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Temporal and spatial distribution of tropospheric NO_2 over Northeast Asia using OMI data during the years 2005–2010

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ABSTRACT

This study aimed to examine the main characteristics of tropospheric nitrogen dioxide (NO_2) concentrations over the Northeast Asia, using the Ozone Monitoring Instrument (OMI) data from 2005 to 2010. The annual mean NO_2 concentrations (AMNC) had an increasing trend mainly due to increasing NO_2 emissions in China except during the 2008 Beijing Olympic Games period, while the reduction policies of South Korea and Japan have led it to be stagnant or decreased. To investigate further regional characteristics of NO_2 increasing trends in China, we divided our study area into 6 geographical regions (sectors 1–6) and then considering 4 different socio–economic levels (groups 1–4) among main cities in Eastern regions (sector 2 and 4) where the concentrations level is the highest in China and NO_2 concentrations show continued increasing trend. Especially OMI NO_2 and emissions consistently showed that metropolitan/big–sized and developed cities (group 1), such as Beijing and Shanghai, had an increasing trend of NO_2 concentrations until 2007 and decreasing thereafter, while small/mid–sized and developing cities (groups 2 and 3) kept a continuous increasing trend over all periods. The seasonal change in NO_2 concentrations showed the apparent increasing trend in winter and no significant trend in summer in all groups except for group 1. These results indicate that an increase in AMNC in Northeast Asia was mainly attributed to the increasing NO_2 concentrations in winter in groups 2 and 3. Therefore, it strongly suggests the importance of the NO_2 management for groups 2 and 3 to improve air quality in the Northeast Asia.

Keywords: *NO_2, OMI, satellite, Northeast Asia, China*

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1. Introduction

Nitrogen dioxide (NO_2) is a major gas that affects the atmospheric environment and indirectly causes climate change. It adversely impacts human health, causing ozone and particulate matter (PM) in the troposphere, along with acid rain and photochemical smog (Vidot et al., 2010; Wang et al., 2011; Geddes et al., 2012; Bechle et al., 2013; Zyrichidou et al., 2013). Besides, in the context of climate change, it reduces the lifetime of methane in the atmosphere, leading to negative radiative forcing (IPCC, 2007). Therefore, investigating the concentrations, the changes and emission sources of NO_2 has become one of the most important environmental issues. NO_2 concentrations in the troposphere show high correlation with emission quantity in source area as it is mainly affected locally rather than by long–range transport because of short lifetime with just 1–2 days (Cheng et al., 2012). NO_2 plays an important role in the chemical process of the major chemicals in the troposphere and also affects the formation of ground–level ozone by involving the photochemical oxidation reactions with CH_4 and CO (Aneja et al., 1996). The dominant sink of NO_2 in the troposphere is its conversion into nitric acid (HNO_3) and peroxyacetylnitrate (PAN), which are eventually removed by dry or wet deposition (Browne et al., 2013).

Major sources of NO_2 include anthropogenic sources, such as stationary sources (industrial facilities), mobile sources (vehicles, ships, airplanes), and small–scale sources (heating facilities, kitchens) and natural sources, such as lightning, volcanic eruption, and bacteria (Sheel et al., 2010). All over the world, various studies have been conducted in the form of surface observations, aircraft measurements, and modeling to assess NO_2 concentrations and identify emission sources (Grice et al., 2009; Anttila et al., 2011; Shon et al., 2011; Xing et al., 2011; Li et al., 2012; Tian et al., 2013). However, surface observations and aircraft measurements have space constraints that could not be identified at the same time in the wide open area, although providing reliable data (David and Nair, 2013). Modeling study also has a disadvantage to be verified by observation, although it can cover an extensive area. Meanwhile, observation of NO_2 concentrations using satellite not only overcomes space constraints, but also can produce data in a stable way.

