Turnaround of Tropospheric Nitrogen Dioxide Pollution Trends in China, Japan, and South Korea

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Abstract

Long-term (2005–2015) tropospheric nitrogen dioxide (NO₂) column data recorded by the satellite-borne Ozone Monitoring Instrument (OMI) in East Asia were analyzed to investigate annual trends on a national basis and their potential causes. We found an evident decrease of 6% year⁻¹ in the NO₂ level over China after 2011. The grid-basis trend analysis implies that the rapid decrease occurred on a provincial or larger spatial scale and was likely due to a nationwide action such as the widespread use of denitrification units. In Japan and South Korea, a turnaround indicating an increase was observed after 2013 and 2012, respectively. As a consequence, the tropospheric NO₂ pollution level in East Asia was found to be recovered to the 5-year-ago level in 2015.

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1. Introduction

Nitrogen oxides (NO_x = NO + NO₂) are released into the atmosphere from both anthropogenic (e.g., fossil fuel combustion) and natural sources (e.g., microbiological processes in soil and lightning) and play crucial roles in tropospheric chemistry, with important implications for air quality and climate change. In the troposphere, NO_x is a critical precursor for ozone, leading to a shorter-lived (and hence localized) positive radiative forcing. In addition, NO_x leads to a longer-lived (and hence more global) negative radiative forcing due to the increased rate of destruction of important greenhouse gases such as methane through feedback on hydroxyl radicals (OH) (Seinfeld and Pandis 2006). Furthermore, NO_x contributes to the secondary aerosol formation, the mechanism of which has been poorly understood. At high concentrations, nitrogen dioxide (NO₂) itself can contribute directly to radiative forcing (Solomon et al. 1999).

Using long-term satellite measurements of tropospheric NO₂ columns by the Global Ozone Monitoring Experiment (GOME) (Burrows et al. 1999) and the SCanning Imaging Absorption spectroMeter for Atmospheric CartograpHY (SCIAMACHY) (Bovensmann et al. 1999), a tripling of the tropospheric NO₂ column was detected over Central Eastern China (CEC; 30°N–40°N and 110°E–123°E) from 1996 to 2011, due to an increase in emissions from industrial and traffic sectors (Hilboll et al. 2013). During the same time period, NO₂ levels in developed countries, including Japan, displayed significant decreasing trends. More recently, Krotkov et al. (2015) and Duncan et al. (2016) analyzed high-spatial-resolution satellite data from the Ozone Monitoring Instrument (OMI) (Levelt et al. 2006) and showed that the NO₂ pollution levels peaked in 2011 over a certain rectangular area situated in an urban environment in China. However, they have not discussed detailed nationwide attribution to the observed trends

for years, including the last year 2015.

In 2011, two events occurred that could represent a turning point for the trend in NO₂ levels in East Asia. One was the establishment of the 12th Five-Year Plan in China (2011–2015) (Tian et al. 2013; Zhao et al. 2013), which aimed to reduce pollution, including NO_x. Even in the latest study for emission inventory during 1990–2010 (Liu et al. 2015), the installed denitrification devices for coal-fired power plants were discussed and assumed not in operation until 2010. The effect of the installed denitrification devices should be confirmed by measurements. The other was a significant substitution in power generation in Japan from nuclear to thermal power plants, as a consequence of the serious damage to the Fukushima Daiichi nuclear power plant by the tsunami generated due to a massive earthquake off the Pacific coast of northern Japan on 11 March 2011 (Takemura et al. 2011). Therefore, this study utilizes the most recent satellite data (until 2015) to investigate how trends in NO₂ levels varied over East Asia focusing on the periods before and after 2011 and make a nationwide discussion about most plausible causes, while proper emission inventory is unavailable.

