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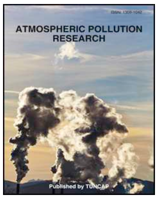

Long-term (2005–2015) trends analysis of OMI retrieved NO_2 columns in Taiwan

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ABSTRACT

In this study, the monthly data retrieved from the Ozone Monitoring Instrument (OMI) have been used to estimate long-term trends of NO_2 (2005–2015) in the entire Taiwan as well as in five representative city areas of Taiwan. The long-term NO_2 data have been used for trend analysis and evaluated to find the extent of the effects of NO_x emission reduction and related environmental regulations on the NO_2 trend. Desirable policies and strategies for further reducing NO_x emission have been also discussed. The corresponding reductions are 15–37% for the five city areas and 24% for the entire Taiwan over the 10-yr span. Clearly, the governmental policies/measures enacted during this 10-yr period are responsible for the reduction of tropospheric NO_2 column values. The OMI NO_2 column values of four cities (i.e., Taipei, Taichung, Kaohsiung, and Douliu) are relatively highly correlated ($r = 0.70$–0.95), indicating the cities have similar emission sources. Furthermore, the correlations between observed OMI NO_2 columns and their emission NO_x are all significant in the three metropolitan cities and the entire Taiwan ($r = 0.66$–0.89). The correlation of OMI-based NO_2 VCDs and ground-based NO_2 levels is high in Kaohsiung ($r = 0.82$) and moderate in Taipei and Taichung ($r = 0.60$ and 0.59, respectively). The satellite retrieved OMI NO_2 data clearly indicate their usefulness in evaluating decreasing trend of the NO_2, a precursor gas of $PM_{2.5}$ during the 10-yr span for the entire region of Taiwan and for the selected five cities.

1. Introduction

Nitrogen oxides ($NO_x = NO + NO_2$) are mainly produced during combustion of fuel and biomass, and contribute to surface ozone formation and aerosol (nitrate) production that may lead to potential health hazards (EC, 2013). For example, the incidence of respiratory diseases in Taiwan significantly correlates with NO_2 pollution at a confidence level of 99% (Lee et al., 2007) and the risk of developing osteoporosis among retired workers with a chronic obstructive pulmonary disease is positively associated with long-term exposure to annual averaged NO_2 (Lee et al., 2014). Furthermore, traffic-related air pollution has increased the risk of Parkinson's disease in Taiwan (Lee et al., 2016). The well-known effect of NO_2 on public health also has been reported elsewhere, e.g., the consistent positive correlations between ambient oxidants (O_3 and NO_x) and daily mortality across the Pearl River Delta cities in China (Tao et al., 2012).

In addition to the formation of secondary O_3 and $PM_{2.5}$, the well-known environmental consequences caused by NO_2 (as well as SO_2) include acidification of lakes, acid rain, reduced visibility, affecting cloud formation, photochemical smog, and, more importantly, deterioration of public health and environment (e.g., Zhang et al., 2017b). In response to several international treaties regarding the reduction of NO_x emission (e.g., 1988 Sofia Protocol concerning the Control of Emissions of NO_x, and 1999 Gothenburg Protocol to Abate Acidification, Eutrophication and Ground-level Ozone), “Acid Program” and “NO_x State Implementation Plan” (Geddes et al., 2016) and different EU Directives (UNECE, 2010) have been established by the USA and the EU, respectively to reduce atmospheric NO_2 levels.

Due to the massive economic growth in Taiwan in the past, the environment has been deteriorated significantly, resulting in sporadic violations of the ambient air quality standards for ambient O_3 and $PM_{2.5}$ concentrations. Despite efforts made by the Taiwan Environmental Protection Administration (TEPA) to enhance the ambient air quality, O_3 and $PM_{2.5}$ pollution problems are still causing

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public awareness and affecting public welfare. Since NO_x plays a role as a precursor gas in the formation of O_3 and secondary $PM_{2.5}$, control of NO_x gas is of importance in reducing atmospheric $PM_{2.5}$ and O_3 concentration.

The underappreciation of mitigation of anthropogenic NOx emissions in Taiwan is reflected by the rather relaxed ambient air quality standard for NO_2 (i.e., 250 ppb (1-h average)), which is much higher than those specified in the US air quality standards (100 ppb; https://www.epa.gov/criteria-air-pollutants/naaqs-table), EU (105 ppb; http://ec.europa.eu/environment/air/quality/standards.htm), WHO guidelines (105 ppb; WHO, 2015), Turkey (100 ppb; Dogruparmak and Ozbay, 2011), and even higher than those in Vietnam, Malaysia and Egypt (Hrebenyk et al., 2013). Nonetheless, a few specific control policies/measures have been implemented in Taiwan to reduce atmospheric NO_x; for example, in order to reduce NO_x in flue gas from power plants and other industrial applications as well as in vehicle exhaust, stricter emission standards have been implemented and 2-wheel motorcycles are being eliminated, among others. Consequently, there has been a reduction in the ambient NO_x concentration in Taiwan. For example, air quality trend from 1994 to 2003 indicated an NO_2 reduction trend of 0.51 ppb/yr based on surface monitoring stations (Chang and Lee, 2007) in Taipei; and NO_2 level was decreased by 16% between 1993 and 2012 also in Taipei (Ding et al., 2016). In their study with surface air-quality stations, Chen et al. (2014a) observed 24.2% decrease of NO_2 and an annual decreasing rate of -0.42 ppb/yr for Taipei and 34.5% decrease and -0.46 ppb/yr for the entire Taiwan during the period of 1994–2012. Meanwhile, NO_2 in the air over Taiwan was transported to the South China Sea. Contribution of Taiwan to NO_2 levels over the South China Sea was estimated 20%. Not surprisingly, that of China was estimated 66% (Zhao et al., 2015).