Developed countries like the U.S. and Europe, have actively used satellites to observe NO_2 concentrations. It was suggested that satellites are reliable through comparing the NO_2 data from OMI with surface observations that displayed high correlation with California (r=0.93) and Toronto (r=0.86) (Geddes et al., 2012; Bechle et al., 2013). Additionally, Zyrichidou et al. (2013) showed that the monthly average NO_2 concentrations of the metropolitan area ($2.0–5.7\pm1.1\times10^{15}$ molecules/cm^2) was higher than that of rural regions ($1.1–2.2\pm0.4\times10^{15}$ molecules/cm^2) in Southeastern Europe. Zhou et al. (2012) pointed out the decreasing NO_2 concentrations in Western Europe from 2004 to 2009 and Ghude et al. (2009) also showed the decrease in NO_2 concentrations in the Eastern U.S. (–2±1.5% 1/year) and Europe (0.9±2.1% 1/year) from 1996 to 2006. The NO_2 concentrations in the atmosphere are increasing in the Asia responding the population growth and rapid industrialization in this region (Kunhikrishnan et al., 2006; Lal et al., 2012; Meena et al., 2013; ul–Haq et al., 2014). He et al. (2007) found a continuous increase of NO_2 concentrations during the past decade with a sharp linear increase rate of 14.1–20.5% per year during 2000–2005 in East Asia using Global Ozone Monitoring Experiment (GOME) and SCanning Imaging Absorption spectro-Meter for Atmospheric CHartographY (SCIAMACHY) observations. NO_X emission from combustion of stationary sources and vehicles has significantly increased in Northeast Asia (Ohara et al., 2007). Richter et al. (2005) also showed that the largest increase in NO_2 concentrations over the industrialized part of China took place from 1996 to 2002. In Beijing, the capital of China, the number of vehicles went up from 0.82 million in 1994 to 3.5 million in 2008 (Sun et al., 2011) and it was also reported that NO_2 increased by 50% in the industrial regions in China from 1996 to 2004 (IPCC, 2013). These reports, however, did not clearly show annual changes in NO_2 concentrations in Northeast Asia. In this study, we examined the temporal and spatial distribution of annual and monthly mean NO_2 concentrations using OMI data. Then, we further assessed the annual and monthly mean NO_2 concentrations by comparing with the emissions inventory in Northeast Asia from 2005 to 2010.

2. Materials and Methods

2.1. The OMI observations

OMI is loaded with the EOS–Aura satellite launched in July 2004 and is capable of resolving a 0.5–nm spectrum, while observing 270–nm to 500–nm ultraviolet and visible ray in nadir view mode (Levelt et al., 2006). UV rays channel consists of two subchannels (UV–1 and UV–2) and their wave spectrum is 270–310 nm and 310–365 nm, while their average spectral resolution is 0.42 nm and 0.45 nm, respectively. The visible channel covers 365–500 nm and the average spectral resolution of 0.63 nm. In addition, the OMI sensor has a wide field of view of 114° and is able to observe the entire global sphere in a day with a swath of 2 600 km, passing the equator at 13:45 local time. The spatial resolution of OMI is 13×24 km^2, enabling it to observe trace gases, such as NO_2, SO_2, O_3, OClO, and BrO.

In order to observe NO_2 from the OMI, Differential Optical Absorption Spectroscopy (DOAS) is deployed to estimate Slant Column Density (SCD), which can be obtained in Vertical Column Density (VCD) using Air Mass Factor (AMF) calculated by a radiative transfer model (Han et al., 2011). For detailed information on NO_2 algorithm, see Boersma et al. (2007) and the DOMINO (Dutch of OMI tropospheric NO_2) Product Specification Document (http://www.temis.nl/docs/OMI_NO2_HE5_1.0.2.pdf). The retrieval of tropospheric column NO_2 takes into account the clear and cloudy conditions in the airmass factor derived for the simulated NO_2 profiles (Bucsela et al., 2006). Tropospheric NO_2 has an uncertainty of 0.1×10^{15} molecules/cm^2 and it is underestimated by 15–30% (Celarier et al., 2008).