2. OMI data and its long-term quality check with MAX-DOAS

The OMI, an ultraviolet/visible (270–500 nm) hyperspectral imaging spectrometer, is one of the instruments onboard NASA's Aura satellite (an equator crossing time of around 13:45 LT) launched on 15 July 2004. Its nominal nadir spatial resolution is 13 × 24 km². The present study analyzed tropospheric NO₂ vertical column density (VCD) products retrieved from the algorithm developed by the Royal Netherlands Meteorological Institute (Dutch OMI NO₂; DOMINO version 2) (Boersma et al. 2011) under cloud-free conditions (i.e., a cloud fraction less than 0.2). Referring to Boersma et al. (2011), data affected by so-called row anomalies and flagged as covered by snow/ice were excluded. According to Boersma et al. (2011), the overall uncertainty for individual retrievals is 1.0 × 10¹⁵ molecules cm⁻² +25% of VCD. It does not affect the conclusions drawn from the present study, because relative changes using data taken for more than one year are discussed.

In this study, a long-term data quality check for OMI data over Japan was performed through long-term validation comparisons between tropospheric NO₂ VCDs derived from OMI and ground-based Multi Axis Differential Optical Absorption Spectroscopy (MAX-DOAS) remote sensing observations at Tsukuba, Japan (36.1°N, 140.1°E) (Irie et al. 2008, 2009, 2012). The operating period of MAX-DOAS (2006–2015) well covers the long-term OMI observations. The Japanese MAX-DOAS profile retrieval algorithm version 2 (JM2) (Irie et al. 2011, 2015), based on the optimal estimation method to solve the nonlinear inversion problem, was applied. Typical uncertainty in MAX-DOAS tropospheric NO₂ VCD data was estimated to be ~15%.

We analyzed the OMI versus MAX-DOAS NO₂ VCD linear correlations and found that they were almost identical for both periods 2006–2011 (slope = 0.50, intercept = 2.8 × 10¹⁵ molecules cm⁻², and determination coefficient R^2 = 0.58) and 2012–2015 (slope = 0.53, intercept = 2.9 × 10¹⁵ molecules cm⁻², and R^2 = 0.52)

Fig. 1. Correlations between the tropospheric NO₂ VCDs measured by OMI and MAX-DOAS at Tsukuba, Japan for 2006–2011 (red) and 2012–2015 (blue). The red and blue lines are linear regression results for respective periods. The 1:1 relationship is shown with the dashed line. Error bars represent error ranges for OMI and MAX-DOAS data.

and were similar to those reported by previous validation studies (Irie et al. 2012) (Fig. 1). These results indicate that OMI data quality remained unchanged before and after 2011.

3. Trend analyses

The general spatial distributions of NO₂, in terms of average tropospheric NO₂ VCDs as measured by OMI over East Asia, for the selected years 2005, 2011, and 2015 are shown in Fig. 2. In contrast to reported trend analyses (e.g., Richter et al. 2005; Hilboll et al. 2013; Krotkov et al. 2015; Duncan et al. 2016), this study estimated tropospheric NO₂ VCDs averaged over the entire national area to enable nationwide discussion in a consistent manner. A country code map created from national boundary data (<http://hydro.iis.u-tokyo.ac.jp/GW/basemap/>) was used to judge the country corresponding to individual OMI data. The number of OMI data used for this analysis varied depending on the year and month, due to the effect by row anomalies and the effect by cloud and snow/ice coverages, respectively. To minimize the contribution of these effects to estimates of national-mean VCDs, the OMI data were first averaged on a 1° × 1° grid for each month and then the gridded values were averaged over the entire national area. We confirmed that the entire national areas of China, Japan, and South Korea were well covered by the OMI.

