

**Variations of surface  
ozone and control  
strategy for Northern  
China**

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# Spatial-temporal variations of surface ozone and ozone control strategy for Northern China

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## Abstract

The Project of Atmospheric Combined Pollution Monitoring over Beijing and its Surrounding Areas, was an intensive field campaign conducted over northern China between June 2009 and September 2011 to provide an in-depth understanding and a comprehensive record of ozone ( $O_3$ ), respirable suspended particulate ( $PM_{10}$ ), fine particle ( $PM_{2.5}$ ), nitrogen oxides ( $NO_x$ ), volatile organic compounds ( $VOC_s$ ) and other air pollutants in this quickly developing region of China. In this campaign, 25 stations in an air-quality monitoring network provided regional-scale spatial coverage. In this study, we analyzed the data on  $O_3$  and  $NO_x$  levels obtained at the 22 sites over northern China between 1 September 2009 and 31 August 2010. Our goal was to investigate the  $O_3$  spatial-temporal variations and control strategy in this area. Significant diurnal, and seasonal variations were noted, with the highest concentrations typically found at around 03:00 p.m. (LT) and in June. The lowest concentrations were generally found during early morning hours (around 06:00 a.m.) and in December. Compared with July and August, June has increased photochemical production due to decreasing cloudiness coupled with reduced  $O_3$  loss due to less dry deposition, inducing an  $O_3$  peak appearing in June. The averaged  $O_3$  concentrations were lower in the plains area compared with the mountainous area due to the titration effects of high  $NO_x$  emissions in urban areas. When the characteristics of  $O_3$  pollution in different regions were distinguished by factor analysis, we found high levels of  $O_3$  that exceeded China's National Standard throughout the plains areas, especially over Beijing and the surrounding areas. An integrated analysis with emissions data, meteorological data, and topography over northern China found that the meteorological results were the main factors that dominated the spatial variations of  $O_3$ , with the presence of abundant emissions of precursors in this area. The smog production algorithm and space-based HCHO/ $NO_2$  column ratio were used to show the  $O_3$ - $NO_x$ - $VOC_s$  sensitivity and examine the control strategy of  $O_3$  over northern China. The results show that summer  $O_3$  productions in the plains and northern mountainous areas were sensitive to  $VOC_s$  and

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NO<sub>x</sub>, respectively. Our results are helpful for redefining government strategies to control the photochemical formation of air pollutants over northern China and are relevant for developing urban agglomerations worldwide.

## 1 Introduction

Increases in tropospheric ozone are a serious environmental concern because of their adverse impacts on human health and vegetation and their resultant greenhouse effect. Photochemical ozone is formed from NO<sub>x</sub> and VOC<sub>s</sub> through non-linear interactions between chemical reactions and meteorology. The relationship between precursor pollutants and photochemical O<sub>3</sub> differs by location due to the emissions distribution and meteorology (NRC, 1991).

Elevated O<sub>3</sub> concentrations that range from local to regional scales have been extensively reported (Lee et al., 1998; Oltmans et al., 1998; Jacob et al., 1999; Lin et al., 2000; Bronnimann et al., 2002; Akimoto, 2003; Lelieveld et al., 2004; Parrish et al., 2004; Jonson et al., 2005; Zhang et al., 2008; Tang et al., 2009). For example, large widespread emission sources in East Asia (Streets et al., 2003; Zhang et al., 2009), especially in China (Li et al., 2010; Huang et al., 2011), have resulted in high concentrations of O<sub>3</sub> precursors, namely NO<sub>x</sub>, VOC<sub>s</sub> and CO. These precursors have, in turn, led to high levels of oxidants (Zhang et al., 1998; Xu et al., 2008; Zhang et al., 2008; Shao et al., 2009; Tang et al., 2009; Wang et al., 2009; Tang et al., 2010a, b; Wang et al., 2010; Xu et al., 2010, 2011; Wang et al., 2011). Therefore, the increase in Asian continental outflows was suggested to account for an increase in O<sub>3</sub> in the northern Pacific Ocean region (Parrish et al., 2004). Jacob et al. (1999) suggested that the tripling of Asian anthropogenic emissions between 1985 and 2010 would lead to an increase in the monthly mean O<sub>3</sub> concentration by 2–6 ppbv in the western US and by 1–3 ppbv in the eastern US. This type of intercontinental O<sub>3</sub> transport has strong effects on climate. These studies emphasize that researches on O<sub>3</sub> pollution in China will provide benefits for the mitigation of global air pollution.

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Xu et al., 2005; Cheng et al., 2007; Chan and Yao, 2008; Xin et al., 2010). Worst of all, in stagnant weather for summer over the BTH region, atmospheric pollutants from various emissions easily accumulate and lead serious pollution accidents, exhibiting decreased visibility, high concentration of particulate matters and ozone in the region (Ying et al., 1999; Song et al., 2002; Ding et al., 2005). Since 2004, Beijing local government realizes the importance of the regional collaborative prevention and control for air pollutants. In other words, three governments should cooperate to decrease the emissions from various sources in order to abate the air pollution and improve the air quality over the BTH region. Therefore, cooperated with other governments and institutes, Beijing local government has implemented 2 projects: study of the Transmission and Transformation of Beijing and Surrounding Area Air Pollution and the Object of the Beijing Air Quality (2005–2007) and Air quality safeguards Research of Beijing during the 29th Olympic games (2006–2008) for monitoring, evaluating and predicting the air quality of Beijing and its surrounding areas for the 29th Olympic Games. Due to the continuous efforts of three local governments, the emission restrictions were notably successful in improving air quality in Beijing during Olympics (Wang et al., 2009; Xin et al., 2010). However, O<sub>3</sub> pollution in the BTH region outside of Beijing has not yet been investigated adequately; large gaps and uncertainties remain in the knowledge of characteristics of regional O<sub>3</sub> pollution and its mitigation strategies in BTH. Previous investigations were fragmented and limited in scope and depth due to a lack of human and instrument resources. Moreover, to develop effective policies for O<sub>3</sub> pollution control, our understanding of O<sub>3</sub>-VOC<sub>s</sub>-NO<sub>x</sub> chemistry over the entire BTH region needs to be improved through more observational and modeling studies.

To further control air pollution in the BTH region over northern China after the Olympics, the “Project of Atmospheric Combined Pollution Monitoring over Beijing and its Surrounding Areas” was organized by the Chinese Ecosystem Research Network (CERN), Institute of Atmospheric Physics (IAP) and the Chinese Academy of Sciences (CAS) to provide an in-depth understanding and a comprehensive record of ozone, PM<sub>2.5</sub>, PM<sub>10</sub>, and other air pollutants in this quickly developing region of China.

Extensive spatial coverage was made possible by the participation of 25 air-quality network stations, which provided a clear picture of the ozone pollution on a regional scale.

In this study, we illustrated the spatial-temporal variations of surface ozone and related components over northern China between 1 September 2009 and 31 August 2011 using data from the air quality monitoring network. Furthermore, a combined approach that incorporates the emissions inventories, meteorological simulations, and topography was used to evaluate the factors that influence the summer O<sub>3</sub> concentrations over northern China. Finally, based on the smog production algorithm and space-based monthly HCHO and NO<sub>2</sub> column concentrations, the sensitivity of ozone production to its precursors was evaluated to identify strategies for controlling photochemical pollution over northern China.

## 2 Methodology

### 2.1 Air Quality Network

#### 2.1.1 Sites

The Air Quality Network over northern China, affiliated with the CAS-IAP-CERN, was expanded based on the established BTH Atmospheric Environment Monitoring Network (Xin et al., 2010) from 17 to 25 sites. The network monitored air pollutants in real-time and provided early warnings of air pollution in the region from June 2009 to September 2011. The network comprises 25 air-quality monitoring stations across this area (Fig. 1), which each measure the ambient concentrations of PM<sub>10</sub>, PM<sub>2.5</sub>, SO<sub>2</sub>, NO<sub>2</sub>, NO, CO, and O<sub>3</sub>. These sites, four in Beijing (BJT, LTH, YF, SQL), two in Tianjin (TJT, TG), seventeen in Hebei (BD, SJZ, CZ, HS, HJ, GA, ZZ, LF, YJ, TS, CFD, QA, LS, XL, QHD, CD, ZJK), and one in Shanxi (DT), are located away from specific point emission sources and were selected to be broadly representative of air pollutants levels

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for a selected site. Based on geopolitical locations, the network includes five sites in Beijing, four sites in the surrounding Beijing area (GA, ZZ, LF, XH, YJ), five sites in the southern plains (BD, SJZ, HJ, HS, CZ), six sites in the eastern coastal area (QHD, TS, CFD, QA, TJT, TG), three urban sites (ZJK, CD, DT) and two rural sites (XL and LS) in the northwestern mountainous area.

In this study, O<sub>3</sub> and NO<sub>x</sub> data from 1 September 2009 through 31 August 2010 were provided by the network. Data from all sites were first screened by inspection the frequency of missing measurements. Except for the sites in QA, XH and SQL, which were temporally interrupted during some periods due to network maintenance and upgrading, all other stations have more than 90 % data capture rates for O<sub>3</sub> and NO<sub>x</sub> during the one year of monitoring. Therefore, the remaining 22 sites were used to illustrate the spatial-temporal variations over northern China in this work.

### 2.1.2 Measurement techniques

The measurement instruments were housed in a laboratory situated at each site. Ambient air samples were drawn through a 3-m PFA Teflon tube (outside diameter: 12.7 mm; inside diameter: 9.6 mm). The sampling tube inlet was located 1 m above the laboratory, and the outlet was connected to a PFA-made manifold with a bypass pump that drew air at a rate of 15 l min<sup>-1</sup>.

Surface ozone concentrations were measured using a Model 49 or 49 C ozone analyzer from Thermo Environmental Instruments (TEI), Inc. NO<sub>x</sub> levels were measured using the TEI Model 42 C and 42 CTL NO and NO<sub>2</sub> analyzers. The TEI Model 49 detector was found to exhibit a detection limit of 2 ppbv and a precision of 2 ppbv, while Model 49 C had a detection limit of 1 ppbv and a precision of 1 ppbv. Both of the NO<sub>x</sub> analyzers had a precision of 0.4 ppbv, with the limit of detection for Model 42 C and 42 CTL being 0.4 ppbv and 0.05 ppbv, respectively.

