East China Plains: A "Basin" of Ozone Pollution

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Economic growth and associated pollution emissions in China are concentrated over three connected plains to the east. In this work, we analyze an episode of highly elevated ozone over East China on June 9−14, 2004, using a 3-D chemical transport model. During this episode, the East China plains were under a high-pressure system, which suppressed the ventilation of pollutants from the boundary layer. Simulated ozone concentrations over a major fraction of East China reached high levels, all the way down to the Pearl River Delta region in the southern border. The convergence of pollutant emissions and population over the vast stretch of the geographically flat plains of East China makes the region susceptible to high-ozone exposure. During this episode, the high-O_3 region extended over an area >1 million km^2, which hosts a population of >800 million people. Model results indicate that controlling anthropogenic NO_x emissions effectively reduces the area with high-ozone exposure.

Introduction

Ozone (O_3), an important chemical compound of the earth's atmosphere, is a greenhouse gas contributing to global warming and poses health and ecology problems on the ground level (*1*). During the past two decades, the rapid economic growth in China has resulted in a significant increase in the emissions of ozone precursors (*2−4*), and these emissions lead to the formation of elevated ozone near the surface. In recent years, more and more observations have found high-ozone episodes in China, mostly over East China (110−120 E, 25−42 N) (*5−9*).

East China hosts the economic engine of the country with >60% of the industries and population (Figure 1a) and most of the major cities. Geographically, this region consists of three plains, the North China Plain, Northeast Plain, and Chang Jiang Downstream Plain, with high mountains and plateaus to the west and ocean to the east (Figure 1b). The elevation over most of the region is less than 200 m. A similar geographical feature exists over the "South Coast Basin" of California in the United States (mountain regions to its east and ocean to its west). The "basin" feature contributes to high-ozone episodes there (*10, 11*). As in the case of California, the East China plains also have large pollutant emissions (*3*). When the atmospheric circulation becomes favorable, widespread high-ozone pollution can form over East China due to the recirculation of pollutants. Unlike the South Coast Basin of California, East China plains are much larger (>1 million km^2) and are occupied by a larger population (>800 million). Thus, the environmental impact of ozone pollution is much greater. Unfortunately, it is difficult to identify such a basin feature of high ozone over East China since ground ozone monitoring sites in China are mostly in major cities and the measurement data are difficult to obtain for researchers outside China. In this work, we identify the basin feature using the simulations from a regional chemical transport model (REAM) and surface ozone measurements.

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Materials and Methods

Model. The 3-D regional chemical transport model (REAM) driven by MM5-assimilated meteorological fields was described by Choi et al. (*12*). Previously, this model was applied to investigate a number of tropospheric chemistry and transport problems at northern midlatitudes (*13−17*) and in the polar regions (*18−20*). In this work, the REAM model is applied to ozone simulations over East Asia. The model has a horizontal resolution of 70 km with 23 vertical layers below 10 hPa. Meteorological fields are assimilated using MM5 constrained by the National Center for Environmental Prediction (NCEP) reanalysis products. The horizontal domain of MM5 has five extra grids beyond that of REAM on each side to minimize potential transport anomalies near the boundary. Most meteorological field inputs are archived every 30 min except those related to convective transport and lightning parametrizations, which are archived every 5 min. Chemical initial and boundary conditions for chemical tracers in REAM are obtained from the global simulation for the same period using the GEOS-CHEM model driven by GEOS-4-assimilated meteorological fields (*21*). A more detailed model description is provided in the Supporting Information.

Emissions. Biogenic emission algorithms and inventories are adapted from the GEOS-CHEM model (*12*). The anthropogenic emissions of tracers other than NO_x are taken from a recent bottom-up Asian emission inventory developed by Streets et al. for the 2006 INTEX-B campaign (the details can be obtained from http://www.cgrer.uiowa.edu/EMISSION_DATA_new/index_16.html). Anthropogenic NO_x emissions for 2004 are obtained by scaling the NO_x emissions derived by Zhao et al. (*22*) for 2007 over East Asia with an annual increasing rate of 8% in China (*22*). The NO_x and CO emissions from biomass burning are obtained from the Global Fire Emissions Database, version 2 (GFEDv2.1) (*23*).

Figure 2 compares the anthropogenic NO_x emissions over China to those over the United States in 2004. The anthropogenic NO_x emission over the United States was prepared by the sparse matrix operator kernel emissions (SMOKE) model (http://cf.unc.edu/cep/empd/ products/smoke/index.cfm) for 2004 projected from the VISTAS 2002 U.S. emission inventory. The estimated total NO_x emission over China is 6.2 Tg/yr, 35% higher than the 4.6 Tg/yr emission over the United States. While high NO_x emissions are mostly over major metropolitan regions in the United States, high NO_x emissions are more spatially spread over East China, corresponding roughly to the connected China plains shown in Figure 1b.