Over China, the annual-mean tropospheric NO₂ VCD, obtained from averages of the monthly means, increased until 2011 and peaked at 3.0×10^{15} molecules cm⁻². By contrast, from 2011 to 2015, the increase slowed, and then was followed by a decreasing trend of $\sim 6\%$ year⁻¹, which is the mean rate of change estimated for 2011–2015. The time period corresponds to the implementation of China's 12th Five-Year Plan. The level recorded in 2015 (2.3×10^{15} molecules cm⁻²) was found to be almost the same as the 2009 level. From 2005 to 2013, a continuous decrease of $\sim 5\%$ year⁻¹ occurred in Japan, whereas almost no systematic tendency ($\sim 0.2\%$ year⁻¹) was detected in South Korea. An emission inventory study indicates that the reduction of NO_x emissions in Japan and South Korea started in 2005 as road transport and power plant emissions fell, respectively (Kurokawa et al. 2013).

Fig. 2. Annual mean tropospheric NO₂ VCDs measured by OMI for 2005 (top), 2011 (middle), and 2015 (bottom). Values are plotted for each 0.25° × 0.25° grid.

The tropospheric NO₂ VCD averaged over the whole of China (2.3×10^{15} molecules cm⁻² for 2015) is similar to that for Japan (2.6×10^{15} molecules cm⁻² for 2015) but much smaller than that for South Korea (9.8×10^{15} molecules cm⁻² for 2015), because only a limited area of the entire national area (e.g., CEC and its surroundings) emitted significant amounts of NO_x in China. The national-mean tropospheric NO₂ VCD for South Korea was greater than that for Japan and China by as much as a factor of ~ 4 as a result of a larger fraction of the entire national area emitting significant amounts of NO_x in South Korea than in China and Japan.

For a more reasonable comparison among countries with respect to national emissions, the tropospheric NO₂ burden (B) was estimated as:

$$B = \text{VCD} \cdot M \cdot S / A, \quad (1)$$

where VCD is the national-mean tropospheric NO₂ vertical column density derived from the OMI observations, M is the molecular weight of NO₂, S is the national area, and A is the Avogadro's number. The S values used here are 959.8, 37.8, and 9.9×10^4 km² for China, Japan, and South Korea, respectively. B values over China were larger than those over Japan and South Korea by a factor of more than 15 throughout the period of 2005–2015 (Fig. 3 and Table 1), reflecting prominent NO_x emissions from China. The B value over China for 2015 was estimated at 17.1 kt, which is below the value for 2010. Similarly, the sum of B values for China, Japan, and South Korea in 2015 dropped below the 2010 level.

Under the assumption that B is proportional to surface NO_x

Fig. 3. Temporal variations in tropospheric NO_2 burdens over the entire national areas of China (red), Japan (green), and South Korea (blue). The burdens were estimated by multiplying the annual averages of tropospheric NO_2 VCDs over the entire national area by the area of each respective country. The burdens estimated using the monthly averages are shown in gray. Error bars represent 1σ standard deviations calculated from monthly averages.

Table 1. Estimated Tropospheric NO_2 Burdens (B in kt) and Those Divided by Populations (B/P in g person^{-1}) in Parentheses

	2005	2010	2011	2015
China	14.4 (11.0)	18.8 (14.1)	21.7 (16.1)	17.1 (12.4)
Japan	0.9 (7.4)	0.7 (5.7)	0.7 (5.6)	0.8 (5.9)
S. Korea	0.6 (13.1)	0.6 (12.9)	0.7 (14.1)	0.7 (14.6)
Sum	15.9 (10.8)	20.2 (13.3)	23.1 (15.1)	18.6 (12.0)

emissions, the B divided by the population (P) of the respective country can be regarded as a qualitative measure of emissions per person, and is therefore of interest. This is likely valid, because satellite NO_2 observations are closely correlated to surface NO_x emissions due to the short lifetime of NO_x in the troposphere. Anthropogenic NO_x emissions and the tropospheric NO_2 VCD simulated by an air quality model for the CEC region, where the anthropogenic NO_x emission was the dominant source of NO_x , were strongly correlated, with a R^2 higher than 0.9 (Itahashi et al. 2014). The annual population data used here were taken from United Nations (2015). Interestingly, the B/P values for the three countries range within a factor of only 4 throughout the period (Table 1), because the ratios of populations among the countries are of the same order of magnitude as the ratios of the satellite-derived NO_2 burdens. In other words, population can be one of the primary factors in determining B values and hence NO_x emissions. The range of B/P values is considered to be due mainly to differences in regulation, control technology, and the economic situation, while NO_x emissions are strongly related to population, regulation, control technology, and the economic situation.

