# ANALYSIS OF LONG-TERM VARIATION OF OBSERVED OMI TROPOSPHERIC NO<sub>2</sub> CONCENTRATION USING CUMULATIVE PERCENTAGE AND AVERAGING

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**ABSTRACT:** In recent years, air pollution has become a serious problem in Northeast Asia due to the rapid growth of population and economy. Especially, the extensive use of fossil fuels has rapidly increased the emission of nitrogen oxides (NO<sub>X</sub>=NO+NO<sub>2</sub>). Nitrogen dioxide (NO<sub>2</sub>) has a detrimental affected on human health. Many studies have been conducted on NO<sub>2</sub> analysis by satellite observations. Previous studies have conducted on tropospheric NO<sub>2</sub> VCD (Vertical Column Density) trend analysis and long-term changes using various satellite data from GOME (Global Ozone Monitoring Experiment), SCIAMACHY (Scanning Imaging Absorption spectrometer for Atmospheric CHartographY), GOME-2 and OMI (Ozone Monitoring Instrument). However, most of the studies on tropospheric NO<sub>2</sub> trend analysis use monthly arithmetic averaging. In general, the use of arithmetic averaged values can be statistically biased if data are asymmetric distributions, which requires careful decision of the researcher. In this study, we analyze the regional (Northeast (NE) China, Southeast (SE) China, Republic of Korea, and Japan) tropospheric NO<sub>2</sub> VCD trend in Northeast Asia over a long-term period 2005-2018, using the monthly arithmetic averaging as well as statistical cumulative lowest 5th (B05), 25th (Q1), 50th (MED), 75th (Q3), and 95th (T95) percentiles of the tropospheric NO<sub>2</sub> VCD using the observed data from the OMI sensor on Aura satellite. As a result, the trends of average, the B05 and T95 trends of NE China, SE China, and Korea tended to increase from 2005 to 2011, and decreased from 2011 to 2018. Japan showed a tendency to decrease continuously. In addition, the average, B05, and T95 range of trend variation were shown to change differently. we found a decreasing tendency of arithmetic means in and around industrial areas and big cities. The B05 trend showed that background concentration has increased due to the rapid economic growth in Northeast Asia (+0.07, +0.06, and +0.03 x 10<sup>15</sup> molecules cm<sup>-</sup> <sup>2</sup>·yr<sup>-1</sup> in NE China, SE China, and Korea, respectively) while T95, representing high concentrations, has decreased in most regions, which is considered attributable to environmental regulations implemented by each country.

## **1. INTRODUCTION**

Nitrogen oxides ( $NO_X=NO+NO_2$ ) are composed of nitrogen monoxide and nitrogen dioxide. NOx are generated by both natural causes such as microbiological processes in soil and lightning, and anthropogenic sources including fossil fuel combustion, power plants and transportation (WHO, 2003; EPA, 2018). The emission of NO<sub>2</sub> affects chemical reactions in the global troposphere. NO<sub>2</sub> plays a catalytic role in forming ozone through photochemical reactions with hydroxyl radicals; produces secondary particulate matter; contributes to acid rain by reacting with water in the atmosphere; and affects climate change by increasing radiative forcing after being absorbed into the atmosphere. In addition, it has been reported that atmospheric  $NO_2$  has adverse effects on human health, such as causing asthma, respiratory or cerebrovascular diseases, and olfactory dysfunction (Filleul et al., 2005; Han et al., 2018). Especially,  $NO_2$  has increased dramatically in the past few decades due to rapid economic growth over Northeast Asia.

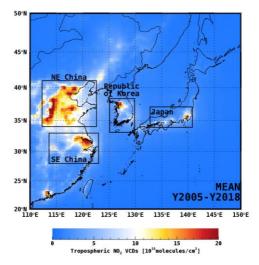
 $NO_2$  can be transported not only to neighboring counties but also to far distant regions, depending on weather conditions. Therefore, long-range transboundary air pollutants including  $NO_2$  have been highlighted as social and

diplomatic issues in Northeast Asia. Currently, a wide range of studies and observations are being conducted to monitor changes in  $NO_2$  concentrations. Ground observations generally have limited spatial coverage and airborne observations have limitations in obtaining continuous monitoring data due to high cost. On the other hand, satellite observation can provide long-term  $NO_2$  measurements on a continuous basis over a broad domain.