Results and Discussion

East China Plains as a Large Ozone Basin. In this work, we identify the basin feature using the simulations from a regional chemical transport model (REAM) (*12*). Measurements from three mountain sites over East China, Mt. Tai (117.10 E, 36.25 N, 1533 m above sea level (asl), center of East China), Mt. Hua (110.09 E, 34.49 N, 2064 m asl, west

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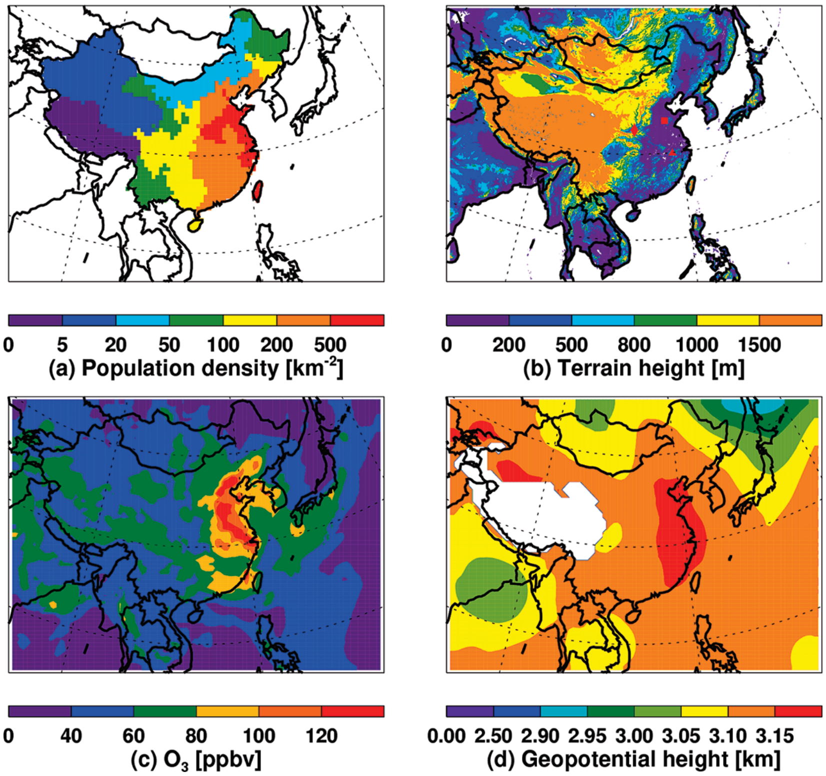

FIGURE 1. (a) Distribution of the provincial population density in China in 2000. (b) Terrain height distribution over East Asia with a 5 km resolution. The red square, diamond, and triangle symbols represent the locations of Mt. Tai, Mt. Hua, and Mt. Huang, respectively. (c) Ground-level MDA8 ozone concentrations over East Asia on June 13, 2004, from the VOC simulation. (d) Simulated 700 hPa geopotential height distribution on June 13, 2004.

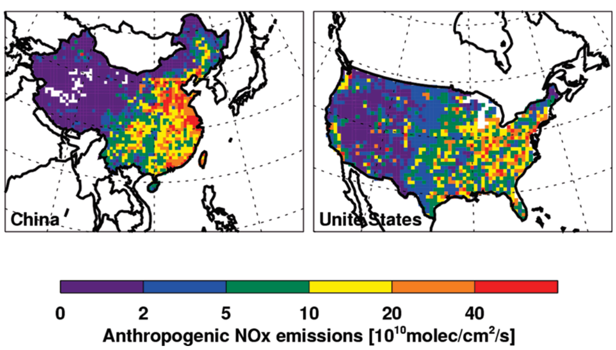

FIGURE 2. Distributions of anthropogenic NO_x emissions over China (left) and the United States (right) in 2004.

edge of East China), and Mt. Huang (118.15 E, 30.13 N, 1836 m asl, south of East China) (Figure 1b) (*8, 24*), are used to confirm that the model simulations generally capture the observed pollution episodes. A larger episode on June 9−14, 2004, is analyzed in this study. While the observation sites are limited, they do represent the regional feature of ozone over East China since they triangulate over the region and the sites at 1.5−2 km are high enough to avoid the influence of local pollutant emissions or distribution features. The model simulation shows high ozone concentrations (>80 ppbv) covering the East China plains in the June episode (Figure 1c). The meteorological condition during this episode is typical for ozone episodes, a high-pressure system controlling the region, which prevents the ventilation of ozone precursors (Figure 1d).

