10} from primary emissions were tagged and its contribution to the PM_{10} the PM_{10} pollution in Beijing were also calculated in this study. The contributions of the secondary particle matter (often refers to secondary aerosol) to the PM_{10} contribution, however, were not included in this study.

Similar to SO_2, the Beijing local contribution to the PM_{10} concentration at the surface layer (23.5 µg m^{-3}) was much higher than that at the 1.1 km layer (1.0 µg m^{-3}) (Fig. 8). The Beijing local sources contributed more than 75 % PM_{10} concentration in Beijing at the surface layer, while only 13 % at the 1.1 km layer. That indicated that the surrounding areas contributed 87 % PM_{10} concentration in Beijing at the 1.1 km layer, with the most contribution from Hebei province (43 %). Thus, the regional transport played an important role in controlling the PM_{10} pollution and visibility in Beijing at the 1.1 km layer. In addition, the regional PM_{10} contribution from the southern surrounding areas appeared to be higher than those from the northern regions, which is similar to that of SO_2, but with relatively lower contribution.

The daily variations of the contribution percentage from different regions to PM_{10} in Beijing are presented in Fig. 9. Similar to SO_2 in Fig. 5, the PM_{10} pollution in Beijing was

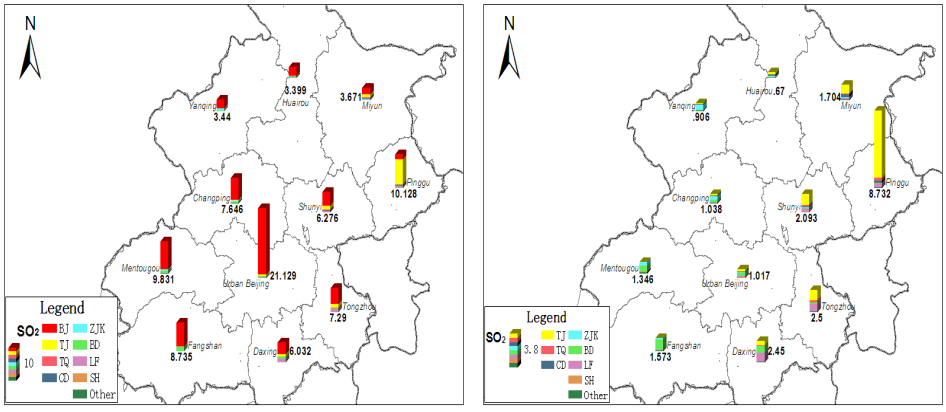

Fig. 7. The contribution to the SO_2 monthly mean concentration (ppbv) over Beijing counties at surface layer, including Beijing local contribution (left) and without Beijing contribution (right).

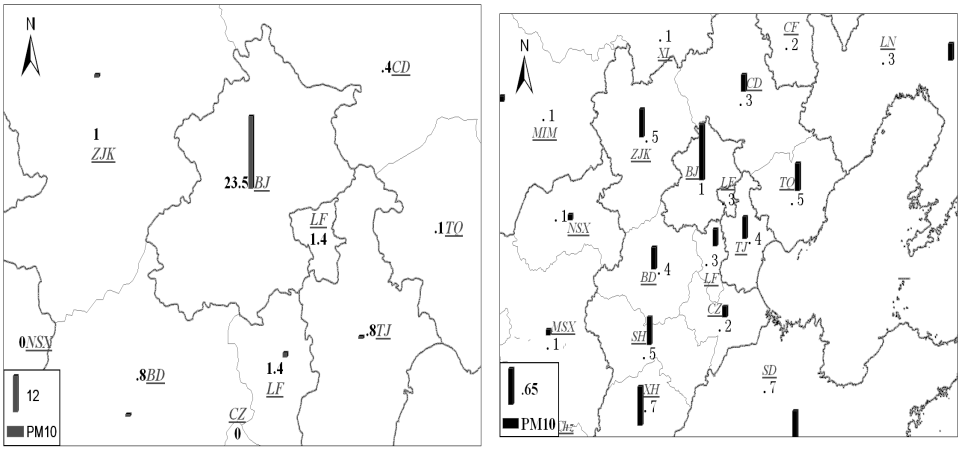

Fig. 8. Same as Fig. 6, but for primary PM_{10}. The PM_{10} concentration unit is $\mu g\,m^{-3}$.