#### 2. DATA AND METHOD

Previous studies have focused on tropospheric NO<sub>2</sub> analysis and its long-term trends in major cities around the world using satellite data from GOME (Global Ozone Monitoring Experiment), SCIAMACHY (Scanning Imaging Absorption spectrometer for Atmospheric CHartographY), GOME-2, and OMI (Ozone Monitoring Instrument) (Richter et al., 2005; Hillboll et al., 2013; de Foy et al., 2016; Duncan et al., 2016; Krotkov et al., 2016; Souri et al., 2017; Cai et al., 2018). In these studies, most of the data used in the long-term trend analysis were simple arithmetic means or level 3.0 data. In general, an arithmetic mean is considered representative when it is located at the center of the Gaussian distribution. However, since most of the data observed in the actual atmosphere show asymmetric distributions, researchers need to make careful judgments about how well the arithmetic mean can represent the data (Grabusts, 2011; Speelman and McGann, 2013). In this study, trends of tropospheric NO<sub>2</sub> Vertical Column Densities (VCDs) were analyzed over Northeast Asia (Northeast (NE) China, Southeast (SE) China, Republic of Korea, and Japan), using monthly arithmetic means as well as 5th (B05), 25th (Q1), 50th (MED), 75th (Q3), and 95th (T95) percentiles of the level 2 tropospheric NO<sub>2</sub> VCDs (OMNO2, version 3.0) retrieved from OMI onboard NASA's Aura satellite.

#### 3. RESULT



# Figure 1. OMI tropospheric vertical column densities (VCDs) of NO2 form 2005 to 2018 over Northeast Asia averaged onto a 0.25°x0.25° grid.

Fig. 1 shows the distribution of arithmetic means of troposphere NO<sub>2</sub> VCDs during the study period (2005-2018), which provides a general picture of how the air pollutant is distributed over the observation domain. Regions with relatively high tropospheric NO<sub>2</sub> VCDs include densely populated and industrial areas such as Beijing, Tianjin, and Hebei in NE China; Shanghai in SE China; Seoul in Korea; and Tokyo in Japan. The average tropospheric NO<sub>2</sub> VCD for each region was highest in NE China followed by SE China, Korea, and Japan (9.92, 6.46, 4.47, and 3.55 x 10<sup>15</sup> molecules/cm<sup>2</sup>, respectively). Except for Japan, the annual averages tended to increase from 2005 to 2011 and then decline to 2018. An interesting finding was that the average, B05, and T95 values showed different trends over the period from 2005 to 2018 (Fig. 2). The averaged tropospheric NO<sub>2</sub> VCDs indicated gradual decreases in industrial and metropolitan areas while B05, considered as background concentration, showed an increasing trend (except for Japan) due to rapid economic growth in the region (NE China:+0.07, SE China:+0.06, Republic of

Korea:+ $0.03 \times 10^{15}$  molecules/cm<sup>2</sup>yr<sup>-1</sup>). T95 representing high concentrations tended to decrease in most regions (NE China: -0.34, SE China: -0.32, Republic of Korea: -0.16, Japan: -0.24 x 10<sup>15</sup> molecules/cm<sup>2</sup>yr<sup>-1</sup>), possibly attributable to stricter environmental regulations of each country.

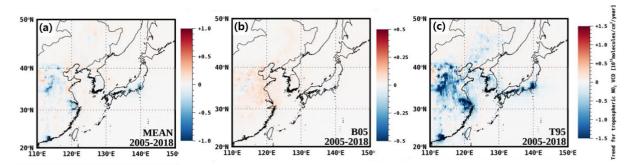


Figure 2. Spatial distribution trends of tropospheric NO2 VCD from satellite observations by OMI for the period 2005-2018 for mean (a), the lower 5th (b), and 95th percentiles (c).

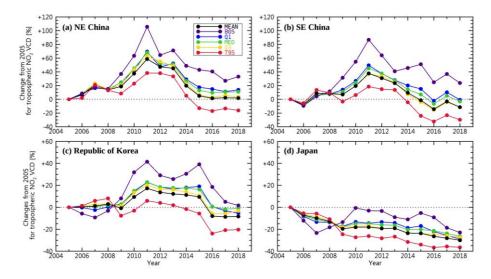


Figure 3. Relative change in the tropospheric NO2 VCD for each year compared to 2005 with the results for mean, the lower percentiles (i.e., 5th, 25th, 50th, 75th, and 95th) in black, purple, blue green, yellow, and, red color, respectively for Northeast China (a), Southeast China (b), Republic of Korea (c), and Japan (d).

Fig. 3 shows the relative change in the tropospheric  $NO_2$  VCDs for each year compared to 2005. Monthly arithmetic means remained relatively similar or decreased, except for Japan (+1.75% in NE China, -11.49% in SE China, and -8.36% in Korea); B05 significantly increased (+33.08%, +23.75%, and +1.68% in NE China, SE Chin, and Korea, respectively); and T95 decreased (-16.19%, -29.75%, and -20.35% in NE China, SE Chin, and Korea, respectively). In other words, background concentrations in NE China and SE China have significantly increased in 2018 compared to 2005, while  $NO_2$  concentrations during severe air pollution events have decreased over the same period.

These results revealed that studies of trend analysis of tropospheric  $NO_2$  VCDs show that statistical values (B05, Q1, MED, Q3, and T95) other than arithmetic averaging values should be taken into account. Also, there is a limit to the use of OMI data from polar orbiting satellites observed once a day. To compensate for this, the use of the Geostationary Environment Monitoring Spectrometer (GEMS), which planned to be launched in March 2020, will enable the monitoring and monitoring of high-resolution tropospheric  $NO_2$  VCD over day time.

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