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The effects of rapid urbanization on the levels in tropospheric nitrogen dioxide and ozone over East China

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HIGHLIGHTS

- Tropospheric columns of NO_2 increased by 82–307% over East China during 1996–2011.
- Rapid urbanization plays a critical role in the long-term changes in tropospheric NO_2.
- Meteorological factors determine the seasonal patterns of tropospheric NO_2.
- Increase in tropospheric NO_2 over Beijing after the 2008 Olympic Games was larger than before.
- Tropospheric O_3 is relatively insensitive to urbanization and changes in tropospheric NO_2.

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ABSTRACT

Over the past few decades, China has experienced a rapid increase in urbanization. The urban built-up areas (population) in Beijing, Shanghai, and Guangzhou increased by 197% (87%), 148% (65%), and 273% (25%), respectively, from 1996 to 2011. We use satellite retrieval data to quantify the effects of rapid urbanization on the yearly and seasonal changes in tropospheric nitrogen dioxide (NO_2) over East China. The results show that rapid urbanization has a profound effect on tropospheric columns of NO_2. During 1996–2011, the tropospheric columns of NO_2 over the surrounding areas of Guangzhou, Shanghai, and Beijing increased by 82%, 292%, and 307%, respectively. The tropospheric columns of NO_2 reach their maximum in winter and minimum in spring. The anthropogenic emissions related to urbanization are a dominant factor in the long-term changes in the yearly and seasonal mean tropospheric columns of NO_2, whereas meteorological conditions such as the prevailing winds and precipitation account for the unique spatial patterns. Around the time of the 2008 Beijing Olympic Games, the tropospheric columns of NO_2 over Beijing urban area significantly reduced by 48% in July, 35% in August, and 49% in September, relative to the same monthly averages over 2005–2007. However, this trend was reversed after the Games, and the increased rate was even larger than before. Our results show that the tropospheric NO_2 above the three regions increased at rates 1.3–8 times faster than the rates in a recent inventory estimate of NO_x emissions for 2000–2010. We also discuss the influence of urbanization on tropospheric ozone and find that the Ozone Monitoring Instrument (OMI) retrieval tropospheric column shows that ozone levels are relatively insensitive to urbanization and changes in tropospheric NO_2.

1. Introduction

The dramatic economic development that China has experienced since the mid-1980s has been accompanied by a rapid increase in urbanization. The combined pace of economic growth and

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urbanization has led to tremendous increases in energy consumption and put millions of cars on the roads. As a consequence, many cities especially the megacities (with populations over 10 million), now suffer from severe air pollution. In Beijing, for instance, the China National Ambient Air Quality Standard Grade II for ozone (hourly average value of 100 ppb) was exceeded on 57 days during 2005, with a maximum hourly average concentration of 212 ppb (Hao and Wang, 2005; Fu et al., 2009), and the ground-level ozone concentrations have risen six-fold in the past three decades; and the daily average concentrations of $PM_{2.5}$ (airborne particles with diameter less than or equal to 2.5 μm) typically range from 100 to 150 $\mu g\ m^{-3}$, which is much higher than the US National Ambient Air Quality Standard value of 35 $\mu g\ m^{-3}$ (Qiu, 2012). The urban population in China has increased from 26% of the total population in 1990 to 47% in 2010 (UN ESCAP, 2011), and is expected to rise to nearly 70% by 2050 (Qiu, 2012). Therefore, an accurate assessment of the effects of urbanization on the atmospheric environment is essential for ensuring efficient energy use and air pollution mitigation.

Nitrogen oxides (NO_x, a mixture of NO and NO_2) are key substances contributing to the formation of ozone and aerosols (e.g., $PM_{2.5}$) in the troposphere. Changes in tropospheric NO_x have important effects on air quality, acid deposition, and the balance of atmospheric radiation (Richter et al., 2005). NO_x are emitted from soil and as a result of fossil fuel combustion, biomass burning, and lightning (Richter et al., 2005; Lin, 2012). While NO_x levels have been decreasing in many industrialized countries, the recent rapid industrialization and urbanization in East Asia has resulted in significant increases in tropospheric nitrogen dioxide (NO_2) (Richter et al., 2005). Emission inventories show that China is the dominant source of anthropogenic emissions of NO_x, sulfur dioxide (SO_2), and carbon monoxide (CO) in Asia (Streets et al., 2003). NO_x emissions in China increased by 280% from 1980 to 2003, and were predicted to undergo another stage of rapid increase from 2003 onwards (Ohara et al., 2007). Nevertheless, urbanization is associated with increased energy efficiency, and the central government of China has set a NO_x emission reduction goal of 10% for the 12th 5-Year (2011–2015) implementation period (Wu et al., 2012). Therefore, an update on the changes in tropospheric NO_2 (a proxy of NO_x) at a regional level will provide an independent assessment of the inventory data and an objective means of evaluating the effectiveness of the government's emission control strategies in relation to the ongoing urbanization process.