Data quality was evaluated and certified by the China National Accreditation Board of Laboratories (CNAL) and was consistent with international requirements. IAP personnel strictly adhered to national environmental monitoring standards. Quality control

checks, including automatic zero-calibration and span checks of gas analyzers, were performed daily, and manual calibrations with standard gases were conducted weekly. The NO<sub>x</sub> analyzers were zero-checked and span-checked using a zero gas generator (TEI Model 111) and an internal O<sub>3</sub> source of a multi-gas calibrator (TEI Model 146 C) with a NO standard (National Centre for Standard Materials, Beijing, China). Multi-point calibrations of the O<sub>3</sub> analyzer were used as an O<sub>3</sub> calibrator (TEI Model 49 CPS). The sampling methods and instrument protocols as well as quality assurance/quality control (QA/QC) procedures for air quality monitoring are described in detail in the Chinese National Environmental Protection Standard, Automated Methods for Ambient Air Quality Monitoring (HJ/T 193-2005; State Environmental Protection Administration of China, 2006). The measurement techniques are the same as in Tang et al. (2009). The description is repeated here for the reader's convenience.

## 2.2 Data analysis

Several different statistics were utilized to investigate the spatial and temporal characteristics of the measured O<sub>3</sub> and NO<sub>x</sub> over northern China. First, we calculated the maximum, minimum, mean and standard deviations for each site to give us a basic knowledge of weather conditions, O<sub>3</sub> and NO<sub>x</sub> variations over northern China. Second, to examine shared variance between different sites and identify homogenous ozone clusters, factor analysis was conducted on the daily maximum O<sub>3</sub> as an initial data reduction tool at all 22 sites using the method in Zheng et al. (2010) and Sarnat et al. (2010). Third, Pearson paired correlation coefficients (*R*) were calculated as a measure of the linear relationship to illustrate the relative temporal differences between sites. Because correlations alone may not sufficiently depict absolute concentration differences between sites (Pinto et al., 2004), the spatial coefficients of variation (CV) and the coefficients of divergence (COD) of the measured O<sub>3</sub> and NO<sub>x</sub> concentrations have been used as supplementary methods to the analysis of correlation, to characterize the spatial variations of O<sub>3</sub> and NO<sub>x</sub> in several multi-site comparative analyses (Wongphatarakul et al., 1998; Pinto et al., 2004; Kim et al., 2005; Cyrus et al., 2008;

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Krudysz et al., 2008; Sarnat et al., 2010). The CV was averaged over the sampling year and defined as the mean of the standard deviation of the spatial distribution divided by the distribution's mean. The COD was characterized by means of coefficients of divergence (Wongphatarakul et al., 1998) which are defined as

$$5 \text{ COD}_{fh} = \sqrt{\frac{1}{n} \sum_{i=1}^n \left( \frac{X_{if} - X_{ih}}{X_{if} + X_{ih}} \right)^2} \quad (1)$$

where  $X_{if}$  is the  $i$ th concentration observed at the  $f$ th site,  $f$  and  $h$  are two monitoring sites, and  $n$  is the sample size of observations. The COD of zero indicates the  $O_3$  and  $NO_x$  measured at two sites are uniform and a value approaching one means absolute dissimilar or heterogeneity. We use the COD of 0.20 as the threshold to identify the heterogeneity or homogeneity between two sites, which is detailed described in Krudysz et al. (2008). In this context, two sites exhibit strong uniformity with each other in  $O_3$  and  $NO_x$  concentrations, showing similar absolute levels, high correlation coefficients, low CV and COD values.

The summary statistics for each sites, including maximum, minimum, mean and standard deviations, were calculated using Microsoft Excel 2003. Factor analysis of the daily maximum  $O_3$  was performed using SPSS 17.0, with varimax rotation, the maximum likelihood extraction method and eigenvalues more than 1.0. The COD of measured  $O_3$  and  $NO_x$  were conducted using the statistical software package SAS 9.1 for Windows.

### 2.3 Model description and experiment design

The Weather Research and Forecasting (WRF, version 3.3) modeling system (Skamarock et al., 2008) was utilized to simulate the meteorological parameters to illustrate the reasons of the  $O_3$  spatial-temporal variations over northern China. The WRF was run with 28 vertical layers and with the following physics options: the Grell-3D cumulus scheme, the Lin microphysics, unified Noah land-surface model, the

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Mellor-Yamada-janjic TKE scheme, a rapid radiative transfer model (RRTM) longwave scheme and Goddard shortwave scheme. The National Centers for Environmental Protection (NCEP)  $1 \times 1$  global reanalysis data and the upper air observation data were used to correct the initial and boundary conditions for the WRF simulations (Fig. 2). The four-dimensional data assimilation (FDDA) technique (“analysis nudging”) was used to nudge the temperature, humidity and 3-D winds every 6 h. Hourly meteorological data from WRF was used to calculate the dry deposition of  $O_3$  using version 3.3 of the Meteorology-Chemistry Interface Processor (MCIP) (Byun and Ching, 1999).

The WRF was configured to have three domains (Fig. 2). The mother domain with a horizontal grid resolution of 81-km, covered the entire area of China; the 27-km grid-spacing inner domain covered Eastern China; and the third domain with 9-km resolution covered Beijing, Tianjin, and Hebei Province. All of the grids had 28 vertical layers extending from the surface to about 15 km above the ground. The vertical layers were unevenly distributed with eight layers in the lowest kilometer, and a surface layer of approximately 38 m. Our WRF simulations are conducted every three and half days from 1 September 2009 to 31 August 2010. To minimize the effect of the initial conditions, a half-day spin-up was used for each simulation.

Predicted meteorological parameters, including temperature, relative humidity, winds and precipitation, were validated against the monthly observations obtained between September 2009 and August 2010 for 10 sites (Fig. 1) over northern China. The meteorological data from 7 of 10 sites (BJ, BT, CD, DT, LT, SJZ and TJ) were available on the China Meteorological Data Sharing Services System website (<http://cdc.cma.gov.cn/>), while data from the other three sites (BJF, LC, YC) were obtained from CERN (Hu et al., 2007a, b). The performance statistics for the WRF calculations were conducted using the Metstat statistical analysis package (Emery et al., 2001).

## 2.4 O<sub>3</sub>-NO<sub>x</sub>-VOC<sub>s</sub> sensitivity diagnosis

### 2.4.1 The smog production algorithm

The smog production (SP) algorithm was used to examine areas of VOC<sub>s</sub> and NO<sub>x</sub> sensitivity, which qualitatively represented the production of O<sub>3</sub> in smog chambers (Blanchard et al., 1999; Blanchard, 2000), in box model simulations (Blanchard et al., 1999), in air quality model simulations (Blanchard and Stoeckenius, 2001) and in comparison with field measurements (Blanchard and Fairley, 2001). Johnson (1984) and Johnson and Quigley (1989) defined the variable SP as

$$SP(t) = O_3(t) - O_3(0) + NO(0) - NO(t) \quad (2)$$

All species are represented in units of volume mixing ratio. SP represents the total amount of NO consumed and O<sub>3</sub> produced and is fundamentally associated with the chemical reactions that enable O<sub>3</sub> to accumulate (Blanchard and Fairley, 2001).

The extent of reaction ( $E(t) = SP/SP_{\max}$ ) was defined as

$$E(t) = \frac{SP(t)}{SP_{\max}} = \frac{O_3(t) + DO_3(t) - O_3(0) + NO(i) - NO(t)}{\beta[NO_x(i)]^\alpha} \quad (3)$$

This equation required values for the unmeasured quantities DO<sub>3</sub>(*t*), O<sub>3</sub>(0), and NO<sub>x</sub>(*i*), which must be estimated from measurements or simulations. We use fixed values of 40 ppbv for O<sub>3</sub>(0), 2/3 for  $\alpha$  and 19 for  $\beta$  according to Blanchard et al. (1999). DO<sub>3</sub>(*t*), which can also be used to estimate NO<sub>x</sub>(*i*), was estimated using O<sub>3</sub>(*t*), dry deposition velocity ( $V_d(t)$ ) and planetary boundary layer height ( $Z(t)$ ) according to equations specified by Blanchard et al. (1999). Both  $V_d(t)$  and  $Z(t)$  parameters can be estimated by WRF simulations.

The  $E(t)$  represented what extent a system had proceeded towards its maximum possible O<sub>3</sub> formation (Johnson, 1984; Blanchard and Fairley, 2001), with the transition

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than 25% (Martin et al., 2004; Duncan et al., 2010). For this work, we used a HCHO/NO<sub>2</sub> ratio < 1 indicate VOC<sub>s</sub>-sensitive conditions, while HCHO/NO<sub>2</sub> ratio > 2 indicate NO<sub>x</sub>-sensitive conditions. HCHO/NO<sub>2</sub> ratio in the range of 1–2 should generally be considered a transition regime (Martin et al., 2004; Duncan et al., 2010; Witte et al., 2011).

### 3 Results

#### 3.1 Model evaluation and meteorology over northern China

In this section, the model results are examined to extend the understanding of the mesoscale structure between September 2009 and August 2010 beyond what can be seen in the more sparse set of observations. Before the specific structures of the numerical solutions are investigated, it is helpful to review briefly the results for the whole year for which the experiment was conducted. Table 1 presents the statistics between monthly observations and simulations of ten weather stations for surface-layer temperature, relative humidity, wind speed and precipitation using several common measures of skill (Stauffer and Seaman, 1990). The table shows mean errors (ME), mean absolute errors (MBE), root mean square errors (RMSE) and correlation coefficients (R) for the former four parameters. The table shows very low bias in the surface temperatures and relative humidity, with typical errors (MBE) of only 1.01 °C and 7.30 %, respectively. However, the surface wind speeds are always overestimated by WRF, which shows high biases of –1.91 % and 1.91 % for MB and MBE, respectively. This overestimation is mostly because the winds in the surface layer tend to be influenced by buildings, small-scale (unresolvable) terrain, and other obstacles or heterogeneity, which is a typical characteristic in a mesoscale meteorological model. Table 1 also shows a small MB for the monthly average precipitation, a modest RMSE and a high correlation of 0.76. All of the comparisons indicated that the WRF performance was well within the typical range of meteorological modeling studies (Hanna and Yang, 2001; Emery et al., 2001).