The ozone measurements in May at the mountain sites were previously published (*8, 24*) and were used in the model evaluation (Supporting Information). The model errors for ozone simulation are within the recommended ranges of the U.S. Environmental Protection Agency (EPA) (Supporting Information). The standard model simulation shows the general characteristics of the high-ozone episodes in May, but significantly underestimates some ozone peaks during the episodes at Mt. Tai, even though it captures the general timing of the episodes.

Ozone is produced by NO_x (NO + NO_2) in the presence of volatile organic compounds (VOCs). The emissions of NO_x over East China used in this model are constrained by satellite measurements (Supporting Information). Anthropogenic emissions of VOCs, which include a large number of species, are subject to a much higher degree of uncertainty (∼130%) over Asia (*3*). Underestimation of VOC emissions leads to underestimation of regional ozone (*25*). Our modeling analysis of observed NO, peroxyacetal nitrate (PAN), and VOCs in Beijing in August 2007 suggests that the model greatly underestimates VOCs that lead to PAN formation (unpublished results). Therefore, more VOCs are added in the model (hereafter referred to as the VOC simulation) in the form of methylglyoxal (MGLY) with the same emission distribution as that of propene over China. MGLY is produced from the oxidation of many VOCs and is an effective PAN precursor. In a previous model analysis, it was significantly underestimated over urban regions such as those in East China compared to limited in situ and satellite measurements (*26*). The scaling factor of MGLY to propene emissions is determined such that PAN simulated in the model in Beijing is in agreement with the measurements. The additional MGLY emission improves the ozone simulations (next section), but its impact on the results presented in this study is limited (Supporting Information).

The evaluation shows that the model performance at Mt. Tai is improved in VOC simulations, especially in producing the high-ozone peaks (Supporting Information). The effects at the other two mountain sites are insignificant, because ozone production in regions around these sites is sensitive to NO_x emissions whereas ozone production around Mt. Tai