As Taiwan island (Fig. 1, left plot) is located off the south-east coast of China, the long-range transport of Asian air pollutants and dusts are still a major environmental concern during the winter season (Lin et al., 2007). Probably, Lin et al. (2005) would be the first research team that investigated the impact of long range transport (LRT) on the air quality (CO, SO_2 and PM_{10}) of Taiwan using ground-based measurements over the winter monsoon periods of 2000 and 2001. Based on concentration differences of pollutants among different meteorological conditions, they estimated that the LRT of particulate pollutants would contribute about 30 mg/m^3 to the PM_{10} concentrations in northeastern Taiwan; a smaller contribution in western plain of Taiwan. Contributions of the LRT to CO and SO_2 were about 230 and 0.5 ppb, respectively. In addition, the impact of the LRT on PM_{10} especially during dust events was estimated to be nearly 100% at windward background stations along the north and northeast coast. However, the impacts decrease to around 60–80% for cities in northern Taiwan as local emissions increase. Due to geographic blocking, a smaller impact is estimated for the western plain of Taiwan. In general, impacts of LRTs on the air quality of coastal areas are greater than that of urban areas. Junker et al. (2009) analyzed the time-series data from 1993 to 2006 and suggested that pollution from the Asian mainland would have significantly deteriorated the background air quality over the Pacific oceanic area (e.g., Taiwan island). For example, a doubling of the NO_x concentration was observed at the Wan-Li (head of Taiwan island) and Heng-Chun (tail of Taiwan island) sites between 2001 and 2006. In particular, Chen et al. (2015) estimated long-term (1994–2010) changes of the relative contributions of domestic and foreign sources of NO_x on the entire Taiwan. They identified the contribution from foreign sources by using data measured at selected coastal monitoring stations in Taiwan under specific meteorological conditions. Then, the domestic contribution was obtained by subtracting the foreign contribution from the overall concentration. Although the trend of ground-based NO_2 concentrations was estimated as -1.4%/yr, that of the background concentration was estimated as 3.8%/yr. Therefore, they concluded that the efforts of Taiwan in reducing air pollution was largely negated by the LRT from China. The NO_2 fraction from the LRT increased twice from 15% to 30% over the period of 1994–2010. Duncan et al. (2016) analyzed Ozone Monitoring Instrument (OMI) NO_2 data from 2005 to 2014 and noted that pollutants transported from China possibly influenced Korea and Japan, diminishing the effectiveness of local pollution controls. Besides, they further found an increase of NO_2 level over the marine area between China and Taiwan. It is probably because changes in NO_2 levels over the west coast of Taiwan were influenced by the NO_2 imported from China. China coastal cities near Taiwan showed large increases in NO_2 levels: for example, an increase of 49.6% and 12.2% over the period between 2005 and 2014 in Quanzhou and Fuzhou, respectively. In their study,

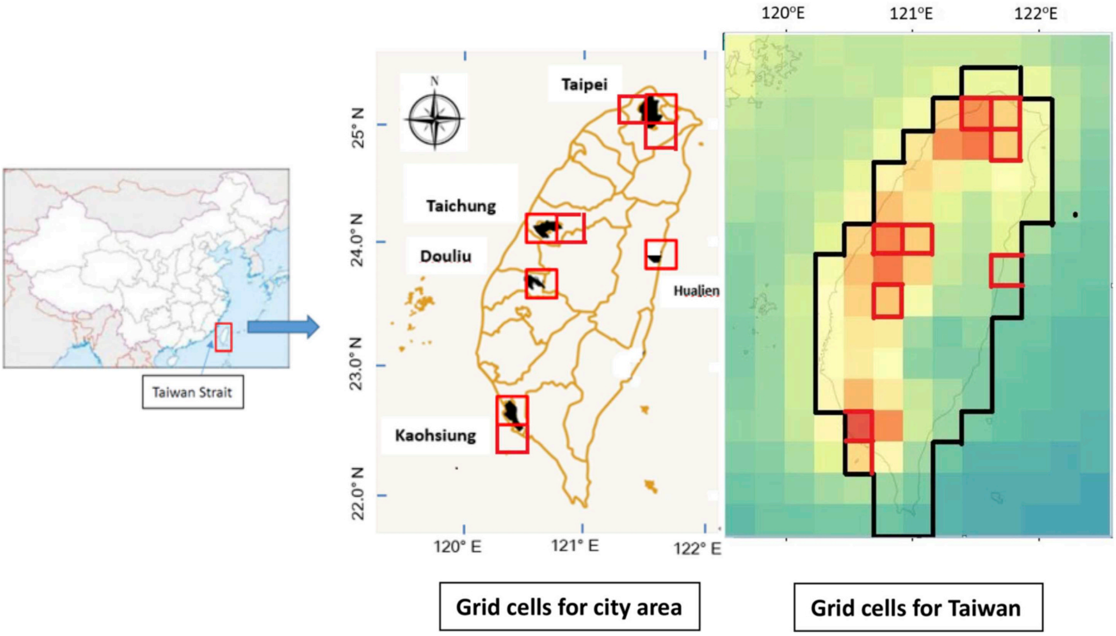

Fig. 1. Map for middle panel: grid cells for selected five cities in Taiwan (grid cells in red square is 0.25° by 0.25° used for each city; black areas within the grid represent city boundary); Map for right panel: grid cells (black contour) for entire Taiwan, the background color of grid cells is the average OMI VCD columns in 2005.

unfortunately, only Taipei (−29.2%) was analyzed in the case of Taiwan.

Although the ground observation data are useful to see whether important pollutants meet the ambient air quality standards, these ground monitoring stations only provide ambient air quality information about the surrounding area of each station and cannot represent the overall air quality in Taiwan. In addition, these stations only monitor near ground air quality and cannot estimate pollutant levels present in the atmospheric planetary boundary layer.

On the other hand, the use of pollutant observations from satellite is able to observe pollution hotspots, including screening high vertical column densities (VCDs) of NO_2 over industrial/power plant regions (Kramer et al., 2008), observing pollution status for special events (e.g., high-pollution episodes, volcanic flux and forest fires), and likely improve the understanding of anthropogenic emissions on regional, continental and global scales (Krotkov et al., 2016). For example, Duncan et al. (2016) used OMI for NO_2 analysis to see changes in urban NO_2 VCDs around the world from 2005 to 2014. They found significant heterogeneity in the NO_2 changes and discussed environmental regulations leading to decreases in NO_x. However, satellite data as they are have limitation. One of the major issues is that only day-time NO_2 data can be obtained by the OMI. Data-gridding and filtering-out of original OMI NO_2 columns also enlarge the uncertainty of data. Nonetheless, in analyzing the long-term trends of NO_2, the OMI NO_2 VCDs can be a good surrogate for ground-based NO_2 monitoring. In short, based on satellite observations, developed countries have exhibited significant reduction in tropospheric NO_2 columns, while some in developing countries (e.g., India, Pakistan, Middle East) are still shown increasing trends (Krotkov et al., 2016; Duncan et al., 2016).

Currently, there is no spatial and temporal study done in areas of Taiwan regarding the tropospheric OMI NO_2 columns, except one paper covering the Eastern Asia mentioning a Taiwan NO_2 trend (Souri et al., 2017), and another global study covering Taiwan (Duncan et al., 2016). Therefore, in this study, the spatial OMI satellite observations were retrieved to observe long term trends of NO_2 (2005–2015) in the entire Taiwan as well as in five city areas representing the typical geographic distribution over Taiwan. The long-term NO_2 data derived from the OMI are conveniently retrieved from the NASA Goddard Earth Sciences Data Active Archive Center (USA) for performing trend analyses and evaluating the extent of effects of NO_x emission reduction and related environmental regulations on the NO_2 trend. Desirable policies and strategies for further reducing NO_x emission (ambient NO_2 levels) are also discussed. It is our hope that these measures/policies should be useful for regulatory agencies in developing countries to combat their air pollution problems.