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Beijing and its surrounding area are in a warm temperate zone and have a typical continental monsoon climate with four distinct seasons. Figure 3 shows the monthly average simulated and observed statistics of ambient temperature, relative humidity, wind speed and precipitation for 10 weather stations. On average, high temperature and relative humidity appeared in summer, and low temperature and relative humidity appeared in winter and spring, respectively. The hottest and the coldest months were found to be July and January, averaging 26.8 °C and -6.5 °C, respectively, while the most humid and the driest months were in August and April, with 73.0 % and 49.9 %, respectively. In other words, even in the winter months, the average RH was over 49 %. The wind speed was much higher in the spring than that in other seasons, with the highest monthly average appearing in April. The lowest monthly average precipitation was 0.44 mm in December, and the highest was 129.2 mm in August, which were due to a Siberian high and the East Asia summer monsoon circulation, respectively. Figure 4 shows the simulated vector winds and planetary boundary layer heights (PBLH) for four seasons between 09:00 and 15:00. The time period is consistent with strong photochemical production of O<sub>3</sub> over the course of the day. During all seasons, the wind speeds were low in North China Plain and high in mountainous areas. In the winter, the northerly and northwesterly winds were much more frequent than that in other seasons. However, the plains and mountainous areas were dominated by southeasterly and southwesterly winds in summer, respectively. In addition, PBLH was highest in the spring and lowest in winter. In the autumn and spring, PBLH of the plains and mountainous areas had good coincidences. However, PBLH in the plains and mountainous areas showed great differences in the winter and summer, whereas PBLH was higher in the winter in the plains areas and in the summer in the mountainous areas, respectively. In summary, the spring showed a high PBLH and westerly, dry and strong winds; summer had high temperature, high humidity and southerly weak winds; winter had low temperature, low PBLH and northwesterly winds; and autumn was similar to spring in temperature, but had characteristically low PBLH, high humidity and northerly weak winds.

## 3.2 Spatial variations of O<sub>3</sub> and NO<sub>x</sub>

Spatial distributions of surface NO<sub>x</sub> and O<sub>3</sub> for the annual mean are presented in Fig. 5 and Fig. 6, respectively. Similar patterns were found for different seasons but are not shown here. The annual mean NO<sub>x</sub> mixing ratios were relatively high over the plains areas, but low over the mountainous areas. The lowest NO<sub>x</sub> concentrations were found in two rural sites over the mountainous areas at LS (8.2 ppbv) and XL (9.1 ppbv), as expected. The average of the other three mountainous sites was 28.5 ppbv, which was also much lower than the average concentration over the plains areas (39.8 ppbv). The highest annual mean NO<sub>x</sub> concentration (55.4 ppbv) appeared in BJT. In TG, the average annual mean NO<sub>x</sub> concentration was 49.7 ppbv, second only to that in BJT. This spatial distribution of NO<sub>x</sub> over northern China did not match well with NO<sub>x</sub> emissions, especially over the red-square areas (Fig. 7a). The total NO<sub>x</sub> emissions (as NO<sub>2</sub>) in Beijing, Tianjin and Hebei are 2.0 Tg in 2006 and accounted for 42.0 % from power plants, 23.1 % from automobiles and 29.0 % from industry (Zhang et al., 2009). Considering the higher stacks of power plants and industry, Hao et al. (2000) indicated that automobile sources contributed 67 % to the NO<sub>x</sub> concentration, which implies that vehicle emissions are the most important air pollutant source in Beijing. This phenomenon was also the main reason for the mismatch between emissions and ambient concentrations.

In contrast to NO<sub>x</sub>, the annual mean O<sub>3</sub> concentrations are relatively high over the northwestern mountainous areas but low over the plains areas. The annual mean O<sub>3</sub> mixing ratio at DT, LS, XL and ZJK were 25.8, 31.8, 49.3 and 28.37 ppbv, respectively, with an average of 33.6 ppbv. However, the average annual O<sub>3</sub> mixing ratio for sites over the North China Plain were lower than 25.0 ppbv, with an average of 21.9 ppbv. Therefore, the high NO<sub>x</sub> emissions over the plains areas, especially in the urban areas, scavenge O<sub>3</sub> from the air in the plains areas, thus decreasing the O<sub>3</sub> levels in these areas.

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### 3.3 Classification of regional sites and O<sub>3</sub> and NO<sub>x</sub> inter-site correlations

Table 2 presents the results of the factor analysis for daily maximum ozone (O<sub>3,max</sub>). Factor analysis (FA) conducted on the daily maximum ozone mixing ratio shows that 29.7% of the variability in the total daily maximum ozone mixing ratio for 7 of the 22 sites was explained by one shared factor; 24.9% of the variability for 5 of the remaining 15 sites was explained by the second shared factor; and 23.6% of the variability for 5 of the remaining 10 sites was explained by the third shared factor. The FA results showed that the ozone patterns in the BTH region could be classified into four homogenous groups. The first group included 7 sites located in Beijing and the surrounding area with more automobiles and fewer industries, marked with solid red circles in Fig. 1. The second group consisted of 5 sites located in the southern plain area of the North China Plain, denoted in Fig. 1 by solid brown circles. The third group, marked with solid blue circles in Fig. 1, is composed of 5 sites located east of Beijing and adjacent to the coastline of the Bohai Sea. The second and third groups were characterized by more industries and fewer automobiles compared with the first group, indicating inconsistent emissions over three different areas. All of the remaining 5 sites belonged to the fourth group, marked with solid black circles, located in the mountainous area with less dense population and industries. Based on these initial FA results for daily maximum O<sub>3</sub>, detailed correlation analysis on O<sub>3</sub> and NO<sub>x</sub> concentrations was conducted for each group.

Based on both the homogenous O<sub>3</sub> temporal variations and the site geographical characteristics, the relative and absolute spatial concentration differences for four categories were separately analyzed using *R*, CV and COD (Table 3). For O<sub>3,max</sub>, the inter-site correlations were the strongest. Correlations ranged between 0.94 and 0.97, with a mean inter-site correlation among all sites of 0.96. O<sub>3,max</sub> also exhibited the least variability in absolute levels, with the mean annual CV ranging from 14.5% to 19.8%, and the inter-site mean COD of each category was less than 0.20. The statistics of O<sub>3</sub> were similar to that of O<sub>3,max</sub>. However, all of the statistics were more heterogeneous than

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that of  $O_{3\_max}$ , with the mean inter-site correlations ranging between 0.82 and 0.97, the CV ranging between 17.8 % to 37.6 % and the COD ranging between 0.12 to 0.23.  $NO_x$ , the primary pollutants, showed mean inter-site correlations for each group that were typically moderate to strong ( $R$  range = 0.67–0.93; mean annual inter-site  $R$  among all sites = 0.84). As expected,  $NO_x$  also exhibited the most variability in absolute levels, with the mean annual CV ranging between 22.1 % and 58.0 % and the inter-site mean COD ranging between 0.17 and 0.36. From the above analysis, two conclusions were made. First, with the high inter-site correlations and the low variability in the absolute levels,  $O_{3\_max}$  showed great homogeneity among each group, indicating homogenous temporal and spatial variations for the  $O_{3\_max}$  of each category. Second, compared with the secondary pollutants ozone,  $NO_x$  was more heterogeneous, with the lowest correlations and the highest CVs and CODs. Considering the homogeneity of  $O_3$  and  $O_{3\_max}$ , the heterogeneity of  $NO_x$  illustrates the non-linearity between the precursors and ozone production. In other words, the homogeneity of  $O_{3\_max}$  among the sites in the different groups probably did not result from the emissions of precursors.

### 3.4 Temporal variations of $O_3$ and $NO_x$

#### 3.4.1 Seasonal variations

Figure 8 displays the monthly variations of  $NO$ ,  $NO_2$  and  $O_3$  between September 2009 and August 2010 at each site. For  $NO_x$  at all of the sites, the seasonal variations seem similar, with the highest concentrations appearing in the winter and the lowest concentrations appearing in the summer. The observed summer valleys of  $NO_x$  can be attributed to stronger vertical mixing due to higher PBLH (Fig. 4d), faster transition from  $NO_2$  to  $O_3$  due to higher temperature and higher wet deposition due to precipitation (Fig. 3). December 2009 was the most polluted month for  $NO_x$ . In this month, the observed monthly mean mixing ratios of  $NO$  and  $NO_2$  were 35.5 and 29.1 ppbv, respectively. The observed winter peaks of  $NO_x$  can be attributed to weaker vertical

mixing due to lowest PBLH (Fig. 4b), slowest chemical loss due to the lowest temperature (Fig. 3), solar radiation and oxidant concentrations (Fig. 8), and particularly, much higher anthropogenic emissions from coal-burning in the winter.

In contrast with  $\text{NO}_x$ , the secondary pollutant  $\text{O}_3$  exhibited a reverse seasonal pattern for all of the sites, with the highest mixing ratio appearing in June and the lowest mixing ratio in December. The observed valleys of  $\text{O}_3$  can be attributed to lower vertical mixing due to lower PBLH (Fig. 4b), stronger titration by  $\text{NO}_x$  due to higher emission related to heating and lower photochemical production due to lower temperature (Fig. 3) and solar radiation. Given that July had the highest temperature, higher  $\text{O}_3$  concentrations were expected in this month. Therefore, the fact that the  $\text{O}_3$  concentration in June (47.2 ppbv) was much higher than that in July (39.7 ppbv) is of great interest. Ding et al. (2008) suggested two causes of the seasonal peak of  $\text{O}_3$  in June over Beijing derived from multi-year records of the MOZAIC aircraft data: more intense crop residue burning in June contributing to emissions of  $\text{O}_3$  precursors; and prevailing southerly winds in June facilitating the long-range transport of regional emissions to Beijing. Our observations at 22 sites in summer 2010 presented two interesting cases. One was that all of the sites had the similar seasonal patterns, even on the eastern coastal areas. The other interesting case was that although  $\text{NO}_x$  levels were not significantly different during these two months (20.2 ppbv for July and 21.3 ppbv for June), the  $\text{O}_3$  levels still decreased from 47.2 ppbv for June to 39.7 ppbv for July. These results suggest that at least in 2010, this phenomenon could not be attributed to long-range transport due to prevailing southerly winds and changes in local and regional precursor emissions. Wang et al. (2008) suggested another cause of the seasonal peak of  $\text{O}_3$  in June at a rural site near Beijing: meteorological influences associated with the summer monsoonal circulation that develops over the North China Plain in July. As shown in Fig. 3, the monthly rainfall for July and August 2010 were 100.2 and 129.2 mm respectively, compared with 32.5 mm for June. As the East Asian summer monsoon circulation prevailed across the entire eastern part of China, we expected the radiative impact of the monsoonal clouds on ozone to be significant on a regional scale during the summer.