Satellite retrieval data provide a feasible basis for revealing spatial and temporal variations in tropospheric columns of NO_2, and evaluating the effectiveness of emissions control measures (e.g., Richter et al., 2005; Witte et al., 2009). This is especially true for regions where there are limited surface observational data. Tropospheric columns of NO_2 ($TCNO_2$) can be retrieved from the ultraviolet/visible measurements observed by satellite instruments, such as the Global Ozone Monitoring Experiment (GOME) (Burrows et al., 1999) and the SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY (SCIAMACHY; Bovensmann et al., 1999). Richter et al. (2005) analyzed the retrieval data from the GOME and SCIAMACHY measurements and found decreasing trends in $TCNO_2$ over North America and Western Europe and increasing trends with accelerating growth rates over China from 1996 to 2002. Their findings are broadly consistent with bottom-up inventory studies (e.g., Zhang et al., 2007) showing a continuous and accelerating growth rate during 1996–2004 over East Central China. Witte et al. (2009) utilized the same satellite measurements and found a 43% reduction in the tropospheric columns of NO_2 over Beijing during July–September 2008 compared to the same months during 2005–2007. Wang et al. (2009b) analyzed the surface observations for several air pollutants at a rural site downwind of Beijing, and found that the mean daytime levels of O_3, SO_2, CO, and NO_y (NO_x plus other oxidized species such as N_2O, N_2O_5 and HNO_3) in August 2008 were 23%, 61%, 25%, and 21% lower, respectively, compared to the same months in 2006–2007. They attributed this change mainly to the strict short-term emissions control measures enforced in Beijing and neighboring provinces to improving air quality during the Beijing Olympic Games in August–September 2008. It is not known whether the reduction in tropospheric NO_2 had lasting effects after the Beijing Olympic Games.

Recently, Schneider and van der A (2012) used global nine-year (2002–2011) SCIAMACHY-derived NO_2 data to compare the trends in tropospheric NO_2 among the world's major megacities and found that Dhaka in Bangladesh had the largest relative increase. They also found a significantly decreasing trend in Europe and strong increasing trends in China and other countries in Asia. Nonetheless, it is still not clear how urbanization quantitatively influences the concentrations of tropospheric NO_2 and ozone.

In this study, we utilize the 1996–2011 retrieval data from both GOME and SCIAMACHY measurements to quantify the relationship between urbanization and changes in the tropospheric columns of NO_2 over East China. The existing related studies are limited to the annual average changes in tropospheric NO_2, and studies on seasonal mean changes and the influence of urbanization are relatively scarce. In this study, we characterize the temporal and spatial variations in the tropospheric columns of NO_2 in three regions of China that have undergone rapid economic growth and examine the factors that determine the temporal variation and spatial patterning of NO_2. We then assess the relationship between the anthropogenic emissions associated with urbanization and the changes in the levels of tropospheric ozone over East China.

2. Data and methodology

Monthly mean tropospheric excess columns of NO_2 ($TECNO_2$) derived from the GOME and SCIAMACHY satellite measurements (Richter et al., 2005) are used to quantify the changes in tropospheric NO_2. The gridded datasets used in this study were provided by the Institute of Environmental Physics (IEP), University of Bremen, Germany (data available at http://www.iup.uni-bremen.de/doas). $TECNO_2$ is the surplus of tropospheric column NO_2 in an area relative to that in a clean region over oceans at the same latitude. GOME was launched in April 1995 and provided global coverage from August 1995 to June 2003. SCIAMACHY was launched in March 2002 and took measurements from August 2002 to April 2012. Grid spacings of $0.5^\circ \times 0.5^\circ$ and $0.125^\circ \times 0.125^\circ$ are used for the GOME and SCIAMACHY data, respectively. The two datasets can be combined to investigate the long-term changes in tropospheric NO_2 as the time series of the two retrieval data fit almost seamlessly despite the different instruments and data resolution (Richter et al., 2005). A detailed evaluation of the resolution effects is given in Hilboll et al. (2013). The $TECNO_2$ products used in this study have been validated (Petritoli et al., 2004) and successfully applied to evaluating changes in tropospheric NO_2 in North America, Europe, and Asia (Richter et al., 2005; Wang et al., 2009a).

Tropospheric ozone (O_3) data retrieved from the Ozone Monitoring Instrument (OMI) are used to assess the influence of changes in NO_2 on that of O_3 in the troposphere. OMI is a nadir-viewing imaging spectrograph that measures the back-scattered solar radiation from the Earth's atmosphere and the surface in the ultraviolet and visible bands (Levelt et al., 2006). Launched in July 2004, the OMI monitors the total column ozone, ozone profiles, other trace gases (e.g., NO_2, HCHO, SO_2), and UV-absorbing aerosols and clouds (Tanskanen et al., 2007). The tropospheric column O_3 levels used in this study are monthly mean data with a spatial resolution of $1.25^\circ \times 1.0^\circ$ in the W–E and S–N directions, respectively (Ziemke et al., 2006) (data available at http://acd-ext.gsfc.nasa.gov/Data_services/cloud_slice/new_data.html).