2. Methodology

2.1. Targeted areas

Five city areas, representing the geographic distribution of Taiwan, were selected to analyze their long-term trends of NO_2 concentrations. The five cities include two urban ones (Taipei, Taichung), one highly industrialized city (Kaohsiung), one remote east coastal city (Hualien), and Douliu, a small city located in the west-center plain of Taiwan. The relevant information for each of the five city areas including the grid location, population, vehicle numbers, and GDP is listed in Table 1 with their geographic location shown in Fig. 1. It is noted that information provided for the cities is for the old boundary since boundaries of some cities have been reorganized or enlarged in 2010.

2.2. OMI NO_2 data product

In this study, the data retrieved from the OMI of the Earth Observation System Aura satellite was used to analyze the long-term NO_2 trends. The Earth Observation System Aura satellite was developed by the Finnish Meteorological Institute and the Netherlands Aviation Authority for detecting a variety of atmospheric compositions (Levelt et al., 2018). Among all the on-board instruments for observing atmospheric pollutants, the OMI has been the most widely used and provided longest data record (Krotkov et al., 2016). Details for the OMI sensor, design and performance are described elsewhere (e.g., Levelt et al., 2006).

The Level 3 (standard product version 3) monthly data were retrieved from the NASA Goddard Earth Sciences Data Active Archive Center (GES DISC; http://disc.sci.gsfc.nasa.gov) using a spatial resolution of 0.25° × 0.25° from the Giovanni interface (Giovanni 4; http://giovanni.gsfc.nasa.gov/giovanni/). In this study, when the OMI tropospheric NO_2 VCDs data were retrieved, a radiative cloud fraction less than 0.3 were screened out (no used flags in the level 3 product) and directly used with negative values discarded. Level 3 products have been produced after filtering for the cross-track scenes mentioned per anomaly (NASA, 2012) and used by many other investigators (e.g., Lin et al., 2010; Prados et al., 2010; ul–Haq et al., 2014; Zhang et al., 2017a).

2.3. Selected grid cells

For Taiwan, the area of grid cells does not match the irregular shape of Taiwan (Fig. 1, right plot). In the present study, we have selected grid cells that covered any parts (including all or part grid covered) of Taiwan and used population density (Fig. SM-1) as weighting for objective representation of Taiwan (see areas of city and grid cells in Table 1 and Fig. 1). In the Fig. SM-1, ten population density ranges have been estimated for administrative districts of Taiwan. This study used the weighting from 1 to 10 for the related population density ranges. The weighting data of population density should represent the weighting of air pollution sources with the proportion of population living in the cell area and with the transportation flows.

It is further noted that since the area of each grid (red square in Fig. 1 middle plot) is larger than that of each selected city (black shaded areas in Fig. 1), the use of city's name in the present study is for convenience only since grid cell covering each city also represents the additional surrounding area. Clearly, the actual NO_2 VCD in each city should be higher than those indicated by the grid cells. Again, we have selected grid cells that cover any part (three grid cells for Taipei, two for Taichung and Kaohsiung, respectively) and used population density (Fig. SM-1) as weighting for various grid cells. However, the data were subsequently used for trend analysis and for comparison with one another, hence based on the above analysis, the systematic error may be reduced to minimum level and the overall error in retrieved NO_2 columns in our trend analysis should be insignificant.

2.4. Deseasonalized time series

The original monthly data from OMI NO_2 clearly showed highly annual seasonal cycle. Thus, the data should be first deseaonalized to remove any seasonal fluctuations and cyclical variations to see the real changes in NO_2 columns. Typically, time series NO_2 data can be deseasonalized in two parts; the long-term trend and irregular components. The detailed methodology used in the present study can be found elsewhere (Hilboll et al., 2013; Zhang et al., 2017a). In this study, the procedure of the deseasonalization was provided in Supplemental Material (SM) (Text SM-1). To obtain long term trend line, this study also used 7-month moving average approach to further smooth data by reducing any random fluctuation. The length of 7-month was designed to cover the length of two seasons (i.e., 6 months). The slope based on the least square analysis of moving average data represents the decreasing or increasing trend of NO_2 data. The corresponding regression coefficient (R^2) of trend line is then used to see the extent of goodness fit between trend line and the moving average data.

2.5. Emission inventories

Anthropogenic emissions in Taiwan were obtained from the Taiwan Emission Data System (TEDS) (TEPA, 2017a), which was published every 3 years since 1988 by TEPA. Three categories of point, line, and area sources are included in the database with spatial resolution of 1 km × 1 km for line and area sources and 1 m × 1 m for each point source. Pollutants included in the database are SO_x, NO_x, TSP, PM_{10}, $PM_{2.5}$, CO, CH_4, NMHC (non-methane hydrocarbon), NH_3, and Pb. Each category of point, line and area sources can be divided into several sectors. Usually in Taiwan point sources are classified into 18 sectors including steel industry, power plant, chemical material manufacture, cement manufacture, food industry, textile industry and etc.; line sources are classified into 10 sectors including heavy duty diesel truck, passenger car, two-stroke motorcycle, four-stroke motorcycle and etc.; and area sources are classified into 18 sectors including vehicle-driven dust, restaurant, construction, agriculture operation, open-burning, bare soil and etc. The amount of NO_x emission for each sector of the three source categories in Taiwan in 2010 and 2013 base year are shown in Table SM-1. In general, the emission amounts for line and area sources are estimated using the emission factor method, while that for point sources are estimated using the measured data requested by TEPA. Each point source in the database contains information for emission rate, stack characteristics, fuel type, operating duration, and process code, among others. All the sources with information in TEDS can be used to calculate the 3-D spatial distribution of pollutant emission rate in Taiwan.

3. Results and discussion

3.1. Trend analysis of OMI NO_2 columns

For Taipei, the irregularly cyclic variations of the original OMI NO_2 data between 2005 and 2015 are shown in Fig. 2a with the deseasonalized NO_2 data shown in Fig. 2b and NO_2 trend line in Fig. 2c. With 7-month moving average, the trend shows decline of 0.13 ± 0.02 × 10^{15} molecules/cm^2/yr ($R^2 = 0.60$) (Table 1), or overall 28% reduction from 2005 to 2015 (2.4% reduction/yr) – results is close to 2.9%/yr decrease in NO_2 in Taipei from OMI data from 2005 to 2014 (Duncan et al., 2016). The annual decreasing rate for each city essentially follows those of reduction percentage between 2005 and 2015, with the highest reduction rate in Kaohsiung (0.19 ± 0.03 × 10^{15} molecules/cm^2/yr, $R^2 = 0.62$), followed by Taichung (0.16 ± 0.02 × 10^{15} molecules/cm^2/yr), Taipei (0.13 ± 0.02 × 10^{15} molecules/cm^2/yr, $R^2 = 0.60$) and Douliu (0.11 ± 0.02 × 10^{15} molecules/cm^2/yr, $R^2 = 0.55$) with slight reduction in coastal city of Hualien (0.013 ± 0.013 × 10^{15} molecules/cm^2/yr, $R^2 = 0.031$) (Table 1). The decreasing trend was also found in developed countries, e.g., decreasing rate of 4.3%/yr from 2005 to 2008 and 2.9%/yr from 2010 to 2013 in Los Angles (Lamsal et al., 2015). On the other hand, some increasing rates were reported in regions in developing countries, e.g., increasing rate of 0.26 × 10^{15} molecules/cm^2/yr in Yangtze Delta between 2005 and 2014 (Wang et al., 2015).

