Atmospheric Research 187 (2017) 128–137

Contents lists available at ScienceDirect

Atmospheric Research

journal homepage: www.elsevier.com/locate/atmosres

Modelling study of boundary-layer ozone over northern China - Part I: Ozone budget in summer

Guiqian Tang [a], Xiaowan Zhu [a], Jinyuan Xin [a], Bo Hu [a], Tao Song [a], Yang Sun [a], Jinqiang Zhang [b], Lili Wang [a], Mengtian Cheng [a], Na Chao [c], Lingbin Kong [d], Xin Li [a,e], Yuesi Wang [a,*]

[a] State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry (LAPC), Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029, China
[b] Key Laboratory of Middle Atmosphere and Global Environment Observation (LAGEO), Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029, China
[c] Environmental Science Research & Design Institute of Zhejiang Province, Hangzhou 310007, China
[d] College of Atmospheric Sciences, Key Laboratory of Arid Climatic Change and Reducing Disaster of Gansu Province, Lanzhou University, Lanzhou 730000, China
[e] Mentougou District Government, Beijing 102300, China

ARTICLE INFO

Article history:
Received 14 August 2016
Received in revised form 15 October 2016
Accepted 25 October 2016
Available online 17 November 2016

Keywords:
Integrated process rate
Ozone sensitivity
Chemical production

ABSTRACT

Regional photochemical pollution caused by ozone (O_3) is serious in northern China during summer. In this study, we combined network observation data with the Fifth-Generation Pennsylvania State/National Centre for Atmospheric Research Mesoscale Model -Community Multiscale Air Quality (MM5-CMAQ) model system to simulate O_3 and its precursors'concentrations over northern China in June 2008. Comparisons of the simulations and observations indicate that the model can accurately reproduce the temporal and spatial distributions of temperature, humidity, and wind as well as the evolution of O_3 and its precursors over northern China. The monthly mean of the total oxidants (nitrogen dioxide + O_3) at 15:00 LT exceeded 90 ppbv across the North China Plain, thereby indicating significant photochemical pollution in this area. Vertical diffusion is the main source of the near-ground O_3, with contributions of more than 20 ppbv h^{-1} in the urban areas. Dry deposition and chemical reactions are the main sinks for O_3, with contributions of more than 20 ppbv h^{-1} and 7 ppbv h^{-1} in the forest and urban areas, respectively. Although vertical diffusion is the main source of near-ground O_3, photochemical reactions dominate the O_3 concentrations in the boundary layer because of the circulation between the lower and upper boundary layers. Considering that O_3 is mainly produced in the upper boundary layer, both nitrogen oxide and volatile organic compounds should be controlled on the North China Plain. The results presented here are intended to provide guidance for redefining strategies to control photochemical pollution over northern China.

1. Introduction

Nitrogen oxides (NOx) and volatile organic compounds (VOCs) are the two main precursors of ozone (O_3) in urban areas. NOx and VOCs react in the presence of sunlight, and mixtures of O_3 precursors and secondary pollutants (e.g., O_3 and peroxyacetyl nitrate) are called photochemical smog. O_3 is the main component of photochemical smog and one of its main indicators (Seinfeld and Pandis, 1998).

In urban areas, the concentrations of precursor pollutants have increased gradually over the past several decades because of increasing human activity, which has enhanced photochemical reactions and induced the deterioration of photochemical pollution (Crutzen, 1988; Houweling et al., 1998; Logan, 1985; Tang et al., 2009; Tang et al., 2012). Under favourable weather conditions, photochemical smog that originates from urban areas can migrate for hundreds of kilometres, which can lead to significant regional photochemical pollution (Ma et al., 2014; Ma et al., 2016; Mckeen et al., 1991; Sillman and Samson, 1995; Vautard et al., 2005; Xu et al., 2011; Xu et al., 2008). Moreover, the impacts of increases in urban O_3 in the boundary layer are becoming increasingly significant and affect the variations of O_3 concentrations globally (Akimoto, 2003; Hough and Derwent, 1990; Jacob et al., 1999; Kley et al., 1994; Levy et al., 1997; Lin et al., 2012; Xu et al., 2016).