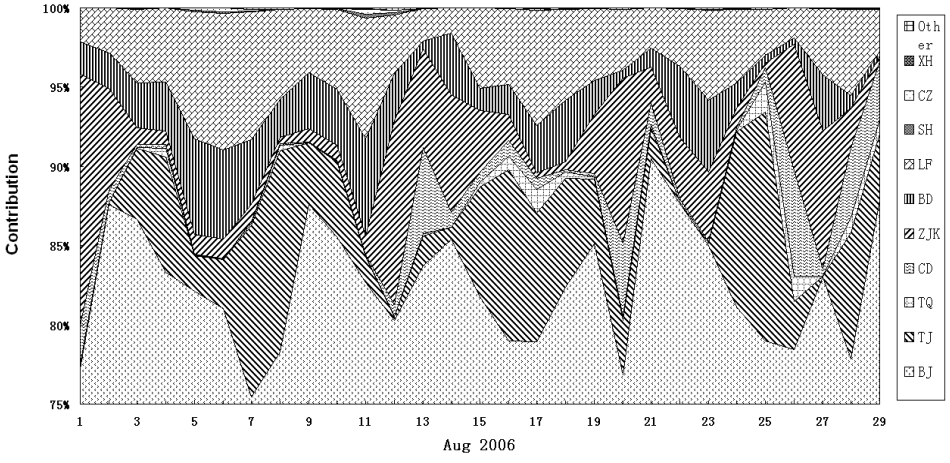

Fig. 9. Percentage of the contributions to the daily mean primary PM_{10} concentration in Beijing at the surface layer.

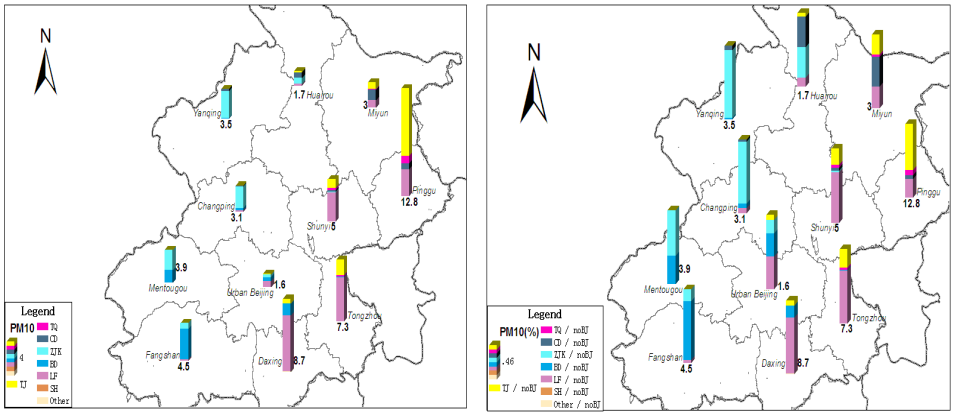

Fig. 10. Surrounding areas contribution to primary PM_{10} monthly mean concentration ($\mu g\,m^{-3}$) over Beijing counties without Beijing local contribution (left) with its percentage (right).

dominated by the contributions from BD, LF, TJ and ZJK on 7, 12, 17, 20, 23 and 25 August respectively. This suggested a typical flux in regional air pollutant transport, including SO_2 and PM_{10}. The finding was also consistent with the typical heavy pollution sequences described in Sect. 2.5.2 (model evaluation). Such convergence reflected the air pollutant tagged method capability to accurately reproduce the air pollutants transport. On the other hand, the lower contribution of the long-range transport from Shandong, Inner-Mongolia and Shanxi Provinces to the PM_{10} in comparison to that of SO_2 was likely due to the fact that the velocity of the transport of trace gases is higher than that of particulate matter which has larger dry deposition velocity (An et al., 2007).