The temporal trend plot for the entire Taiwan is shown in Fig. 2g (right column) with the decreasing trend of 0.09 ± 0.01 × 10^{15} molecules/cm^2/yr ($R^2 = 0.80$) which lies in between the values of five cities (0.013–0.19 × 10^{15} molecules/cm^2/yr). For comparison, Wang et al. (2015) showed decreased rate of 0.1 × 10^{15} molecules/cm^2/yr in western Taiwan between 2005 and 2014 and the decreasing rate of 0.07 × 10^{15} molecules/cm^2/yr for entire Taiwan from 2004 to 2014 (Souri et al., 2017). Similar figures for other 4 cities are illustrated in Figs. SM-2 to SM-7.

Finally, the spatial distributions of average NO_2 over Taiwan in 2005, 2015 and "difference between 2005 and 2015" are illustrated in Fig. 3. It is noted that scale for color intensity for "difference between 2005 and 2015" (far right) is different than those of 2005 and 2015 plots. Clearly, a significant reduction is noticed between 2005 and 2015 which shows the highest reduction occurring in southern part of Taiwan (Kaohsiung) in the "difference between 2005 and 2015" plot as reflected by the largest reduction in Kaohsiung (Table 1). In brief, the long-term trends of decreasing NO_2 in the 10-yr span clearly indicates the successful policy execution of air pollution control, such as the effectiveness of NO_x emission reduction (more to be discussed later) by upgrading vehicle emission standards (e.g., NO_x emission standards had been upgraded to 0.07 g/km in 2008 from 0.25 g/km in 1999 for gasoline passenger car, while for light diesel trucks the standards were upgraded by one order of magnitude, or from 0.68 g/km in 1999 to 0.07 g/km in 2008 (TEPA, 2013)), new flue gas standards (100–500 ppm of NO_x depending on types of combustion fuels and the flue gas flow rate). It is noted that successful application of denitrogenation processes as an air pollution control device in power generation plants in Taiwan certainly reduces NO_x emissions (TEPA, 2017a). Hence, the amount of emissions from point and line sources decreased over the period of 2010 and 2013, especially in the sectors of chemical material manufacturing, metal manufacturing, and automobile manufacturing. In addition, the trends of NO_2 VCDs and NO_x emission will be discussed in Section 3.3.

Specifically, in 2006, the Air Pollution Control Act was amended to include emission standards from stationary sources; the values of emission standards depended on the type of industry, facility and locality, among others (Tsai, 2016). Starting in 2004, the vehicle emission standards for US Federal Test Procedure (FTP) Transient Cycle driving

Table 1
Relevant information for studied cities.

City	Area (km^2)	Population[a]	Vehicles[b]	GDP[c]	OMI NO_2			
		(10^3)	(10^3)	(10^9 USD)	2005 VCDs[e] (10^{15} molecules/cm^2)	2015 VCDs[e] (10^{15} molecules/cm^2)	Annual decreasing rate[f] (10^{15} molecules/cm^2/yr)	Reduction % between 2005 and 2015
Taipei	276	2705	1820	81.7	5.3 ± 2.6	3.8 ± 1.3	0.13 ± 0.02 ($R^2 = 0.60$)	28
Taichung	163	1155	1032	20.9	5.7 ± 2.6	4.4 ± 2.4	0.16 ± 0.02 ($R^2 = 0.78$)	23
Kaohsiung	154	1516	1654	32.9	6.2 ± 3.8	3.9 ± 2.6	0.19 ± 0.03 ($R^2 = 0.62$)	37
Douliu	94	108	107[d]	1.8[d]	4.8 ± 3.0	3.7 ± 2.3	0.11 ± 0.02 ($R^2 = 0.55$)	23
Hualien	29	106	103[d]	2.1[d]	2.0 ± 0.5	1.7 ± 0.7	0.013 ± 0.013 ($R^2 = 0.031$)	15
Entire Taiwan	36,014	23,347	21,401	474.0	4.1 ± 1.8	3.1 ± 1.3	0.09 ± 0.01 ($R^2 = 0.80$)	24

[a] 2015 data Source: http://statdb.dgbas.gov.tw/pxweb/Dialog/varval.asp?ma = CS0201A1A&ti = &path = ../database/CountyStatistics/&lang = 9.

[b] 2010 data including motorcycles; source: http://erdb.epa.gov.tw/DataRepository/ReportAndStatistics/StatSceMotors.aspx.

[c] 2011 data for 2011, no update from 2012 for Taiwan Government; source: http://bbs.tianya.cn/post-333-303106-1.shtml.

[d] Estimated from ratio of the population of these cities and their counties.

[e] Data are presented as mean ± standard deviation.

[f] Data are presented as mean ± confidence level of 95%.