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More rainfalls was correlated with more cloud fraction (Fig. 9). The induced less short-wave radiation because of cloudiness in July then decreased the  $O_3$  photochemical production in northern China. While we focused on the advantages of meteorology for  $O_3$  photochemical production in June, the process of loss for  $O_3$  should not be ignored.

5 Figure 6 also shows the monthly variation of dry deposition velocity ( $V_d$ ) for ozone from MCIP, with the highest value appearing in August and the lowest value appearing in December. The simulated  $V_d$  for  $O_3$  in June ( $2.3 \text{ mm s}^{-1}$ ) was lower than that in July ( $2.8 \text{ mm s}^{-1}$ ) by about 22.9%. From the above analysis, we arrived at a major conclusion. A decrease in regional  $O_3$  photochemical production was coupled with an increasing  $O_3$  dry deposition in July, and such changes led to the highest  $O_3$  concentrations in June over northern China. Although some interpretations for the June  $O_3$  peak are given, detailed sources and sinks for surface  $O_3$  over northern China still need to be deeply studied using the air quality model.

### 3.4.2 Seasonal average diurnal variations

15 For each season, the average diurnal variations of  $NO_x$  and  $O_3$  mixing ratios were conducted from the hourly average values in the corresponding season. Figure 10 shows the average diurnal variations of the mixing ratios of  $NO_x$  in different seasons at the 22 sites. The  $NO_x$  mixing ratio shows a diurnal pattern with the maximum appearing in the early morning when the mixing layer is low and stable and the minimum appearing in the afternoon when the photochemical formation and vertical mixing is strong. Figure 10 shows that the mixing ratios of  $NO_x$  at night in the winter stayed at high levels, which resulted from strong temperature inversion and stable conditions in combination with larger emission from heating in the winter (Fig. 4b). Figure 11 shows the average diurnal variation of the mixing ratios for  $O_3$  in different seasons at the 22 sites.  $O_3$  increased in the early morning after sunrise, reaching a maximum around 03:00 p.m., and then  $O_3$  decreased in the evening, reaching a minimum around 06:00 a.m., which is immediately before sunrise. Typically, the ozone at low-elevation sites peak in the afternoon, resulting from photochemical formation of ozone

over the course of the day (Logan, 1989). The diurnal variations of both pollutants were much less significant in winter than those in other seasons. The highest average diurnal amplitude for ozone (51.9 ppbv) appeared in the summer, and the lowest average diurnal amplitude (11.6 ppbv) appeared in the winter. Spring and autumn showed modest amplitudes of 28.8 ppbv and 28.6 ppbv, respectively. This strong diurnal variation in the summer over northern China is a result of strong photochemical production due to high temperature and high solar radiation. Interestingly, the average diurnal variations for  $\text{NO}_x$  and the ozone concentrations at XL in the summer were different from those of other seasons and other sites. The  $\text{NO}_x$  concentration at XL showed a diurnal pattern with the maximum appearing in daytime and the minimum appearing at night. Ozone at XL increased in the morning after sunrise, reaching a maximum around 05:00 p.m. and then decreased at night, reaching a minimum around 08:00 a.m. Both the peak and valley hours were delayed by about 2 hours compared with other sites. The high mixing ratios of ozone occurred at 05:00 p.m., combined with the daytime peak in  $\text{NO}_x$  mixing ratios (Fig. 10), indicating the arrival of more polluted air, consistent with the change in mean flow pattern in the summer (Fig. 4d). In addition, the diurnal variations of ozone were much less significant at XL than those at other sites. Considering the delayed ozone peak and mean flow pattern in the summer, the weak diurnal variation of ozone indicated the main source of ozone at XL may be transport. Consistent with our speculation, previous observations at other surface sites near northerly mountainous area over northern China showed a similar diurnal cycles in summer (Wang et al., 2008; Lin et al., 2008).

## 4 Discussions

### 4.1 Exceedances of $\text{O}_3$

Compared with the Chinese Ambient Air Quality Standards (CAAQS, revised GB 3095–1996 of Chinese National Air Quality standards), hourly  $\text{O}_3$  mixing ratio exceeding

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Grade II (102 ppbv) occurred mainly between May and September. During the observation period, the average exceedances at the 22 sites of the daily maximum  $O_3$  when the  $O_3$  level exceeded Grade II were found to be 0.7 %, 0.0 %, 1.9 % and 18.2 % in the autumn, winter, spring and summer, respectively. The value of the WHO interim target-1 (IT-1) for daily maximum 8-h mean (82 ppbv), an intermediate target for populations with  $O_3$  concentrations above this level, was proposed as one of the indexes for the protection of human health effects (WHO, 2006). This average index value was also calculated for the  $O_3$  at the 22 sites and found to be 1.4 %, 0.0 %, 2.8 % and 26.2 % in the autumn, winter, spring and summer, respectively. The daily maximum 1-h and 8-h mean exceeded CAAQS and IT-1 mainly occurred in the summer, with the largest appearing in June. According to these statistics,  $O_3$ , which threatens human health, is one of the main pollutants over northern China in the summer. In the following sections, we will focus on the spatial distribution for summer  $O_3$ , and then give some interpretations for summer  $O_3$  spatial distribution over northern China. Finally, combined with the smog production algorithm and space-based HCHO/ $NO_2$  column ratio, detailed control strategies for summer  $O_3$  will be obtained for different areas over northern China.

## 4.2 Spatial variations and interpretations of summer $O_3$

Spatial distributions of exceedances for average daily 1-h maximum ozone in the summer are presented in Fig. 12. Similar patterns were found for the average daily 8-h maximum ozone, but those results are not shown here. Inconsistent with the spatial distribution of ozone annual mean, the spatial distributions of the exceedances for daily 1-h and 8-h maximum ozone in the summer were consistent with the classification of the regional sites, with the highest values over Beijing and the surrounding areas and the lowest values over the northwestern mountainous area. The exceedances of daily 1-h maximum ozone at DT, CD, LS and ZJK in the mountainous area were 3.3, 3.3, 5.5 and 11.1 %, respectively, with the average being 5.8 %. However, the highest exceedances of daily 1-h and 8-h maximum ozone appeared at XL (45.3 % for 1-h and 70.9 % for 8-h maximum ozone) in the mountainous area, which are downwind

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of Beijing, Tianjin and Hebei, showing the strong influences of upwind areas. At CFD, QHD, TG, TJT and TS in the eastern coastal areas, exceedances of daily 1-h maximum ozone were 15.2, 3.3, 4.9, 8.0 and 5.8 %, with the average of 6.1 %, the second lowest only to the one in the mountainous areas. The relative highest exceedances of daily 1-h maximum ozone appeared over Beijing and the surrounding areas with the average of 31.3 %. The southern plains area presented modest exceedances of daily 1-h maximum ozone with an average of 15.7 %. According to the above analysis, we found that the average daily maximum ozone concentrations for the summer were distinct over different areas. Therefore, what are the sources in different areas? Kley et al. (1999) indicated  $O_x$ , defined as:  $O_x = O_3 + NO_2$ , a quasi-conservative quantity, where the concentration of  $O_x$  represented the photochemical production of ozone. Figure 13 shows the spatial distribution of the average daily  $O_x$  range for summer. As shown in Fig. 13, the spatial distribution of the average daily  $O_x$  range in the summer was similar to the spatial distribution of exceedances for daily 1-h maximum ozone except at XL. XL, a regional site influenced by transport, exhibited the lowest  $O_x$  production rate ( $25.3 \text{ ppbv d}^{-1}$ ), as expected. The average photochemical production rate of four other mountainous sites and the eastern coastal sites were  $35.9 \text{ ppbv d}^{-1}$  and  $35.9 \text{ ppbv d}^{-1}$ , respectively, which were also much lower than that over the southern plains areas ( $48.8 \text{ ppbv d}^{-1}$ ) and Beijing and its surrounding areas ( $61.2 \text{ ppbv d}^{-1}$ ). These results exhibit the strongest ozone photochemical productions in plains areas, especially over Beijing and its surrounding area.

Soloman et al. (1999) indicated that among the four factors, meteorology, atmospheric chemistry, emissions and deposition, which lead to the accumulation of  $O_3$  in the troposphere, meteorological processes are dominant, causing large day-to-night, day-to-day, season-to-season, and year-to-year variations. High concentrations of  $O_3$  observed during stagnant conditions are characterized by high ambient temperatures, low wind speeds, ample sunlight, and the presence of abundant local or regional emissions of precursors species. While all of the temporal variations are mainly controlled by meteorology, what controls  $O_3$  spatial variations? Do meteorological processes

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also control  $O_3$  spatial variations and  $O_3$  production in these four different areas over northern China? Figure 14 shows the simulated average temperature, relative humidity, downward shortwave, and cloud fraction between 09:00 and 15:00 for the summer. As shown in Fig. 4d and Fig. 14, four different areas exhibit inconsistent meteorology.

Meteorology simulated over Beijing and its surrounding areas is characterized by high temperatures, high downward shortwave, low cloud cover, low relative humidity, weak southeast winds and relatively low PBLH compared with the southern plains areas. Similar to Beijing and its surrounding areas, the southern plains areas have a meteorology that is characterized by high temperatures, low relative humidity and weak southeast winds, but relatively low downward shortwave, high PBLH and high cloud cover. However, the meteorology of the eastern coastal area is characterized by low temperatures, low downward shortwave, high cloud cover, high relative humidity, low PBLH and strong southeast winds. In addition, meteorology over the northwestern mountainous area shows another feature, which is characterized by the lowest temperature, lowest relative humidity, highest downward shortwave, strong west winds and high PBLH.