Similar to SO_2 pollution in Beijing, county scale analysis of the PM_{10} contribution without the local contribution is presented in Fig. 10 and the results are summarized in Table 6. Again, our results showed the significant PM_{10} contributions from TJ to Pinggu County (63 % of 12.8 $\mu g\,m^{-3}$), from LF to Daxing (76 % of 8.7 $\mu g\,m^{-3}$), Tongzhou (71 % of 7.3 $\mu g\,m^{-3}$) and Shunyi (68 % of 5.0 $\mu g\,m^{-3}$) counties, and from BD to Fangshan County (80 % of 4.5 $\mu g\,m^{-3}$). The surrounding areas contributed more to the PM_{10} pollution over the eastern counties (7.3 $\mu g\,m^{-3}$~12.8 $\mu g\,m^{-3}$) of Beijing than the western (3.1 $\mu g\,m^{-3}$ ~3.9 $\mu g\,m^{-3}$) counties, which might be associated with Beijing's semi-basin topography and the weather patterns of the summer monsoon. And the most regional contribution to PM_{10} in Beijing at the surface layer is from LF (1.4 $\mu g\,m^{-3}$, Fig. 8), accounting for more than 50 % of the surrounding contributed PM_{10} concentration over Daxing, Tongzhou and Shunyi counties as mentioned above. The most important contributor to urban Beijing at the surface layers was the pair LF and BD with 72 % of the surrounding contributed PM_{10} concentration.

Our results are also consistent with Streets et al. (2007) analysis on $PM_{2.5}$, but more detailed, despite the different methods, simulations periods and the meteorological conditions between these two studies. Given that PM_{10} is often the primary pollutant in Beijing (Hao et al., 2008; An et al., 2007), the sectoral analysis in this study brought out the substantial information for relevant environmental implications by specifying areas of direct influence of each potential contributor. Our results support the strategies that controlling the PM_{10} pollution in Beijing should be implemented at the county scale.

3.3 The contribution of the NO_2 and ozone (O_3) concentration to Beijing

Nitrogen oxides ($NO_x = NO + NO_2$) play a key role in tropospheric chemistry, e.g., the production of ozone in the troposphere (Seinfeld and Pandis, 2006; Zhang et al., 2009).

The surrounding area's contribution on NO_2 pollution over Beijing showed overall similar patterns with those on PM_{10} and SO_2 pollution. The percentage of the surrounding area's contribution on NO_2 pollution at the 1.1 km layer were much higher than at the surface layer, essentially from the southern regions (Fig. 11). The contribution from the regional transport accounted for 10 % of NO_2 at the surface layer, while up to 40 % at the 1.1 km layer, which are all lower than those of SO_2 and PM_{10}. Yet, TJ, BD and ZJK were still the most significant surrounding contributors, accounting for 11 %, 9 % and 8 % respectively to the NO_2 in Beijing at the 1.1 km layer.

The county scale analysis of the NO_2 contribution is presented in Fig. 12. The local contribution was dominant in all counties of Beijing, including Pinggu County, which is slightly different from the SO_2. The urban Beijing showed the highest NO_2 concentration of 36.13 ppbv (36.53 ppbv–0.40 ppbv) due to the contribution from local NO_x emissions. Without considering the contribution from Beijing local sources, TJ was the most important regional

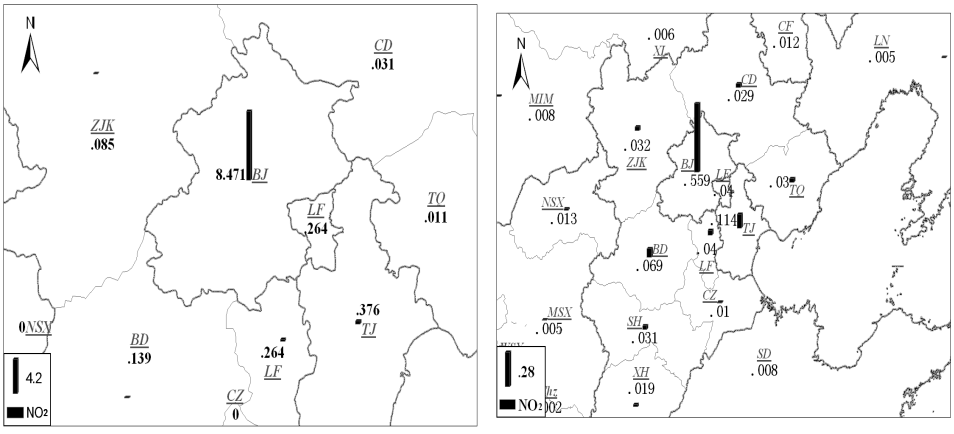

Fig. 11. Same as Fig. 6, but for NO_2.