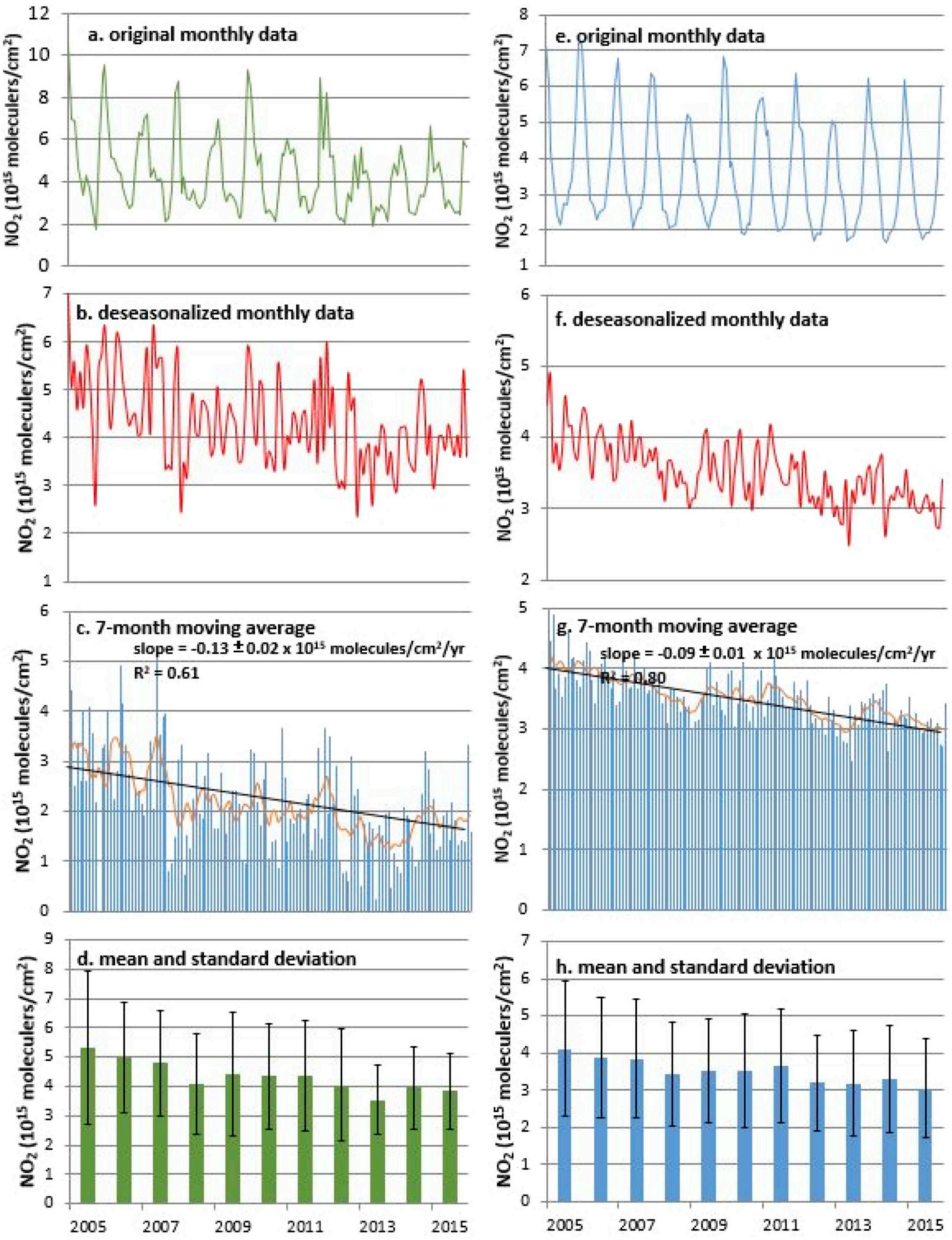

Fig. 2. Long term raw monthly NO_2 VCD data (a), deseasonalized (b), 7-month moving averages with a yellow trend line (blue bars: deseasonalized monthly data) (c) and annual means with standard deviations (d) analyzed for Taipei, and Long term raw monthly NO_2 VCD data (e), deseasonalized (f), 7-month moving averages with a yellow trend line (blue bars: deseasonalized monthly data) (g) and annual means with standard deviations (h) analyzed for Taiwan.

tests had been upgraded frequently for new vehicles (TEPA, 2013). In 2012, the standard for buses and trucks, subject to either European Stationary Cycle (ESC) or European Transient Cycle (ETC) test was 2 g NO_x/kWh. The emission standard set in 2001 for powered two-wheel vehicles was equal to Euro 3 (UDC cycle; Delphi, 2016). Clearly the progresses made in reducing vehicle NO_x emission partially explain the decreasing OMI NO_2 column trend. Furthermore, the use of the newly expanded subways in two major cities (Taipei and Kaohsiung) certainly reduced emission from private vehicles as noted by Ding et al. (2016). The installation of public transport systems, such as mass rapid transit railway and the rapid transit bus system might also contribute to the decreasing NO_2 reduction trend.

On the other hand, the number of registered light motorcycles in Taiwan, mostly two-strokes, had dropped from 4.6 million in 2003 to 1.88 million in 2015, a decrease of nearly 2.72 million vehicles in a period of 12 years (TMTC, 2016). On the other hand, the 4-stokes motorcycles may be increased during the same time span due to its much less pollutant emissions. Recently, the administrator of TEPA has announced that it is planning to get rid of 300,000 two-stoke motorcycles in 2017 and to impose complete ban using 2-stroke motorcycles in Taiwan in 2020 (TEPA, 2017b). According to Table SM-1, the NO_x emission of vehicles was 217 thousand tons with 50% of total amount in 2010 and reduced to193 thousand tons with 47% of total amount in 2013, meanwhile the NO_x emission of motorcycles with both two- and

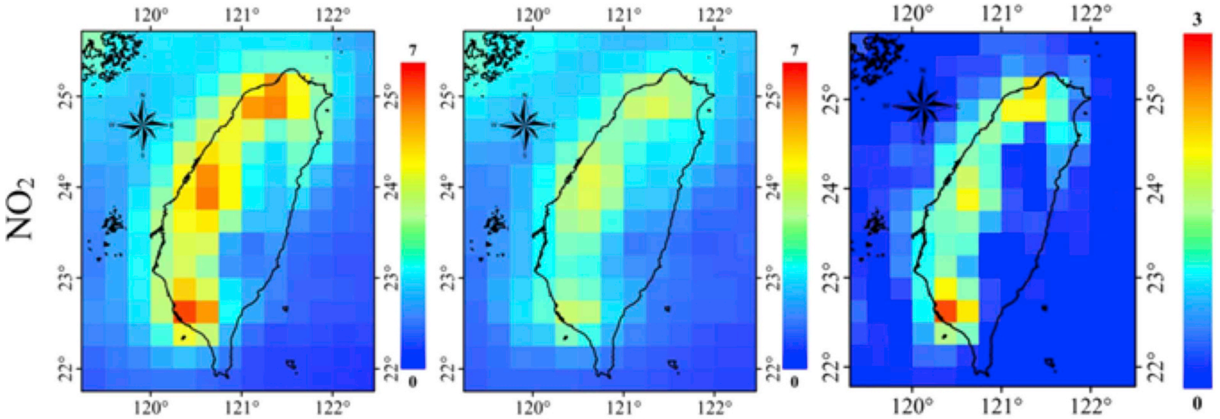

Fig. 3. Spatial distributions of average NO_2 (10^{15} molecules/cm^2) over Taiwan at 2005 (left), 2015 (middle) and "difference between 2005 and 2015" (right).

four-strokes was 17.6 thousand tons in 2010 and decreased to 12.9 thousand tons in 2013. The contribution of motorcycle emission to the vehicle emission was 8.1% in 2010 and 9.1% in 2013.