From the above analysis, we found that the meteorology over Beijing and its surrounding area and the northwestern mountainous area are the most and the least fit for  $O_3$  photochemical production, respectively. In addition,  $O_3$  clearly displayed an increasing trend from eastern coastal area to the mountain front area. The trend of increasing ozone along the direction of the prevailing wind is consistent with the widespread regional-scale transport of oxidants. These results, consistent with spatial variations of  $O_3$  concentrations and  $O_x$  production rates, illustrate that the spatial distribution of summer  $O_3$  is dominated by the meteorology of different areas over northern China. However, what are the roles of emissions over northern China? As shown in Fig. 7 (Zhang et al., 2009), the plains area corresponds to more industrialization and automobiles, and the  $NO_x$  and  $VOC_s$  emissions for most of the grid (0.5 degree  $\times$  0.5 degree) over the plains areas are both over  $18 \text{ kton yr}^{-1}$ . However, in the mountainous areas,  $NO_x$  and  $VOC_s$  for most of the grid are both less than  $6 \text{ kton yr}^{-1}$ . The emissions of

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precursors are weakly correlated with  $O_3$  concentrations over northern China, especially in the eastern coastal area, which exhibits high emissions of precursors but low  $O_3$  concentrations and  $NO_x$  production rates. Although there is no direct relationship between the emissions of precursors and  $O_3$  concentrations, it should be noted that abundant emissions over the plains areas are essential conditions for  $O_3$  photochemical production over Beijing and its surrounding area. In summary, we arrived at two major conclusions. First, the  $O_3$  photochemical production efficiency was highly non-linear and abundant  $NO_x$  and  $VOC_s$  emissions over the eastern coastal area could not result to high  $O_3$  concentrations in this area, indicating that the observed daily maximum  $O_3$  concentrations are dominated by meteorological processes. Second, the abundant  $NO_x$  and  $VOC_s$  emissions are essential for ozone production. Therefore, the domination of meteorological process is based on the present spatial distribution of emissions. Considering the similar year-to-year meteorology in the summer, abatement of emissions of precursors is a unique and effective method to change the spatial variations of summer  $O_3$  and diminish the regional photochemical pollution over northern China.

### 4.3 $O_3$ - $NO_x$ - $VOC_s$ sensitivity diagnosis and control strategy of summer $O_3$

Figure 15 shows the spatial distribution of the extent of reaction over northern China between June and August 2010. The  $E(t)$ s of nearly all of the sites except XL were less than 0.4, which indicated  $NO_x$ -saturated conditions at these sites. As discussed above, the main source of ozone for XL is the transport from upwind areas. Photochemical production in XL was lower than that at other sites, as shown by the much higher  $E(t)$  value (0.74) at XL. This high value of  $E(t)$  illustrates that the  $O_3$ - $NO_x$ - $VOC_s$  sensitivity of XL should generally be considered transitional or indeterminate.

Figure 16 shows the GOME-2 HCHO/ $NO_2$  column ratio over northern China between June and August 2010. The resulting gradient of  $NO_x$ -saturated conditions during the summer over North China Plain, but  $NO_x$ -sensitive conditions over the northern mountainous areas, were consistent with the results from the SP algorithm and photochemical model calculations of Tang et al. (2010b). However, GOME HCHO/ $NO_2$  ratios



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greater than one during the summer over eastern China indicated  $\text{NO}_x$ -sensitive conditions in 1997 (Martin, et al., 2004), which is inconsistent with our analysis. Since 1997, with the strong economic and industrial development in China, the anthropogenic  $\text{VOC}_s$  and  $\text{NO}_x$  emissions significantly changed. Smedt et al. (2010) showed that the HCHO trend in the BTH area was  $4 \pm 1.4 \% \text{ yr}^{-1}$  between 1997 and 2009, while the  $\text{NO}_2$  trend in Beijing is  $11 \pm 4 \% \text{ yr}^{-1}$  between 1996 and 2006 (Van der A et al., 2006, 2008). Calculated using the above HCHO and  $\text{NO}_2$  trends, the HCHO column concentrations increased 1.7 times from 1997 to 2010, while the  $\text{NO}_2$  column concentrations increased 3.9 times in the same period. In other words, the growth rate of  $\text{NO}_2$  was much higher than that of HCHO, indicating a decreasing trend of HCHO/ $\text{NO}_2$  between 1997 and 2010, which induced a clear transition from  $\text{NO}_x$  sensitivity in 1997 to  $\text{NO}_x$  saturation in 2010 over the plains areas.

The above finding is extremely important to the design of an effective  $\text{O}_3$  control strategy for northern China. Our results suggest that the control of  $\text{VOC}_s$  would be most effective for controlling the ozone over North China Plain, while  $\text{NO}_x$  control would be counterproductive in this area. In addition,  $\text{NO}_x$  control would be the most effective measure for controlling ozone over the mountainous areas, related to the lower emissions in this area. In this context, as noted earlier, the concentrations of  $\text{O}_3$  in the upwind region are low. However, the high  $\text{VOC}_s$  and  $\text{NO}_x$  emissions in this area (Fig. 10) contribute significantly to the  $\text{O}_3$  precursors in the downwind regions. Therefore, an effective  $\text{O}_3$  control strategy also needs to consider  $\text{O}_3$  formation from the reaction of its precursors during the transport from the upwind region.

## 5 Conclusions

As one of the developing urban agglomerations in China, the BTH area has been confronted with severe air pollution for several decades. The air pollutants originate from sources both in the urban areas and in the countryside, particularly in the polluted North China Plain. To make effective polices for controlling the photochemical pollution over northern China, it is crucial to know the air quality in this area. During the Project

of Atmospheric Combined Pollution Monitoring over Beijing and its surrounding areas, 22 sites over northern China were selected as representative sites. For the first time, the concentrations of ozone and nitrogen oxide were observed at the ground level from 1 September 2009 to 31 August 2010.

During the observation period, we examined the seasonal, diurnal and spatial variations of meteorological factors, ozone and  $\text{NO}_x$  over northern China. Our results suggest distinct seasonal and diurnal cycles in meteorology, ground-level ozone and  $\text{NO}_x$  over northern China. Lower ozone concentrations were generally observed in the winter and during early morning hours, while high ozone levels were typically found in the summer and in the afternoon. The annual average ozone concentrations were lower in plain areas compared with the sites in the northwestern mountainous areas. The lower ozone concentrations in the plains areas were due to the titration effect of high  $\text{NO}_x$  emission in this area. When the summer ozone was separately analyzed with the meteorological model, the emissions, the smog production algorithm and space-based observations, four major conclusions were concluded:

1. A decrease in regional  $\text{O}_3$  photochemical production seems to be coupled with an increasing ozone dry deposition in July, and this combination leads to the highest ozone concentrations in June over northern China.
2. The 22 sites can be aggregated into four categories based on daily 1-h maximum ozone: are the eastern coastal area, southern plains area, Beijing and the surrounding area and the northwestern mountainous area. These results indicate inconsistent ozone maximum concentrations over different areas, with the highest and lowest exceedances in Beijing and its surrounding area and the northwestern mountainous area, respectively.
3. Spatial variations in the daily maximum ozone are dominated by meteorological processes over northern China. The highest daily maximum ozone concentrations observed over Beijing and the surrounding area are dominated by high temperatures, high downward shortwave, low cloud cover, low relative humidity, weak

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southeast winds and low PBLH, with the presence of abundant  $\text{NO}_x$  and  $\text{VOC}_s$  emissions over the plains areas.

4. The low extent of reaction and  $\text{HCHO}/\text{NO}_2$  ratio suggest that  $\text{O}_3$  photochemistry was usually in the  $\text{VOC}$ -limited regime over the plains area in the summer. Our results suggest that control of  $\text{VOC}_s$  would be most effective for controlling ozone over North China Plain, while  $\text{NO}_x$  control would be counterproductive in this area. However, considering the high emissions in the upwind area, an effective  $\text{O}_3$  control strategy will also need to consider the  $\text{O}_3$  formation from reactions by its precursors during the transport from the upwind region.

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**Table 1.** Summary of statistics for surface-layer temperature, relative humidity, wind speed and precipitation.

Parameter	ME	MBE	RMSE	<i>R</i>
Temperature (°C)	0.24	1.01	1.29	0.99
Relative humidity (%)	5.85	7.30	9.26	0.77
Wind speed (m s <sup>-1</sup> )	-1.91	1.91	2.14	0.56
Precipitation (mm)	3.64	20.27	32.65	0.76

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**Table 2.** Results of factor analysis for daily maximum ozone.

Sites	Factor1	Factor2	Factor3
BD	0.44	<b>0.77</b>	0.29
BJT	<b>0.87</b>	0.33	0.32
CD	0.43	0.47	0.49
CFD	0.37	0.46	<b>0.75</b>
CZ	0.17	<b>0.64</b>	0.55
DT	0.39	0.56	0.31
GA	<b>0.60</b>	0.64	0.36
HJ	0.45	<b>0.63</b>	0.49
HS	0.22	<b>0.68</b>	0.40
LF	<b>0.69</b>	0.47	0.48
LS	0.23	0.52	0.24
LTH	<b>0.83</b>	0.32	0.37
QHD	0.40	0.36	<b>0.61</b>
SJZ	0.47	<b>0.71</b>	0.28
TG	0.46	0.38	<b>0.69</b>
TJT	0.37	0.34	<b>0.69</b>
TS	0.45	0.37	<b>0.74</b>
XL	0.54	0.46	0.57
YF	<b>0.85</b>	0.33	0.32
YJ	<b>0.64</b>	0.37	0.52
ZJK	0.48	0.42	0.36
ZZ	<b>0.69</b>	0.52	0.32

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**Table 3.** Inter-site uniformity statistics for four categories.