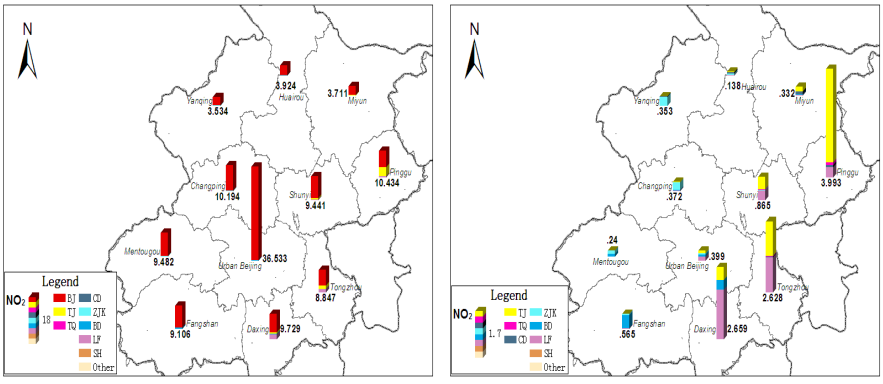

Fig. 12. Same as Fig. 7 but for NO_2, concentration unit: ppbv.

contributor to Pinggu County with an average contribution of 84 % (monthly mean 3.99 ppbv); comparatively, LF was the primary contributor to Daxing County accounting for 68 % of 2.66 ppbv. TJ and LF together contributed 97 % of the surrounding NO_2 concentration (2.63 ppbv) to Tongzhou County (Fig. 12 and Table 6). Similar to PM_{10}, the surrounding areas showed more impact on NO_2 pollution in the eastern counties, especially in Pinggu, Daxing and Tongzhou counties with the contribution range of 2.63 ppbv∼3.99 ppbv.

Therefore, the NO_2 pollution over Beijing was primarily from local NO_x emission, consistent with the results reported previously (Streets et al., 2007; Zhang et al., 2009). However the environmental strategy in the eastern counties should also take efforts to control the NO_x emission in the regions close to Beijing.

The ozone was also tagged after being produced by the photochemical reaction, and the results are shown in Fig. 13. It appeared that the contributions of ozone from both Beijing local sources and regional transport were strongly associated with the meteorological conditions and compatible

Table 7. Percentage of Beijing local production and surrounding areas contribution to daily ozone concentration in urban Beijing at the surface layer.

Region	Mean (%)	Maximum (%)	Number of days(>5 %)
BJ	53	75	30
BD	19	44	22
LF	4	12	17
ZJK	4	16	8

with the present analysis. The mean monthly and maximum daily contribution to Beijing is presented in Table 7. The local production of ozone concentration to urban Beijing was about 53 %, with maximum daily percentage of 75 % at the surface layer. Similar to other species, the ozone pollution at surface layer is dominated by the local production. However, the contribution from surrounding areas at the surface layer was estimated to be about 47 %, indicating a considerable contribution from the regional transport, especially from the