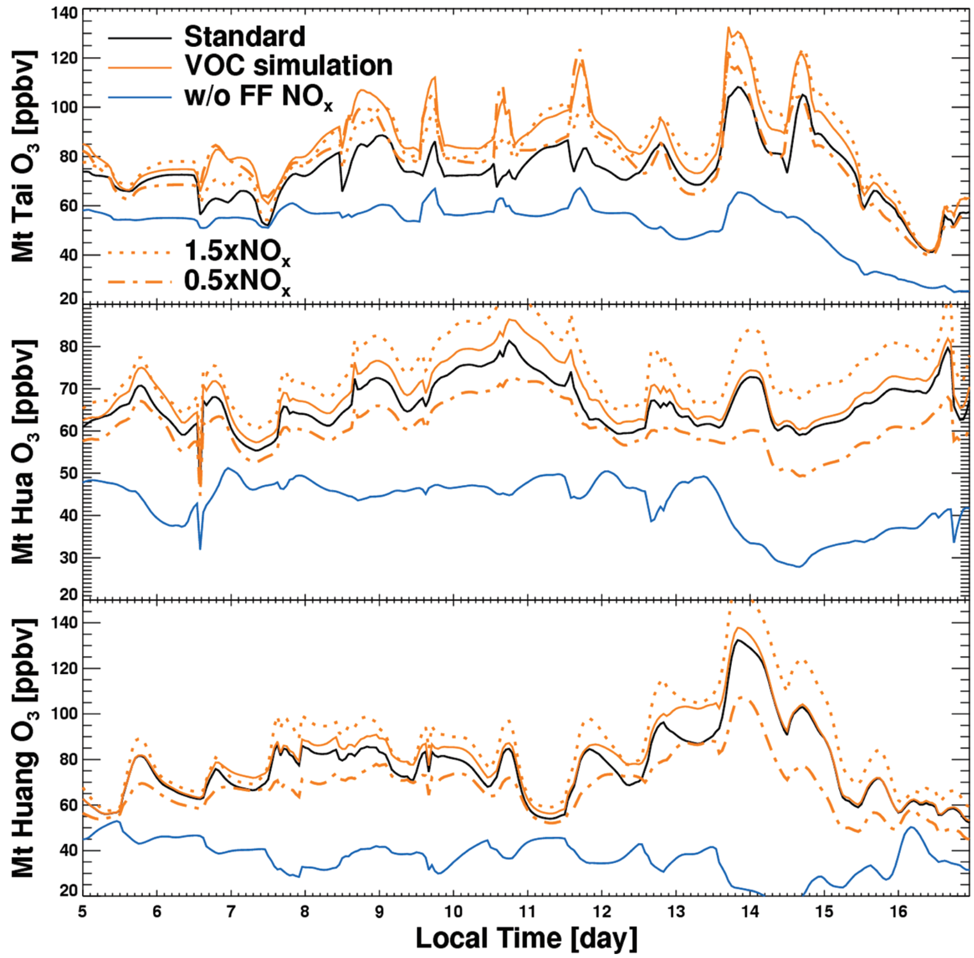

FIGURE 3. Simulated hourly O_3 concentrations on June 5−16, 2004, at three mountain sites (from top to bottom, Mt. Tai, Mt. Hua, and Mt. Huang). Five model results are shown: the standard simulation (black line), three VOC simulations (standard, solid orange line; 50% increase of Chinese NO_x emissions, dotted orange line; 50% decrease of Chinese NO_x emissions, dashed orange line), and the simulation without anthropogenic NO_x emissions over China (blue line).

is sensitive to VOC emissions (next section). Therefore, we use the modeling results from the VOC simulation in this study. In the results shown, the difference between the VOC and standard simulations is small except the intensely polluted areas such as the vicinity of Beijing (Supporting Information) where ozone production is sensitive to VOC emissions (next section). Due to the restriction of the agreement between two institutes in China and Japan, we were asked not to show the measurement data in June in this work. The model performance during the June episode is similar to that shown for May, and the simulated results are also better correlated with the measurements with correlation coefficients of 0.60, 0.54, and 0.80 for Mt. Tai, Mt. Hua, and Mt. Huang, respectively.

The simulated hourly ozone concentrations at three mountain sites on June 5−16, 2004, are shown in Figure 3. Among the three mountain sites, Mt. Tai is located in an area with a high population density (Figure 1a), and the regional emissions nearby are also higher (Figure 2). Ozone exceeded 80 ppbv from June 9 to 14, reaching up to 120 ppbv. The VOC simulation had higher ozone concentrations than in the standard simulation especially during the high-ozone episode because of ozone production sensitivity to VOCs in the surrounding area. The rapid decrease of ozone to 50 ppbv on June 15 was driven by the onset of the Asian summer monsoon (*24*), when relatively clean maritime air is transported to the region. Mt. Hua is located at the western edge of the Northeast Plain. The surrounding population and regional emissions are lower than those of the other two sites. As a result, the average ozone concentrations during the episode from June 9 to June 14 were lower than those of Mt. Tai. Mt. Huang is located to the south of the Chang Jiang Downstream Plain. The model simulation shows that the episode started in the northern plains and then migrated southward. As a consequence, ozone concentrations at Mt. Huang were significantly higher on June 12−14 than the first half of the episode, reaching a maximum of ∼140 ppbv, much higher than the summer average of ∼45 ppbv (*24*).

We also conducted another simulation, in which anthropogenic NO_x emissions over China were removed in the model. In this sensitivity simulation, O_3 concentrations mainly reflect the transport of ozone produced in the other regions of the world and from the stratosphere and are significantly lower than those of the standard simulation. This demonstrates that the high-ozone episodes are not driven by ozone transport from the regions outside China or the stratosphere but from ozone production inside China. The larger impact of anthropogenic NO_x emissions at Mt. Huang than Mt. Tai reflects in part ozone production sensitivity to NO_x emissions in areas around Mt. Huang, whereas ozone production around Mt. Tai is more sensitive to VOC emissions (next section). The local ozone production sensitivity is modulated by regional recirculation of ozone.

The high-ozone peaks (up to 140 ppbv) at midnight of June 14 at the three mountain sites were simulated as observed. These high-ozone peaks at midnight reflect the amounts of high ozone produced photochemically on June 13 in polluted regions. The spatial distribution of the maximum daily 8 h average (MDA8) ozone concentrations on June 13 over China from the VOC simulation is shown in

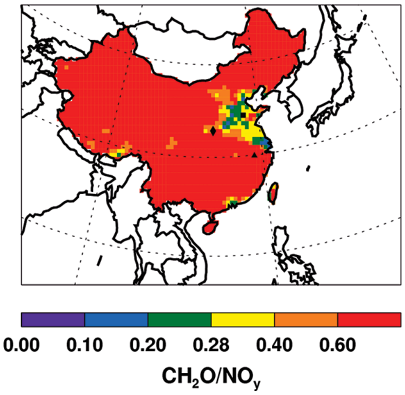

FIGURE 4. REAM-simulated mean daytime ratios of CH_2O to NO_y for June 2004 over China in the VOC simulation. The black square, diamond, and triangle symbols represent the locations of Mt. Tai, Mt. Hua, and Mt. Huang, respectively.