Based on retrieved satellite data, Zien et al. (2014) have reported about 3800 global events of LRT over the ocean during the period 2007 to 2011. In fact, the $PM_{2.5}$ levels around the Taiwan Strait were influenced by LRT, mainly from North China, the eastern coast of China, Korea Peninsula, and South Japan (Li et al., 2017). The effect of LRT of pollutants is also reflected in $PM_{2.5}$ levels in Tainan City (between Taitung and Kaohsiung) where each of trans-boundary pollution from neighboring cities, LRT, and local emissions contributed to about one-third of $PM_{2.5}$ levels (Lu et al., 2016). The Chinese contribution of NO_3^- in $PM_{2.5}$ during the winter and spring to Japan and Korea due to westerly wind is significant (Qu et al., 2016), or contributing to about 38% of NO_3^- in Japan and 67% in Korea in winter (Duncan et al., 2016). Thus it is possible that changes in the OMI NO_2 VCDs in western coast of Taiwan (Fig. 1) are influenced by the import from mainland NO_2 as reported by Duncan et al. (2016). Chen et al. (2014b) further stated that LRT of NO_3^- via indirect means (i.e., transport of NO_x aerosols and then forming secondary $PM_{2.5}$ locally) to Taiwan contributed to $PM_{2.5}$ levels in Taiwan. Clearly, the impact of LRT on air quality in Taiwan needs to be further investigated. Therefore, this study simply used the OMI VCDs data and chose 19–35 grid cells for open-ocean areas surrounding Taiwan as background areas. The baselines of the west, east, north and south were 120° E, 122° E, 25.5° N, and 22° N that can minimize the influence of sources from the island of Taiwan. Results clearly showed all city areas and the entire Taiwan were significantly affected by LRT. All the NO_2 VCDs significantly decreased after the effect of LRT had been eliminated (LRT-affected vs. LRT-free, Table SM-2). The annual NO_2 decrease in the major cities and the entire Taiwan becomes even more pronounced, once the LRT influence is accounted for (Table SM-2), thus signifying the effectiveness of environmental policies and regulations. For example, the annual NO_2 decreasing rate of Kaohiung was $0.33 \pm 0.04 \times 10^{15}$ molecules/cm^2/yr in the LRT-free case. On the other hand, the NO_2 decreasing rate was only $0.19 \pm 0.03 \times 10^{15}$ molecules/cm^2/yr in the original data (Table SM-2). Result of detailed analysis is summarized in Text SM-2.

In summary, comparison of NO_x trend in different countries/regions/cities listed in Table SM-3 may provide readers as to the magnitude of decreasing trend in developed/developing countries. It is noted that during the earlier period (2005–2011), China as a developing country encountered increasing NO_2 percentage of 53%, but later on (2011–2015), the NO_2 in fact decreased by 32% (Liu et al., 2016).

3.2. OMI NO_2 columns in 2015

The annual average of NO_2 VCDs in five cities as well as in the entire Taiwan from 2005 to 2015 is shown in Table SM-4. The OMI NO_2 VCDs in 2015 in five cities of Taiwan ranged from 1.7 to 4.4×10^{15} molecules/cm^2 with the standard deviation from 0.7 to 2.6×10^{15} molecules/cm^2 (Table 1). The standard deviations of NO_2 VCDs in Taipei (1.3×10^{15}) and Hualien (0.7×10^{15}) were both lower than other cities ($2.3–2.6 \times 10^{15}$), meaning low variation around year. With location in rural eastern coast of Taiwan, the mean of NO_2 VCDs in Hualien also very low. In addition, the reduction ranged from 15 to 37% between 2005 and 2015. The order of selected cities for 2015 NO_2 VCDs is: Taichung ($4.4 \pm 2.4 \times 10^{15}$ molecules/cm^2) > Kaohsiung ($3.9 \pm 2.6 \times 10^{15}$) > Taipei ($3.8 \pm 1.3 \times 10^{15}$) > Douliu ($3.7 \pm 2.4 \times 10^{15}$) > Hualien ($1.7 \pm 0.7 \times 10^{15}$) (also in Table 1). However, Kaohsiung exhibited the highest reduction of 37% between 2005 and 2015, followed by Taipei, Taichung and Douliu (23–28%). In the global study of OMI data, Duncan et al. (2016) reported 29% reduction in Taipei between 2005 and 2014 – very close to our 28%, considering actual Taipei data should be much higher than 28% as mentioned before. As would be expected, the values and reduction percentage in the entire Taiwan (24%) lie within range of above five cities (15–37%). For comparison, Krotkov et al. (2016) showed 40% decrease in eastern US from 2005 to 2015 with no changes in Eastern Europe and North China Plain, and 20% increase in Middle East.

It appears that NO_2 data in three metropolitan areas is related to density of vehicles (number of vehicles divided by area) with 10.7 vehicles/km^2 for Kaohsiung, 6.6 and 6.3 vehicles/km^2 for Taipei and Taichung, respectively, since mobile sources constitute a major fraction of NO_x pollution in these cities (Kurokawa et al., 2013; TEPA, 2017a). In addition, Kaohsiung and to some extent Taichung have other industrial NO_x emission sources (e.g., power plants and petrochemical complexes), thus Taichung exhibited the highest NO_2 2015 VCDs (4.4×10^{15} molecules/cm^2), followed by Kaohsiung (3.9×10^{15} molecules/cm^2). The remote area of coastal city of Hualien had the lowest NO_2 VCDs (1.7×10^{15} molecules/cm^2) among five cities evaluated. The Taiwan Cement Co. in Hualien may contribute the portion of observed NO_2 level, based on US EPA NO_2 emission factors (0.67 kg/ton clinker; Edwards, 2014). The relatively high NO_2 VCDs in Douliu (3.7×10^{15} molecules/cm^2) is influenced by the transport from Taichung. The seasonal trends and pollution levels of the two city-areas are similar. In addition, the lower dispersion rate due to the effect of nearby mountains may partially explain the observed NO_2 VCDs.

3.3. Comparison with NO_x emission

The main sources for NO_2 emission include power plants, industrial sectors and transportation. Vehicle emission accounted for 85% NO_x emission in Taipei, and 50% overall emission in Taiwan (TEPA, 2017a). The contribution from Taipower (state-owned electric utility company) to the entire Taiwan's NO_x emission was 16% in 2010 (ESC, 2015). The trend of NO_x emissions in three major cities as well as in the entire