The monthly mean tropospheric NO_2 column data (Level 3) from OMI were analyzed from January 2005 to December 2010 considering available periods for OMI data and emission inventories. Level 3 is the monthly DOMINO ver. 2.0 collection 3, with a cloud radiance fraction <50%, corresponding to cloud fractions approximately <20%, with a 0.125°×0.125° grid resolution, available from the TEMIS website (http://www.temis.nl/airpollution/no2col/no2regioomimonth_col3.php; Boersma et al., 2011) for KNMI–OMI.

2.2. Methods

In this study, as Figure 1 indicates, changes in tropospheric NO_2 over the Northeast Asian regions were observed (north latitude 20–45° and east longitude 105–145°). The spatial and temporal distribution of NO_2 concentrations from 2005 to 2010 was analyzed by each region, sector, and group as follows.

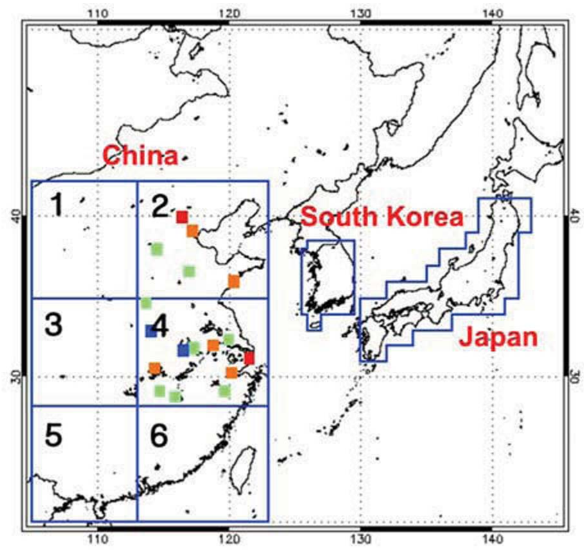

Figure 1. *Target domain in this study. The blue line indicates the areas by region for China, South Korea, and Japan. China is divided into 6 sectors and 4 groups (red box is group 1, orange box is group 2, light green box is group 3 and blue box is group 4).*

First, the region was divided into three parts; (a) the whole Northeast Asia (all areas), (b) China, Japan, and South Korea (China + South Korea + Japan; CKJ) and (c) background area (the rest of the area except for CKJ). To investigate further regional characteristics of NO_2 increasing trends in China, we divided it into 6 geographical regions (sectors 1–6). Because the area of China is much bigger than other countries, there is a huge difference in NO_2 levels by each sector. To investigate the characteristics by each city in sectors 2 and 4, China was divided again into 4 groups (Qiu and Liu, 2011) depending on gross domestic product (GDP) (GDP is the income level of the city) and population (Table 1). Cities in group 1 is saturated because of the slow economic and population growth, while cities in groups 2 and 3 still rapidly grow with steady population growth. Cities in group 4 are at the early stage of urbanization and economic growth with very little movement on their population.

Table 1. *Classification of cities by population size and GDP*

GDP per capita (Unit: Dollar)	Population Size (Unit: Million)				
	Metropolis (>10)	Big City (5–10)	Middle City (1.5–5)	Small City (0.5–1.5)	Rural (<0.5)
>10 000	a	a	b	c	c
5 000–10 000	a	b	c	c	d
3 000–5 000	b	c	c	d	d
1 000–3 000	c	c	d	d	d
<1 000	c	d	d	d	d

[a] *Beijing, Shanghai*
[b] *Tianjin, Nanging, Qingdao, Hangzhou, Wuhan*
[c] *Jinan, Shijiazhuang, Hefei, Zhengzhou, Taizhou, Jinhua, Changde, Nanchang*
[d] *Luan, Zhuma dian*

3. Results

3.1. NO_2 concentrations over Northeast Asia

Figure 2 shows a time series of the annual mean NO_2 concentrations (AMNC) in troposphere over Northeast Asia. In the whole Northeast Asian regions (all areas), the AMNC showed an increase of 0.11×10^{15} molecules/cm^2 per year with 2.05–2.61×10^{15} molecules/cm^2. In China, South Korea, and Japan (CKJ