Figure 1c. The ozone basin feature of East China is evident. All of East China suffers from high ozone concentrations with an average MDA8 value of 93 ppbv, which normally occurs in the urban and suburban regions in the United States (*27*). This episode is fostered by the synoptic meteorological condition. A high-pressure system was located over East China from June 11 to June 13 (Figure 1d). Under the control of the stable high-pressure system which suppresses the ventilation of pollutants from the boundary layer, photochemistry produces large amounts of ozone in the polluted regions, and the meteorological condition favors the recirculation of high ozone through the connected East China plains.

Effectiveness of NO_x Emissions Control. Ozone production is driven mainly by NO_x and VOCs. Photochemically, ozone production is sensitive either to NO_x emissions or to VOC emissions. One way to diagnose the ozone production sensitivity is through the ratio of CH_2O to NO_y (*28*), where NO_y is the sum of total reactive nitrogen. The critical value determined by Sillman et al. (*28*) is 0.28. Above this value, ozone production is sensitive to NO_x emissions; otherwise, it is sensitive to VOC emissions. Figure 4 shows that the simulated CH_2O/NO_y ratios are >0.6 over most regions of East China except the intensely polluted areas such as the vicinity of Beijing, indicating ozone production over a large part of East China is sensitive to NO_x emissions. Both standard and VOC simulations show similar results (Supporting Information). The NO_x-sensitive region over China is consistent with the results of Tie et al. (*29*) in summer. Ozone at Mt. Tai located in the VOC-sensitive region is less sensitive to NO_x emissions than at the other two mountain sites located in the NO_x-sensitive region (Figure 3).

We apply the model to evaluate the effectiveness of reducing NO_x emissions in controlling regional ozone over East China. The changes of the MDA8 ozone concentrations from two sensitivity results (case 1, increasing anthropogenic NO_x emissions by 50%; case 2, decreasing anthropogenic NO_x emissions by 50%) from the VOC simulations are shown in Figure 5. Both the magnitude and the coverage of the high ozone over the East China plains on June 13 were significantly affected by the change of the NO_x emissions, since ozone production over most regions of East China is sensitive to NO_x emissions. The MDA8 ozone concentrations over East China change by +9% (an increase of 5−15 ppbv) and −16% (a decrease of 10−40 ppbv) with a 50% increase and decrease of anthropogenic NO_x emissions, respectively. More sensitive to NO_x emissions than MDA8 ozone concentrations are the areas affected by high ozone concentrations. If we use the U.S. EPA MDA8 ozone standard of 75 ppbv, the areas with high ozone concentrations correspond linearly and increase

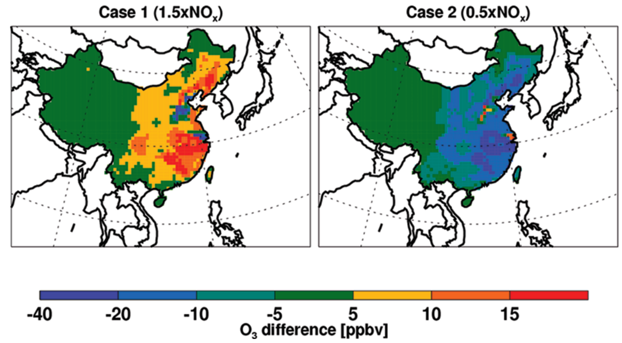

FIGURE 5. Ground-level MDA8 O_3 concentrations over East Asia on June 13, 2004, from the VOC simulations by increasing (case 1) or decreasing (case 2) the anthropogenic NO_x emissions over China by 50%.

or decrease by 50% from 1.3 million km^2 as the changes of the anthropogenic NO_x emissions in the model.

Satellite measurements indicate the NO_x emissions over East China have been increasing at an annual rate of 6−10% (*4, 30*). The estimated total fossil fuel NO_x emission in China was 35% higher than in the United States in 2004. The high-emission regions in China are more widespread than in the United States. The geographically flat East China plains coupled with high anthropogenic emissions lead to high-ozone episodes, which affect >60% of the population in China. The basin feature of the connected plains allows the recirculation of pollutants over the high-emission regions under high-pressure systems. Model sensitivities indicate that ozone concentrations are very responsive to NO_x emission reductions in terms of high-ozone areas and hence population exposure.

Acknowledgments

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Supporting Information Available

REAM model description, model performance evaluation using satellite NO_2 columns and surface O_3 measurements, and assessment of the impact of MGLY emissions on model simulations. This information is available free of charge via the Internet at http://pubs.acs.org.

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