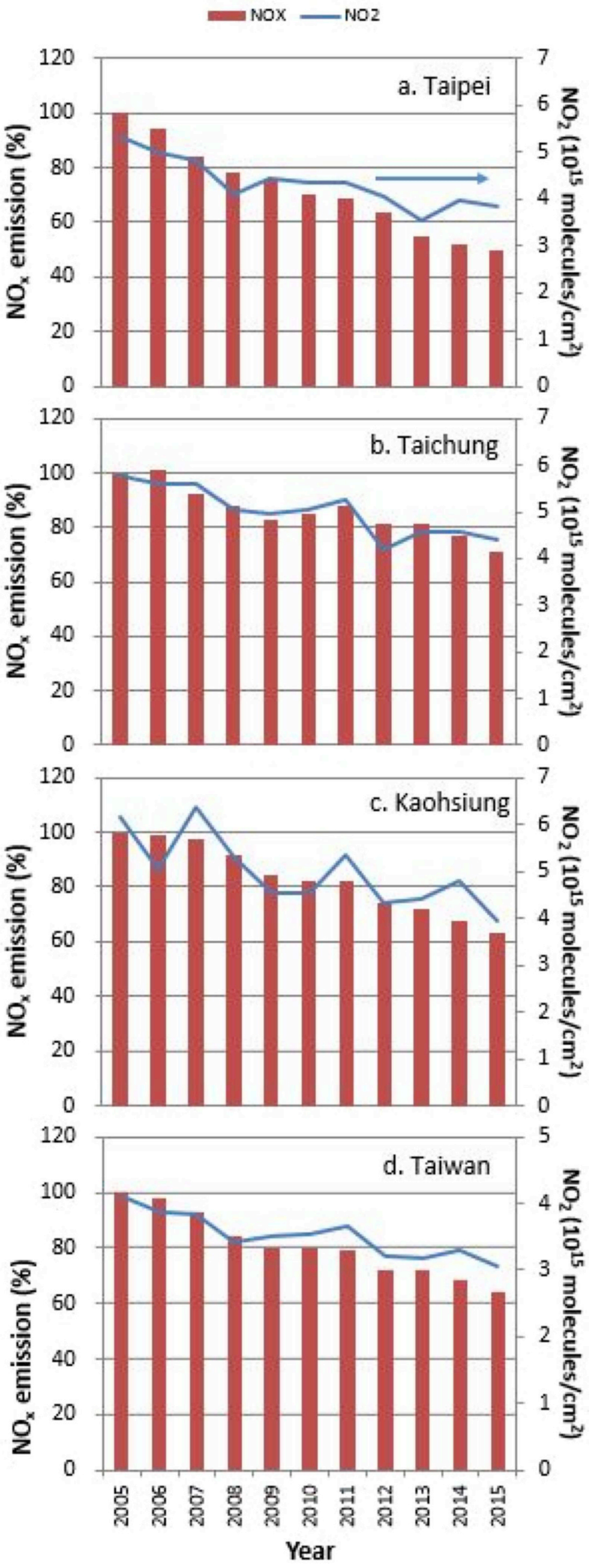

Fig. 4. NO_x emission trend (bar) for threes cites and entire Taiwan with OMI NO_2 VCDs (line) shown for comparison.

Taiwan are shown in Fig. 4 with corresponding OMI NO_2 VCDs shown for comparison. Clearly, the highest percentage reduction from 2005 to 2015 is in Taipei (50%) while the highest annual decreasing rate is in Kaohsiung (1.69 ± 0.18 kt/yr, $R^2 = 0.98$; Table 2) with the overall reduction of 37% between 2005 and 2015 (Table 2, Fig. 4). For the entire Taiwan, the decreasing rate is significant (18.7 ± 2.97 kt/yr, $R^2 = 0.96$ or overall reduction of 36%). For comparison, the EU-27 member countries exhibited 27% emission reduction of NO_x between 2002 and 2011 (Guerreiro et al., 2014); 43% reduction in USA, or 20.3 Mt in 2005 to 11.5 Mt in 2015 (https://www.epa.gov/air-trends/nitrogen-dioxide-trends#nonat); and 30% NO_x reduction between 2004 and 2013 in Europe (EEA, 2015).

Table 2 showed NO_x reductions in Taipei (−50%), Taichung (−29%), Kaohsiung (−37%) and the entire Taiwan (−36%) over the 10-year period of 2005–2015. However, OMI NO_2 VCD (Table 1) exhibited less percentage reductions for Taipei (−28%), Taichung (−23%), Kaohsiung (−37%) and the entire Taiwan (−24%) over the same period. Only Kaohsiung had a consistent reduction rate (37%). Not only the entire Taiwan (Fig. 4d), but also Taipei (Fig. 4a) show the same tendency of faster NO_x emission reduction compared to the slower changes in NO_2 VCD. A similar observation was made in a study with five cities of Texas by Choi and Souri (2015). During the period, the Taiwanese government did not relax any of relevant regulations, but put more effort to eliminate two-stroke motorcycles and provide a subsidy for electronic motorcycles from 2008. Unfortunately, the number of diesel-driven vehicles increased by 15.5% from 2012 to 2015 (data not available between 2005 and 2011; see Table SM-5) as well as the total number of vehicles in Taiwan (16.1% from 2005 to 2015). Besides, the possible reasons leading to the discrepancy are (1) a shorter lifetime of NO_2 in day-time when OMI NO_2 VCDs are measured, (2) a missing ratio of NO_2 VCD density to NO_x emission (NO_2/NO_x), (3) lack of information about non-surface sources, and (4) inaccuracies in the inventory of anthropogenic emission sources (Choi and Souri, 2015).

The decreasing trend of NO_2 emission for the entire Taiwan between 2007 and 2011 was 2.1% per year based on satellite observations (Mijling et al., 2013), as compared to average value of 3.3% per year for the entire Taiwan and 3.9% per year for 3 major cities (Taipei, Taichung and Kaohsiung) in the present study (2005–2015). The reasons of explaining the decreasing trend of the OMI-derived NO_2 VCDs can certainly apply to the reduction of NO_x emissions. The existing fifth-stage emission standards for powered two-wheelers in Taiwan is equal to Euro 3 standard effective in 1998, and Taiwan's sixth-stage standard, effective in 2017, is similar to the Euro 4 standards (Liang, 2014). In 2007, Taiwan initiated the CNS cold start cycle for 150 cc cycles with NO_x 0.15 g/km limit (MECA, 2014). The ban of 2-stoke motorcycles is a sensible approach to reduce NO_x emission. For that matter, reduction of NO_x from vehicles is the key to reduce NO_2 level since the secondary NO_2 formation caused by the titration reactions of NO with O_3 and peroxy radicals is responsible for the major fraction (approximately 70%) of the measured NO_2 in Germany as reported by Kurtenbach et al. (2012). Furthermore, use of electric vehicles in complete replacement of light duty vehicles can surely reduce ambient NO_2 level, e.g., reduction of 3.3 ppb NO_2, albeit with increase of 0.1 ppb SO_2 due to electricity generation (Li et al., 2016). Lastly, energy intensity in Taiwan had been reduced from 9.42 to 7.37 LOE/NT$1000 (1 USD ≈ 30 NT$) between 2005 and 2015, or 22% reduction (TBOE, 2016) – this indirectly also reduced NO_2 emission.

Since power plants emit large amounts of NO_x, Taipower (the monopoly power company in Taiwan) has made significant efforts in addressing pollution abatement. For example, Taipower has installed advanced low-NO_x burners on all new thermal units, utilizing selective catalytic reduction (SCR) that lowers the concentration of NO_x emissions, as well as adopting advanced Ultra Supercritical units for reducing both NO_x as well as CO_2 emission (Taipower, 2015). Also, it has employed automated combustion tuning technology and optimized loading for NO_x reduction. On the average, NO_x emission intensity in Taiwan was 327 kg/GWh in 2015 (Taipower, 2016), within the range of NO_x best available technology of 160–420 kg/GWh (Ito, 2011). The emission intensity reduction of 34% from 2005 level for NO_x (Taipower, 2007) is comparable to those of early stage (1998–2008)

Table 2
Emission of NO_x in three metropolitan cities with decreasing rate from 2005 to 2015.