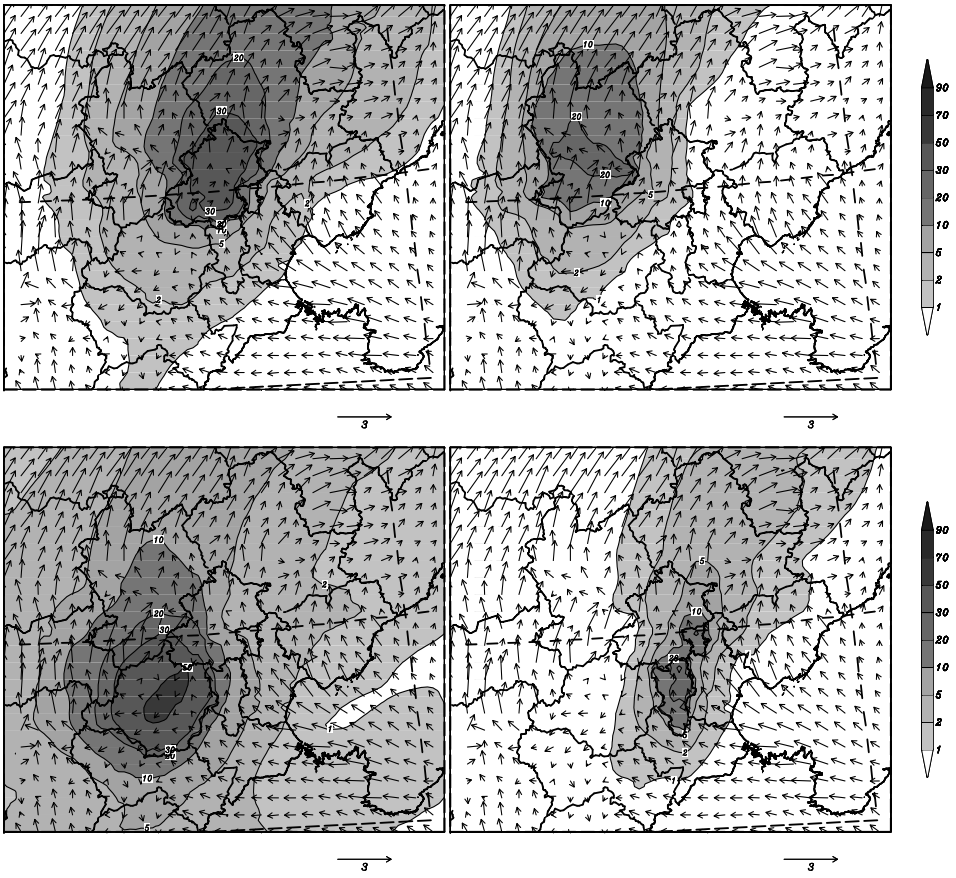

Fig. 13. Horizontal distribution of the percentage of Beijing local production and potential surrounding areas contributions to ozone monthly mean concentration over Beijing: upper left: from BJ; upper right: from ZJK; lower left: BD; lower right: LF. The unit of the wind vector is $m\,s^{-1}$.

southern regions. Without considering the local production, BD was the principal direct contributor to ozone pollution in urban Beijing, with a monthly average of 19 % and maximum daily percentage of 44 %. In total, 22 days with the contribution from BD exceeding 5 % were observed.

Overall, a significant amount of ozone produced in surrounding regions can be transported to Beijing, which has large influence on the pollution level of NO_x and VOCs. Our results also highlight the crucial role of transport and other meteorological conditions in photochemical pollution of ozone as previously reported by Streets et al. (2007), Hao et al. (2008) and Tang et al. (2010). The contribution of NO_2 from surrounding areas to Beijing discussed above were relatively smaller compared to that of ozone pollution. Further study on the transport of ozone precursors (NO_x and VOCs) is therefore needed to properly characterize the ozone contribution.

4 Conclusions

The NAQPMS model coupled with an on-line air pollutant tagged module has been developed to investigate the contributions of local sources and regional transport to the air pollution in Beijing during CAREBeijing-2006. An in-depth analysis through this tagged method, with 22 tagged emissions regions, was performed to quantify the influence of various emissions regions on SO_2, PM_{10}, NO_2 and ozone pollution over Beijing. The surrounding areas contribution to air pollutant in Beijing was estimated at the surface and the 1.1 km layer, in addition, the detailed information on the contribution from different surrounding regions to the air pollution over Beijing at county scale was present in this study. Results were summarized as following:

– The NAQPMS model showed highly improved model performance in simulating SO_2, PM_{10}, NO_2 and O_3 as suggested by the overall good agreements between the simulated and the observed data provided by CAREBeijing-2006. The model had better performance

at the IAP tower station in the urban Beijing than Yufa and Xinglong station, which are located at the rural areas. And the simulated SO_2 at Xinglong station in the east of Beijing was much higher than the observed, likely due to the overestimated source emissions in this region.