It should be noted that rigorous comparisons between B/P values for different countries should be made with caution, because the NO_x lifetime could differ at different latitudes. For individual countries, however, comparisons between B/P values for different years can be interpreted with reduced effect of lifetime, as solar illumination conditions should have been almost un-

changed. The B/P values for China showed that a reduction started in 2011. However, when effects by regulation, control technology, and economic situation become insignificant, a future increase in China's population could potentially lead to an increase in NO_x emissions, as the population can be one of the primary factors in determining NO_x emissions.

To determine the potential causes of the trends revealed by the national-basis analyses, a grid-basis linear regression analysis was conducted for each $0.50 \times 0.50^\circ$ grid (Fig. 4). Linear regression was made using data containing in-between years. To ensure that sufficient grid cell coverage was achieved, the gridding was made for different grid sizes (e.g., 0.10, 0.25, 0.50, 1.00°). From this test, we found that grid sizes smaller than 0.50° were inappropriate as the trend distributions showed significant noise. From 2005 to 2011, increases in NO_2 VCDs occurred over almost all of CEC. At the same time, distinct reductions were seen around Hong Kong. In addition, reductions occurred in the Pacific Belt Zone over metropolitan areas between Tokyo and Fukuoka.

In contrast, from 2011 to 2015, most Chinese areas that had experienced an increase before 2011 exhibited a decreasing trend (Fig. 4). This was an unexpected result, particularly considering the fact that the population increased by about 2% from 2011 to 2015 (United Nations 2015). It should be noted here that the increasing tendency still appeared in Sichuan province (around Chendu), Shanxi province (around Taiyuan), Liaoning province (around Shengyang), and Inner Mongolia (Fig. 4), all of which are out of CEC or NCP (North China Plain) regions usually investigated for reported trend analyses. Thus, the turnaround in 2011, when the increasing trend ended and the decreasing trend started, occurred on a provincial or multi-provincial scale in China. This is one of the most important results of this study as it offers a unique observational hint to understand the causes of the observed trends, while emission inventory data for the most recent years were unavailable.

The Ministry of Environmental Protection (MEP) of China reported that the total load of NO_x was reduced by $\sim 6\%$ due to the widespread use of denitrification units in the first 6 months of 2014 compared with the 2013 level, laying a solid foundation for achieving the pollution reduction targets set by the 12th Five-Year Plan. For this period, the reported reduction in the total load of NO_x was comparable to the OMI-observed reduction in the NO_2 burden, suggesting that the observed decreasing trend was at least partly due to the widespread use of denitrification units in China. This nationwide action is consistent with our finding that the turnaround in trends occurred on a provincial or multi-provincial scale. The maximum NO_2 observed in 2011 may suggest that the denitrification devices have been effectively in operation since 2011, while the installed denitrification devices might not be in operation until 2010 (Liu et al. 2015).

Other underlying reasons for the observed reduction were the installation of low- NO_x burners, especially in new large power plants after 2005 (Zhang et al. 2007), and the implementation of new vehicle emission standards (Zhang et al. 2009). The National Western Development Strategies (the "Go West" movement) implemented in 1999 may have been effective because a significant NO_2 increase ($\sim 9\% \text{ year}^{-1}$ on average) was reported over Western China from 2005 to 2013 (Cui et al. 2015). The reduction in the economic growth rate in China may have made an additional contribution, because the economic situation was suggested to be a factor influencing emissions just after the summer of 2008 when emissions from power plants and industry decreased due to the global recession (Kurokawa et al. 2013). However, we note here that the observed reduction in NO_2 levels was likely much larger than the recent slowdown in the China's Gross Domestic Product (GDP) growth rate. Rapid reductions in NO_2 under the conditions of a growing Chinese economy and population might reflect the reduced coal demand, with increased diversification of power sectors such as hydro, wind, solar, and nuclear power sectors (IEA 2015).