Since the 1980s, the economic development and increased energy use in northern China generated the photochemical smog pollution in Beijing (BJ) (Tang et al., 1995). To mitigate the photochemical pollution, the Beijing Environmental Protection Bureau and other research institutes established an observation network to study the photochemical pollution in BJ since 2001 (Tang et al., 2009). O_3 production is sensitive to the VOCs concentrations in the urban area of BJ (Chou et al., 2009; Sun et al., 2006; Tang et al., 2009; Xu et al., 2006; Wang et al., 2009). According to these researches, the BJ municipal government has gradually tightened regulations to alleviate emissions from motor vehicles and

* Corresponding author.
E-mail address: wys@mail.iap.ac.cn (Y. Wang).

http://dx.doi.org/10.1016/j.atmosres.2016.10.017
0169-8095/© 2016 Elsevier B.V. All rights reserved.

industrial and domestic sources in the city. However, because of the rapid industrial development of Tianjin (TJ) and Hebei (Langfang (LF), Baoding (BD), Shijiazhuang (SJZ) and Tangshan (TS), among other aeras), primary and secondary pollutants are transported by southerly or easterly winds and react over the North China Plain, thereby resulting in photochemical pollution with obvious regional characteristics (Dufour et al., 2010; Han et al., 2013; Han, 2011; Ran et al., 2012; Tang et al., 2012; Wang et al., 2014; Wang et al., 2015; Xue et al., 2014). Because the North China Plain is located west of the Taihang Mountains and south of the Yan mountains, the mountain plain winds dominate the North China Plain under weak synoptic systems, and southwest and northeast winds occur during the day and night, respectively (Tang et al., 2016b). Peak O_3 concentrations occur in the afternoon, and the piedmont areas are influenced by urban plumes because of the plain winds in the North China Plain (Ge et al., 2012; Ma et al., 2013; Xu et al., 2011; Wang et al., 2006; Wang et al., 2009). For example, the urban plumes of BJ contributed 21.8 ppbv at the northeast station of Shangdianzi from 2004 to 2006 (Lin et al., 2008). Therefore, coordinated regional emission reduction measures are the most effective methods of controlling regional O_3 pollution over northern China.

Cooperation between other governments and research institutes produced air quality safeguards research implemented form 2006 to 2008 prior to the 29th Olympic Games in BJ, and the Beijing-Tianjin-Hebei Atmospheric Environment Monitoring Network was established by the Institute of Atmospheric Physics, Chinese Academy of Sciences (Xin et al., 2010). After the Beijing Olympics, the network was expanded from 17 to 25 stations to develop an in-depth understanding of photochemical pollution over northern China on a regional scale (Tang et al., 2012; Wang et al., 2014). Because additional regulations were conducted to reduce the emissions over northern China after 2006, a decreasing trend in the ratio of formaldehyde (HCHO) and nitrogen dioxide (NO_2) was observed between 1997 and 2010, thus indicating a clear transition from NOx sensitivity to VOCs sensitivity over the North China Plain (Martin et al., 2004; Tang et al., 2012). In addition, different O_3 sensitivity were observed in the plains and mountainous areas, with VOCs sensitivity and NOx sensitivity observed in the North China Plain and mountainous areas, respectively (Tang et al., 2012). As a result, changes in the NOx and VOCs emissions have led to increases in O_3 concentrations year-by-year over northern China, which indicates that the atmospheric oxidation capacity may increase with on-going urbanization and industrialization (Fu and Tai, 2015; Hayashida et al., 2015; Huang et al., 2015; Ma et al., 2016; Wang et al., 2012a).