– Analysis indicated that the local sources were dominant contributions at the surface layer, accounting for 65 % of SO_2 concentration. On the episode day of 25 August, there was a significant contribution (about 25 %) from TJ. The local contribution at the 1.1 km layer was lower than that at the surface layer. However, the surrounding areas, especially the southern regions, played a dominant contribution to the pollution at the 1.1 km layer, for example, 50 % of the total SO_2. County scale analysis indicated that the regional transport from TJ contributed the most to SO_2 over Pinggu County, accounting for 86 % of the SO_2 from the total surrounding areas.

– Similar to SO_2, the Beijing local contribution to the PM_{10} at the surface layer (75 %) was much higher than that at the 1.1 km layer. At the 1.1 km layer, the contributions from surrounding areas were more significant. The PM_{10} contribution from the southern surrounding areas also seemed to be higher than those from the northern regions. In addition, the contributions from surrounding areas showed more impacts on PM_{10} in eastern counties of Beijing than the western counties. The most important surrounding contributor to Beijing at the surface layer was LF, which contributed more than 50 % of the surrounding PM_{10} contribution over Daxing, Tongzhou and Shunyi counties.

– The local contribution to the NO_2 concentration at the surface layer was dominant in all counties of Beijing, including Pinggu County, while the surrounding areas contributed 10 % NO_2 concentration versus 40 % at the 1.1 km layer, much lower than those of SO_2 and PM_{10}. The surrounding areas also contributed more to the NO_2 pollution in the eastern counties, especially in Pinggu, Daxing and Tongzhou counties.

– The local production of ozone contributed about 53 % of total ozone at urban Beijing at the surface layer. The contribution from surrounding areas was also significant and estimated to be about 47 %, of which BD was the primary contributor.

In summary, the local contribution played the actual key role in controlling the air pollution over Beijing at the surface layer, while the surrounding areas contribution to air pollution in Beijing were more significant at the 1.1 km layer. Results in this study suggest that long-term efficient air quality control strategies in Beijing should be based on regional-scale collaborations, by not only abating the local emission, but also controlling emissions from the surrounding areas. However, achievement of sustainable ozone control over Beijing needs further long-term study on ozone sensitivity to precursor emissions distribution over Beijing and concerned surrounding areas.

Appendix A

The statistical parameters

Mean bias:

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

Mean absolute gross error:

$$\mathrm{MAGE} = \frac{1}{n}\sum_{i=1}^{n}|\mathrm{Sim}(i) - \mathrm{Obs}(i)|$$

Root mean squared error:

$$\mathrm{RMSE} = \sqrt{\frac{1}{n}\sum_{i=1}^{n}(\mathrm{Sim}(i) - \mathrm{Obs}(i))^2}$$

Normal mean bias:

$$\mathrm{NMB} = \frac{1}{n}\sum_{i=1}^{n}(\mathrm{Sim}(i) - \mathrm{Obs}(i))/\mathrm{Obs}(i)$$

Normal mean error:

$$\mathrm{NME} = \frac{1}{n}\sum_{i=1}^{n}|\mathrm{Sim}(i) - \mathrm{Obs}(i)|/\mathrm{Obs}(i)$$

Correlation coefficient:

$$R = \frac{\sum_{i=1}^{n}(\mathrm{Sim}(i) - \overline{\mathrm{Sim}})(\mathrm{Obs}(i) - \overline{\mathrm{Obs}})}{\sqrt{\sum_{i=1}^{n}(\mathrm{Sim}(i) - \overline{\mathrm{Sim}})^2 \sum_{i=1}^{n}(\mathrm{Obs}(i) - \overline{\mathrm{Obs}})^2}}$$

Fractional Bias:

$$\mathrm{FB} = \frac{1}{n}\sum_{i=1}^{n}\frac{\mathrm{Sim}(i) - \mathrm{Obs}(i)}{0.5 \times (\mathrm{Sim}(i) + \mathrm{Obs}(i))}$$

Supplementary material related to this article is available online at: http://www.atmos-chem-phys.net/11/5997/2011/acp-11-5997-2011-supplement.zip.

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