Site Category	NO <sub>x</sub>			O <sub>3</sub>			O <sub>3,max</sub>		
	<i>R</i>	CV(%)	COD	<i>R</i>	CV(%)	COD	<i>R</i>	CV(%)	COD
Southern plain area	0.93	27.9	0.19	0.96	19.3	0.14	0.97	16.0	0.12
Beijing and surrounding area	0.87	31.4	0.22	0.97	18.0	0.12	0.97	14.5	0.09
Northwest mountainous area	0.67	58.0	0.36	0.82	37.6	0.23	0.94	19.8	0.13
Eastern coastal area	0.91	22.1	0.17	0.96	17.8	0.13	0.96	13.0	0.09

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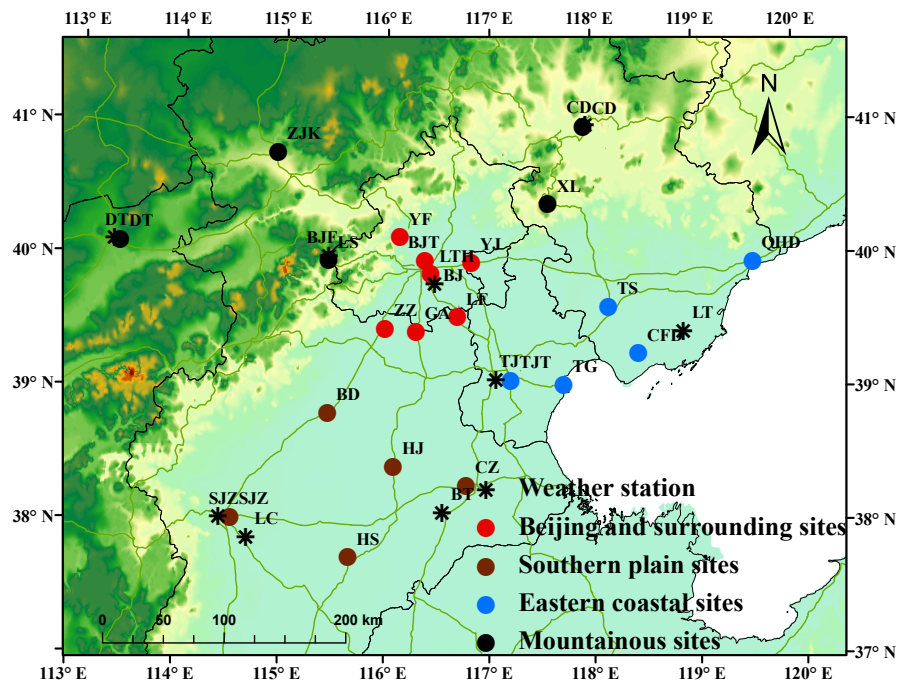
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**Fig. 1.** The Air Quality Network and weather stations over northern China.

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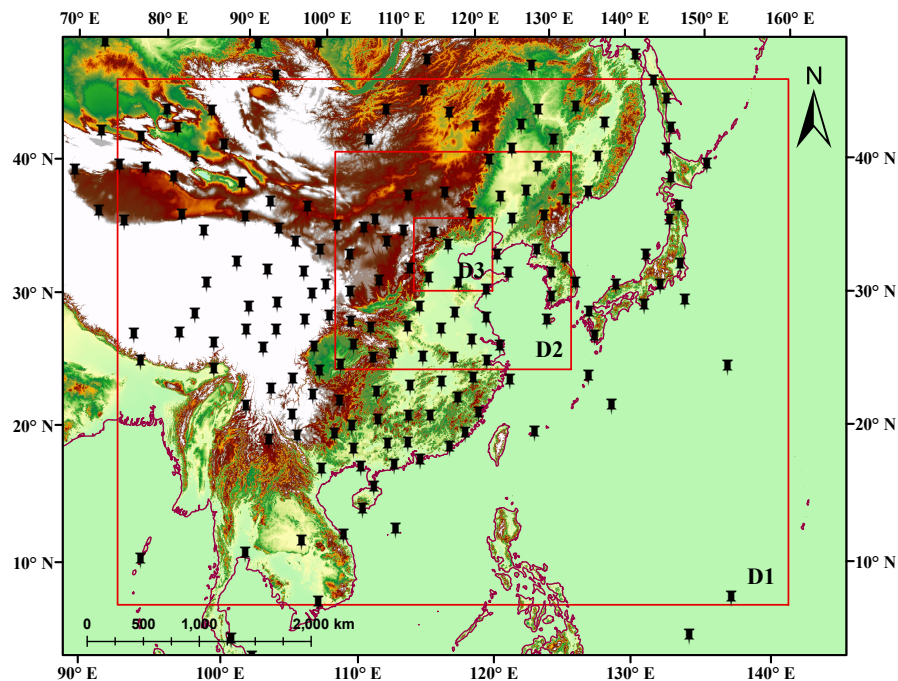
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**Fig. 2.** Topography, domain settings and upper sites used to correct the first guess fields in WRF.

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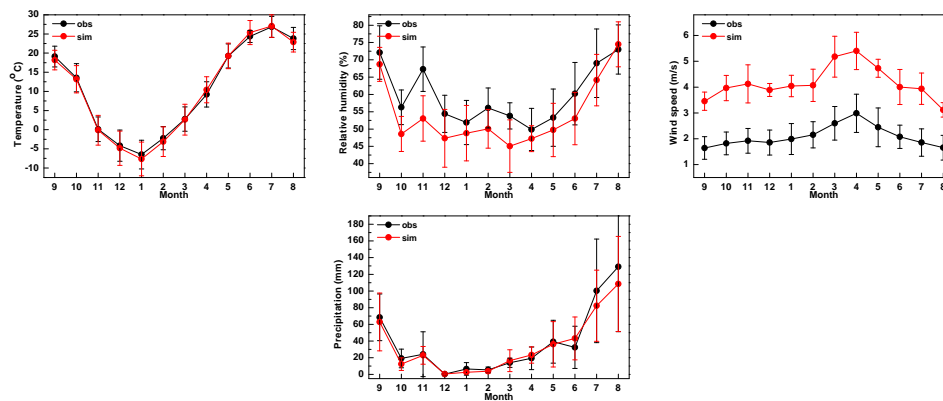
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**Fig. 3.** Simulated and observed monthly variations of temperature, relative humidity, wind speed and precipitation. The value of each species represents an average of measurements and simulations taken from ten weather stations over northern China.



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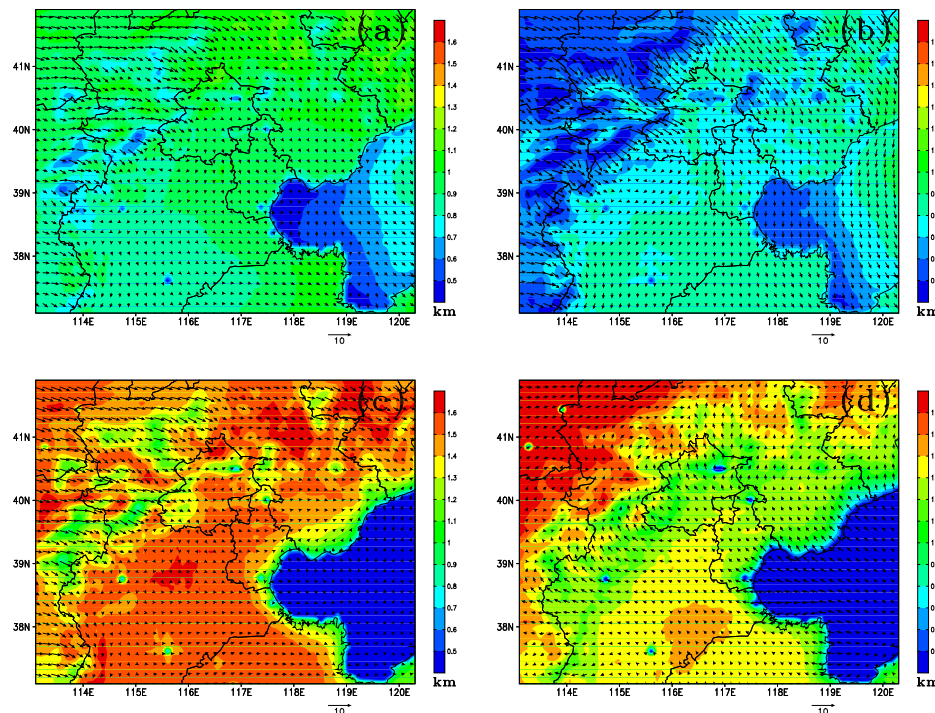
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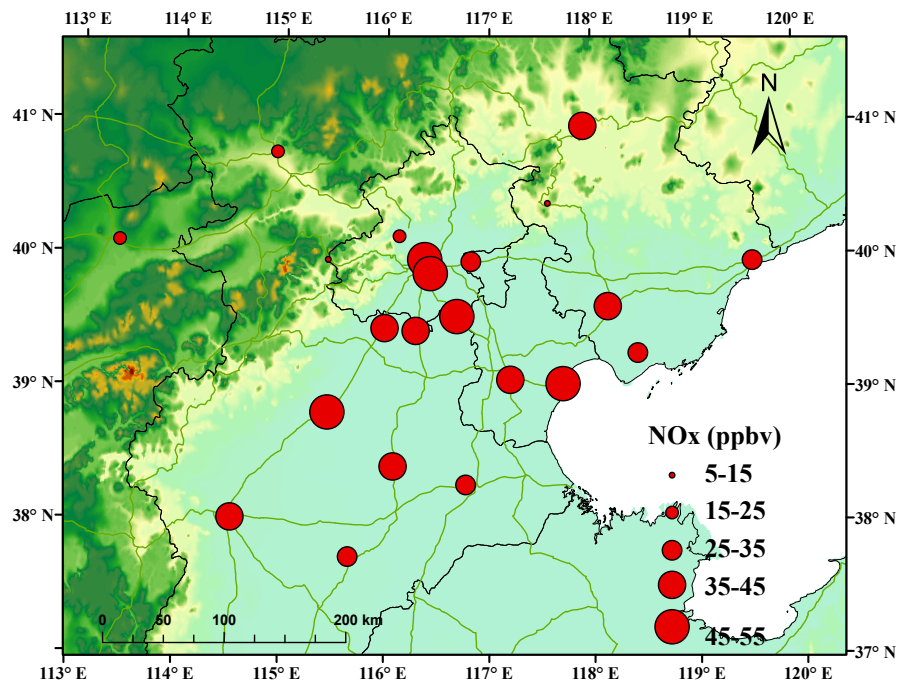
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**Fig. 4.** Averaged planetary boundary layer height and vector winds between 09:00 and 15:00 for autumn (a), winter (b), spring (c), and summer (d).

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**Fig. 5.** Spatial distribution of average NO<sub>x</sub> over northern China.