Because of the increasingly serious photochemical pollution over northern China, control strategies are urgently required. However, inappropriate policies may be implemented because of the large research gaps in the O_3 budget over northern China. Previous studies have indicated that the lower tropospheric O_3 maximum in summer is a result of strong photochemical production and regional transport (Ding et al., 2008). Recently, higher concentrations of O_3 were observed in the upper boundary layer, especially above the stable boundary layer at night (Chen et al., 2013; Ma et al., 2011; Ma et al., 2013; Sun et al., 2006; Zheng et al., 2005). However, past investigations were fragmented and mostly limited in scope to the near-ground level. Large gaps and uncertainties remain in our understanding of the characteristics of the O_3 distribution in the vertical direction, which leads to a lack of specificity in the O_3 control strategies over northern China.

To further control the regional O_3 pollution over northern China, we used an air quality model to simulate the spatial and temporal distributions of O_3 and its precursors in June 2008. Using data from the observation network, the model results were validated in the horizontal and vertical directions to illustrate the O_3 budget in the boundary layer over northern China. Finally, based on the O_3 budget at different heights in the boundary layer, the sensitivity of O_3 production to its precursors was evaluated to identify strategies for controlling photochemical pollution over northern China.

2. Methodology

2.1. Air quality model simulation

2.1.1. Model configuration

We used the Community Multiscale Air Quality (CMAQ) model (version 4.6) to simulate the boundary layer O_3 and its precursors over northern China. The CMAQ is a comprehensive Eulerian grid model that simulates the complex interactions among multiple air pollutants between regional and urban scales (Dennis et al., 1996). In this study, the model is configured to include a detailed implementation of the piecewise parabolic method for horizontal and vertical transport as well as the RADM2 (Regional Acid Deposition Model 2) chemical mechanism with isoprene extensions (CIS4) (Zimmermann and Poppe, 1996), dry deposition, and cloud physics and chemistry (Dennis et al., 1996).

The CMAQ is configured to have 3 domains (Fig. 1). The mother domain with a horizontal grid resolution of 81 km covers the entire area of China, the 27-km grid-spacing inner domain covers eastern China, and the third domain with a resolution of 9 km covers BJ, TJ, and Hebei Province. All the grids have 15 vertical layers extending from the surface to approximately 15 km above the ground. The vertical layers are unevenly distributed with eight layers in the first kilometre from the ground, and a surface layer of approximately 38 m. Because June is the month with the most photochemical pollution over northern China (Tang et al., 2012), we selected the period of June 2008 to analyse the regional photochemical pollution. Our CMAQ simulations were conducted from

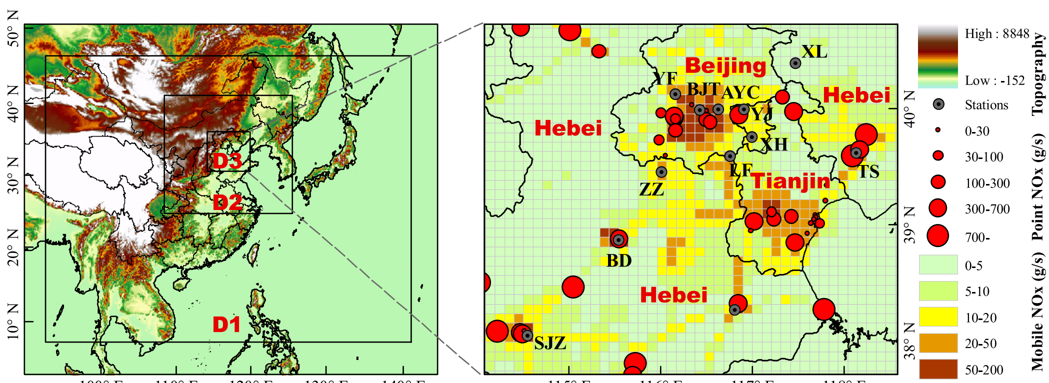

Fig. 1. Topography, domain settings, observation stations and emissions over northern China.