In Japan, decreases in NO_2 levels were observed from 2005 to 2013 (Figs. 3 and 4). Larger decreases tended to occur in the Pacific Belt Zone over metropolitan areas between Tokyo and

Fig. 4. Absolute (left) and relative (right) annual changes in tropospheric NO₂ VCDs measured by OMI from 2005 to 2011 (top) and from 2011 to 2015 (bottom). Inset maps display changes over South Korea from 2012 to 2015 and over Japan from 2013 to 2015. Values are plotted for each $0.50 \times 0.50^\circ$ grid. Insignificant changes smaller than $\pm 1 \times 10^{14}$ molecules $\text{cm}^{-2} \text{year}^{-1}$ are omitted for clarity.

Fukuoka (Fig. 4). From 2013 to 2015, however, the NO₂ level increased at $\sim 13\% \text{ year}^{-1}$. In the period, since the implementation of stringent vehicle emission standards should have been effective in reducing NO_x emissions, it is likely that the increase was due to elevated NO_x emissions from the power plant sector as the significant substitution of thermal power generation for nuclear power occurred after 2011. Indeed, such a power substitution appeared to have a greater impact on NO₂ trends in rural regions than in urban regions (e.g., Tokyo), where the road transport sector is the dominant NO_x emission source (Fig. 4). More detailed attribution analysis on finer spatial scales was difficult, because the spatial resolution of OMI and statistics on such fine spatial scales were insufficient.

In South Korea, OMI NO₂ data showed an increasing trend from 2012 to 2015 (Figs. 3 and 4). Although the trend is not very significant, the NO₂ level recorded in 2015 was the highest in 11 years of OMI observations (Fig. 3). No significant positive correlation between NO₂ levels in China and South Korea was observed, indicating that changes in annual mean NO₂ levels primarily correspond to the change in domestic NO_x emissions even in a country situated downwind of large emission sources. Increases tend to have occurred in suburban and rural regions (Fig. 4). Increases observed from 2012 to 2015, particularly in suburban and rural regions, probably resulted from an energy consumption increase, as projected by Wang et al. (2014). As in Japan, this impact exceeded the potential reduction in vehicle emissions in suburban and urban regions.

To confirm the conclusions drawn from satellite observations, a detailed inventory, including top-down estimation by inverse modeling, is needed. Further investigation of the observed dramatic changes in tropospheric NO₂ levels will provide an opportunity to test our understanding of physical-chemical processes and emissions. This is particularly important for East Asia, widely recognized as the largest emission source of NO_x in the world.

4. Conclusions

This study utilized the most recent satellite observations and conducted a nationwide trend analysis to report the most recent change that the tropospheric NO₂ pollution level in East Asia was improved to the 5-year-ago level in 2015. We found that the NO₂ level over China decreased at $\sim 6\% \text{ year}^{-1}$ after 2011. While emission inventory data for the most recent years were unavailable, additionally-performed grid-based trend analysis suggested that the decrease was due to a nationwide action such as the widespread use of denitrification units. In Japan and South Korea, increasing tendencies in the NO₂ levels were observed after 2013 and 2012, respectively. While the tendency seen in South Korea was not very significant, it is likely that the increase found in Japan was due to increased NO_x emissions from the power plant sector as the significant substitution of thermal power generation for nuclear power occurred after 2011. Further investigation of the observed dramatic changes will provide an essential opportunity to test our understanding of physical-chemical processes and emissions.

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