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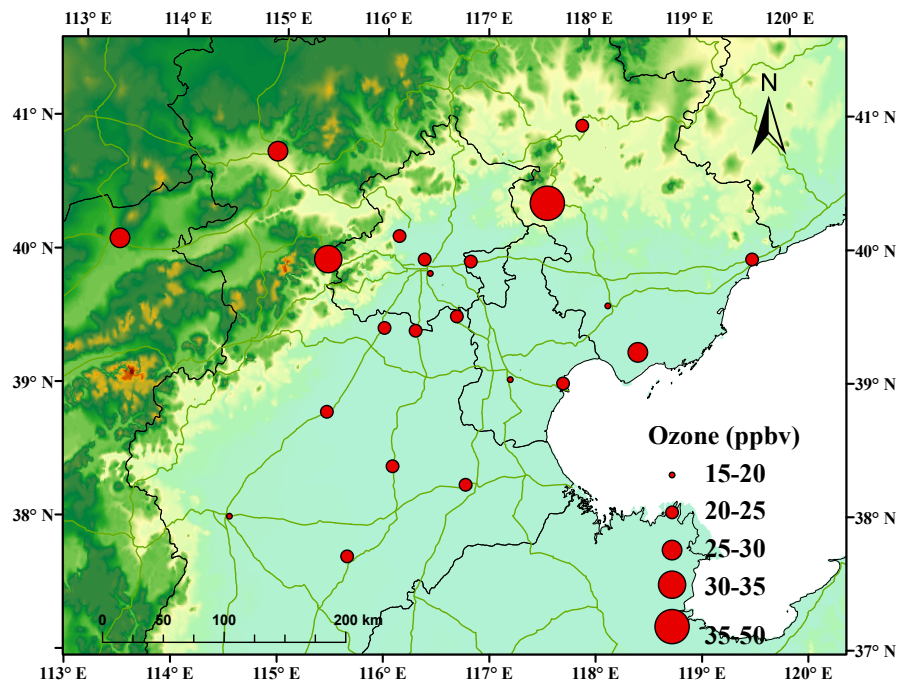
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**Fig. 6.** Spatial distribution of average  $O_3$  over northern China.[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

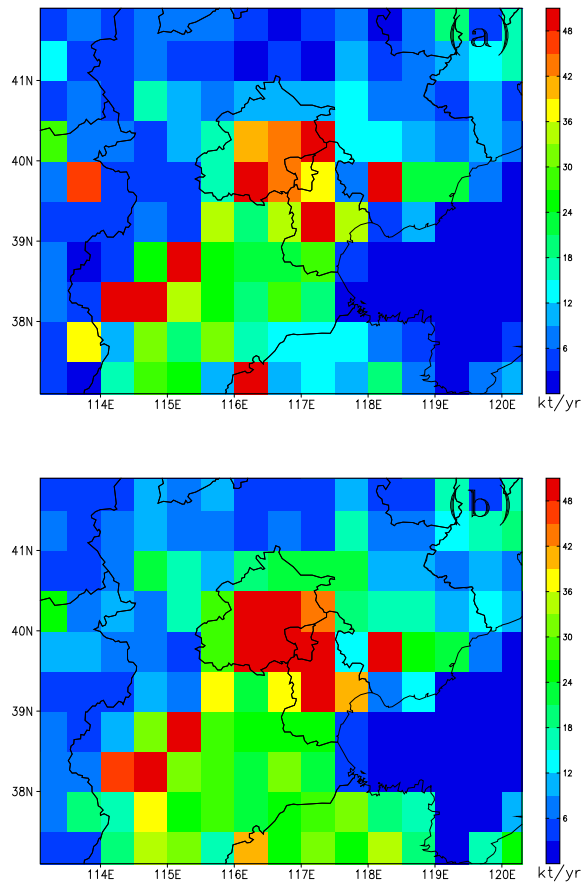


Fig. 7.  $\text{NO}_x$  (a) and  $\text{VOC}_s$  (b) emissions in 2006.

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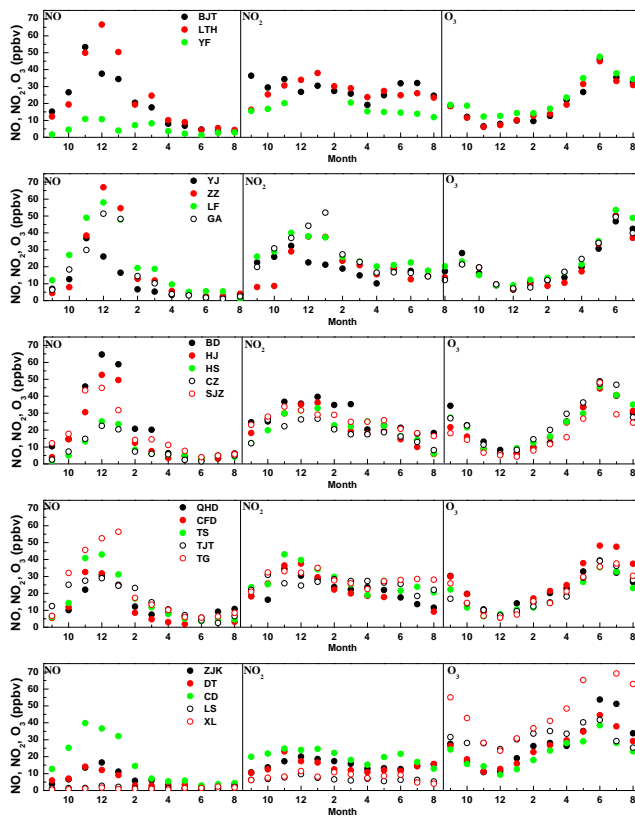
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**Fig. 8.** Seasonal variations of NO<sub>x</sub> and O<sub>3</sub>.

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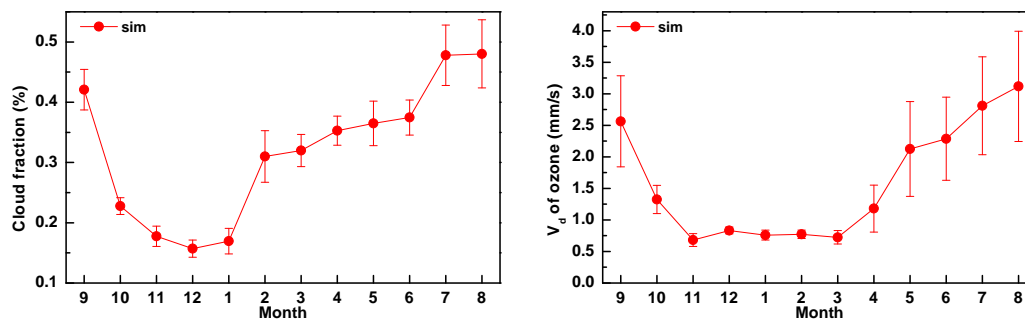
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**Fig. 9.** Simulated monthly variations of cloud fraction and dry deposition of ozone. The value of each species represents an average of measurements and simulations taken from ten weather stations over northern China.

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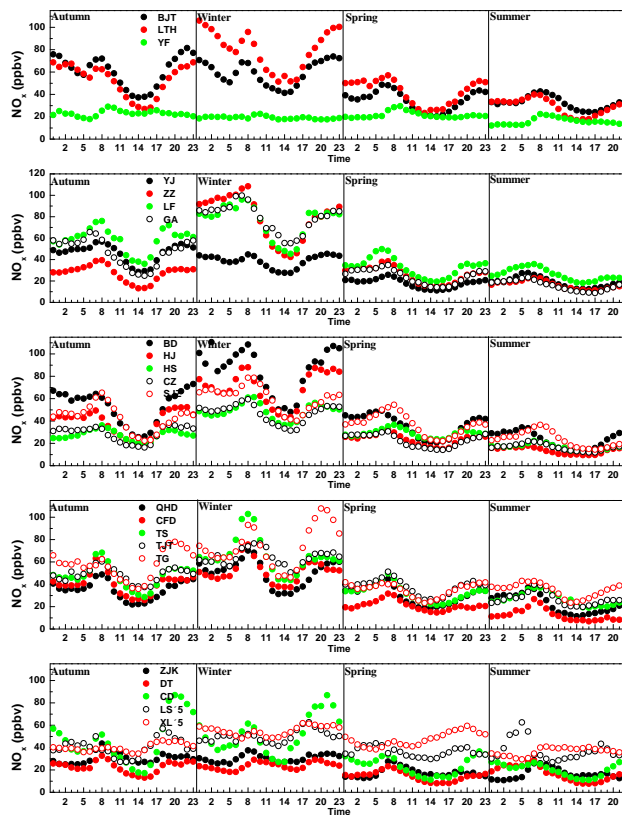


Fig. 10. Diurnal variations of  $\text{NO}_x$  for four seasons.

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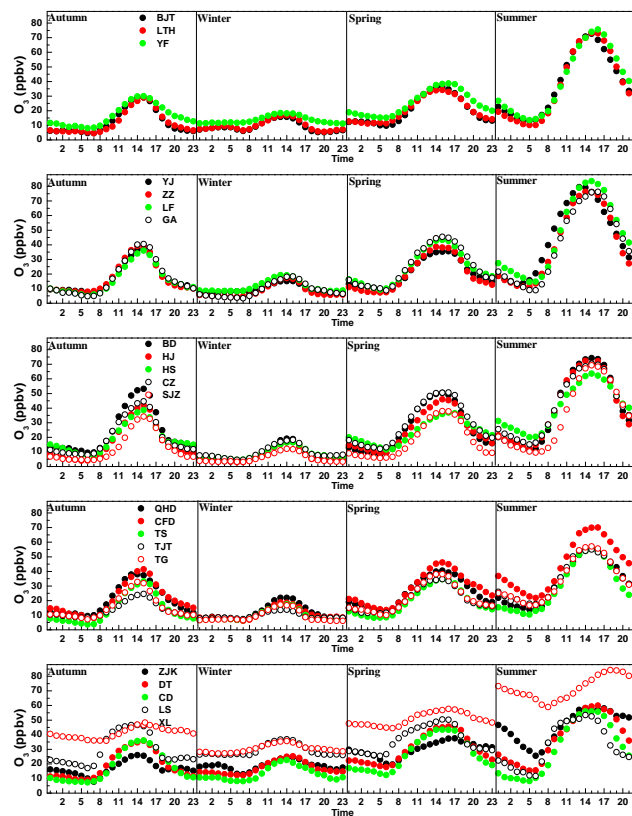


Fig. 11. Diurnal variations of  $O_3$  for four seasons.

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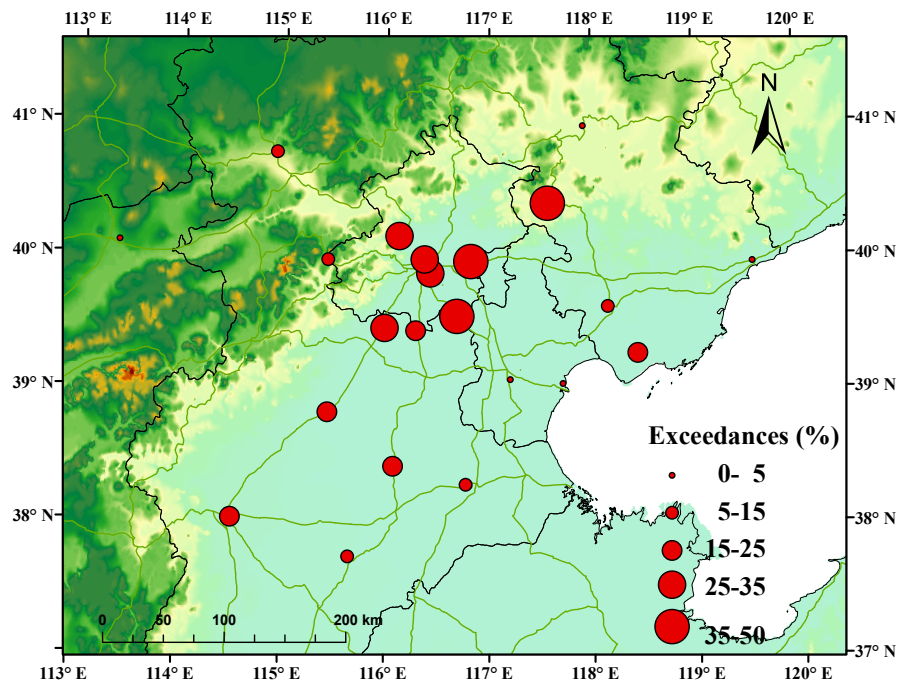
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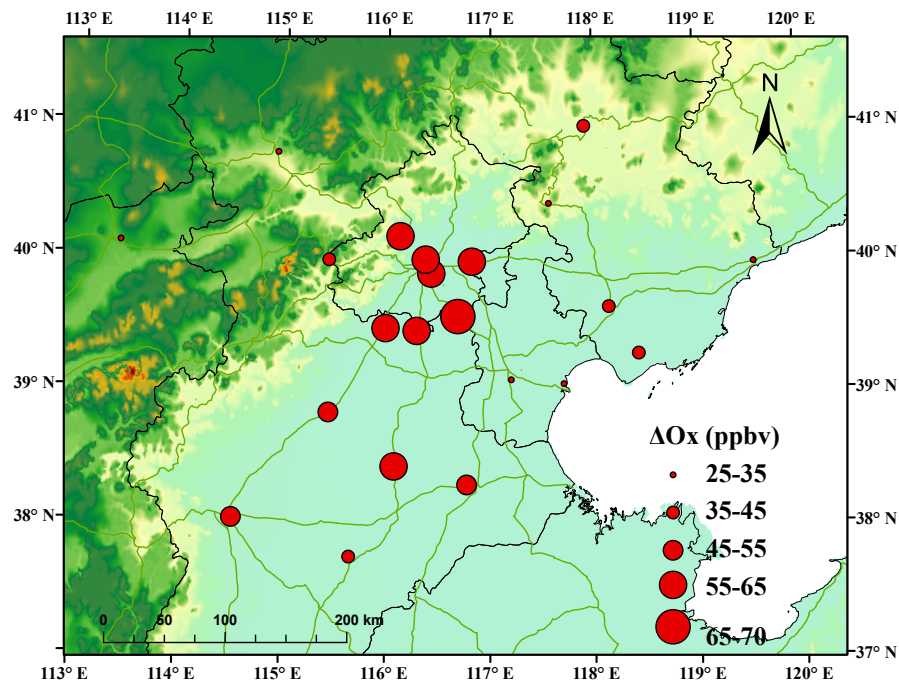


**Fig. 12.** Spatial distribution of exceedances for average daily 1-h maximum O<sub>3</sub> in the summer.

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**Fig. 13.** Spatial distribution of averaged daily  $O_x$  range for the summer.

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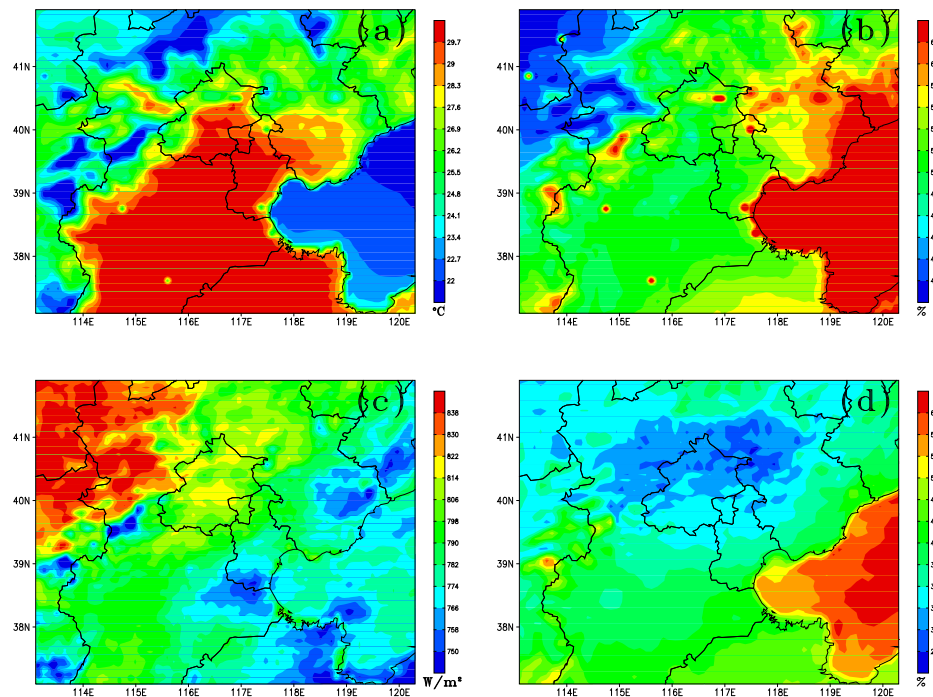
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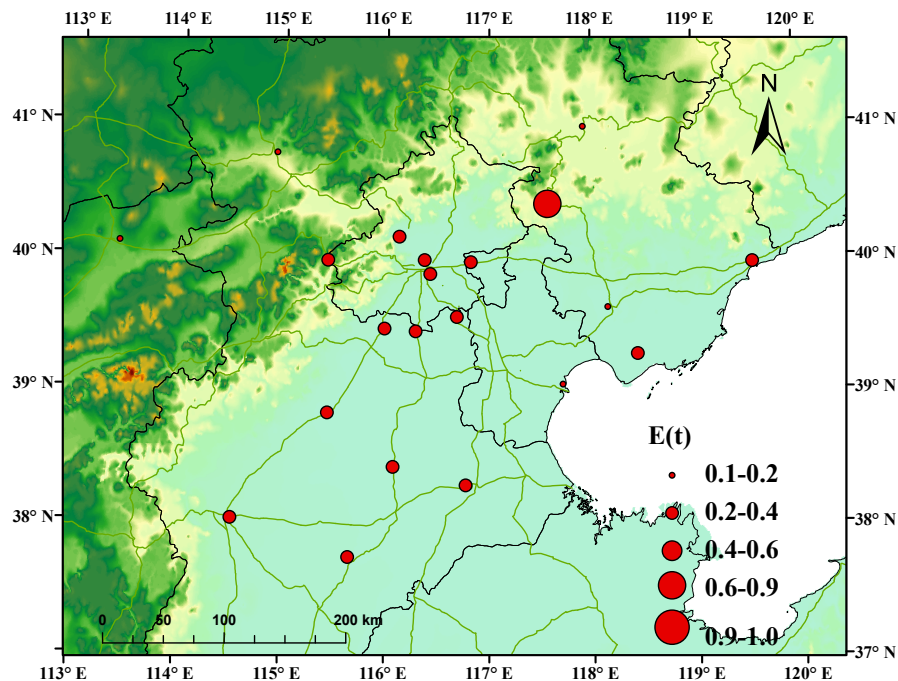
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**Fig. 14.** Simulated average temperature (a), relative humidity (b), downward shortwave (c), and cloud fraction (d) between 09:00 and 15:00 for summer.

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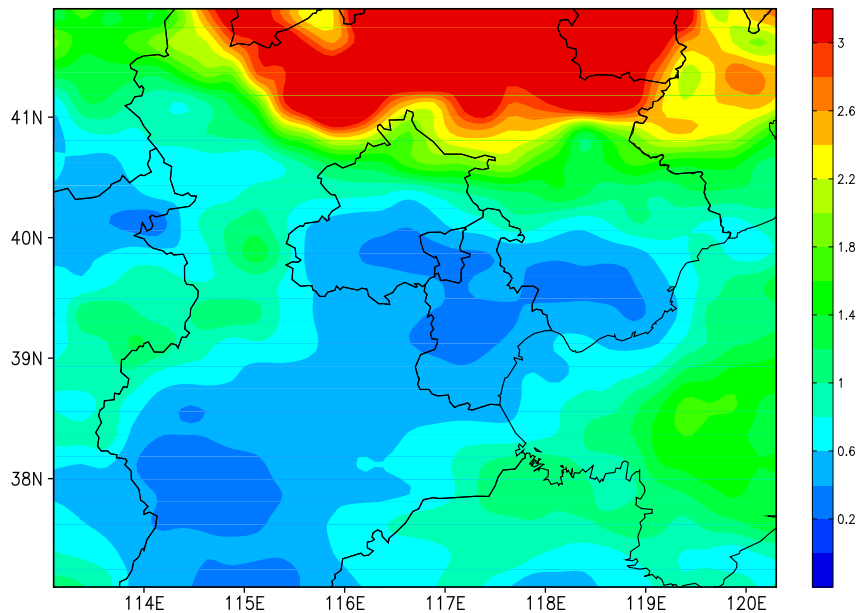


**Fig. 15.** Spatial distribution of the  $E(t)$  over northern China for summer.

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**Fig. 16.** HCHO/NO<sub>2</sub> ratio of GOME-2 for the summer.

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