City	NO_x emission			
	2005 (kt)	2015 (kt)	Annual reduction rate (kt/yr)	Percentage reduction from 2005 to 2015 (%)
Taipei	17.9	8.9	0.89 ± 0.10 ($R^2 = 0.98$)	50
Taichung	8.0	5.7	0.20 ± 0.06 ($R^2 = 0.86$)	29
Kaohsiung	44.1	27.8	1.69 ± 0.18 ($R^2 = 0.98$)	37
Taiwan	534.5	344.3	18.7 ± 2.97 ($R^2 = 0.96$)	36

efforts in UK and Canada (Ito, 2011). In the future, it is expected that some of the existing coal-fired power generators (32% of all power plants) will be replaced with natural gas to further reduce NO_x. A recent study has indicated that by replacing 8–10 existing coal-fired power plants with modern natural gas combined cycle units in the Electric Reliability Council of Texas region, significant reductions of NO_x by 51–55% would be expected (Peer et al., 2016). Furthermore, setting much stringent emission standards for existing coal-fired plants (currently, NO_x is 144 mg/m^3 (Pan, 2016), retrofitting SCR and electrostatic precipitator as well as using clean fuel can reduce NO_x emissions. It is noted that often the reduction of NO_x emission can increase the ozone concentration unless volatile organic compound (VOC) emissions are also reduced (Huang et al., 2001).

Remarkably, there is an excellent correlation between the NO_x emission and the OMI NO_2 columns ($R^2 = 0.82$ for Taipei, 0.78 for both Taichung and Kaohsiung and 0.79 for the entire Taiwan) as shown in Fig. 5. The NO_x emission data were obtained from governmental document (TEDS 9.0; https://teds.epa.gov.tw/new_main2-0-1.htm). The annual NO_x emission and OMI NO_2 data show consistency between emission rates and NO_2 air quality. Good agreement between OMI NO_2 observations and emission rates were also reported in China in 2005–2015 (Liu et al., 2016). Unlike the study by Tong et al. (2015) reporting that % reduction in the OMI-derived NO_2 VCD was higher than that of NO_x emission in eight major cities in the US, the OMI NO_2 VCD decreasing trend from 2005 to 2015 in the present study (24% in Taipei, Table 1) lags behind the corresponding NO_x emission reduction (Table 2; 50% in Taipei) as also reported in the EU (EEA, 2015) and the US (https://www.epa.gov/air-trends/nitrogen-dioxide-trends#nonat). The regional transport should have contributed to the observed NO_2 level, although the exact extent of its contribution was not quantified. However, NO_x emission from diesel vehicles may be the main cause (EEA, 2015). Nonetheless, the pollutant emission reduction rate does not always yield the same OMI derived VCD reduction (e.g., Hilboll et al., 2013) due to complex atmospheric reaction, meteorological conditions, and LRT impact as also reported by Guerreiro et al. (2014) who state that there is an apparent mismatch between the trends in the anthropogenic emission of precursors and the observed trends (or lack of trends) in ozone.

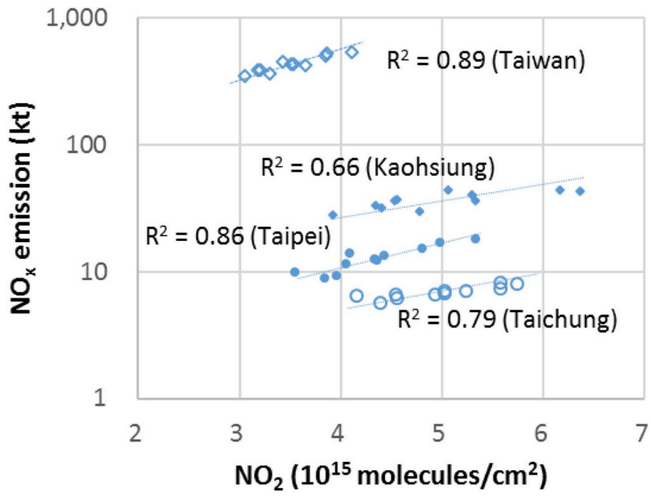

Fig. 5. Regression of annual OMI NO_2 VCDs with the corresponding annual emissions for three cities and Taiwan.

Again, this study also showed that the entire Taiwan and five cities were significantly affected by LRT. In particular, most OMI NO_2 VCDs were from LRT in Hualien which is located in the east cost of Taiwan.

As mentioned before, Taipei and Kaohsiung had the similar NO_2 VCDs in 2015, yet with different emission amounts (Table 2). In general, the stacks in the cluster of petrochemical plants and three power plants near Kaohsiung are high enough (typically 130–250 m height used in coal-fired steam plant while 60–130 m used in natural gas plant) that emitted pollutants may be blown to ocean under right weather conditions and not reach the areas close to the factories. In addition, the ocean surrounding Kaohsiung contains some significant NO_2 VCDs as clearly shown in the satellite OMI image (Fig. 3, middle plot). The detailed emission transport along with the shipping emission in Kaohsiung harbor away from local emission sources into ocean requires further modelling study.

3.4. *Comparison with ground monitoring data*

It is interesting to find whether there is any correlation between original monthly OMI NO_2 columns and ambient NO_2 concentrations derived from ground-based monitoring stations (for details of these monitoring stations see Lee et al., 2018). For ground-based NO_2 data, a total of thirteen ground monitoring stations were selected; four stations in Kaohsiung, two in Taichung, five in Taipei, and one each in Douliu and Hualien. It must be noted that the data from the ground stations cannot represent the overall air quality of these cities; they can only represent the average concentration of the surrounding area of each ground monitoring station. Thus we select three cities and performed correlation analysis. In the three cities evaluated, Kaohsiung shows very high correlation between original monthly OMI and ground-based NO_2 ($r = 0.84$, Fig. 6c) with moderate correlation in Taipei and Taichung ($r = 0.60$ and 0.59, respectively; Fig. 6a and b). This study also compared the decreasing trends of surface NO_2 level and OMI NO_2 columns (Table SM-6). In short, the percentage reduction of ground NO_2 (data from Lee et al., 2018) and that of the OMI NO_2 columns in the three city areas were very consistent. In the case of Kaohsiung, for example, a high percentage reduction both in the ground NO_2 and the OMI NO_2 columns was observed: 27% reduction for ground NO_2 and 37% for the OMI NO_2. The high reduction is definitely attributed to the continuously applied environmental regulations and policies.

3.5. *Correlation*

In this section, we wanted to investigate if the original monthly OMI NO_2 VCDs of different cities would be similar or not and if their values could be correlated. The original monthly NO_2 data show good correlation among 4 cities and entire Taiwan with correlation coefficients (r) ranging from 0.70 to 0.95 (Table 3). Such good correlation indicates similar NO_x emission sources (e.g., power plants and industrial stationary sources as well as from mobile sources). The good correlation was also reported elsewhere (e.g., Guo et al., 2010; Zhang et al., 2014;

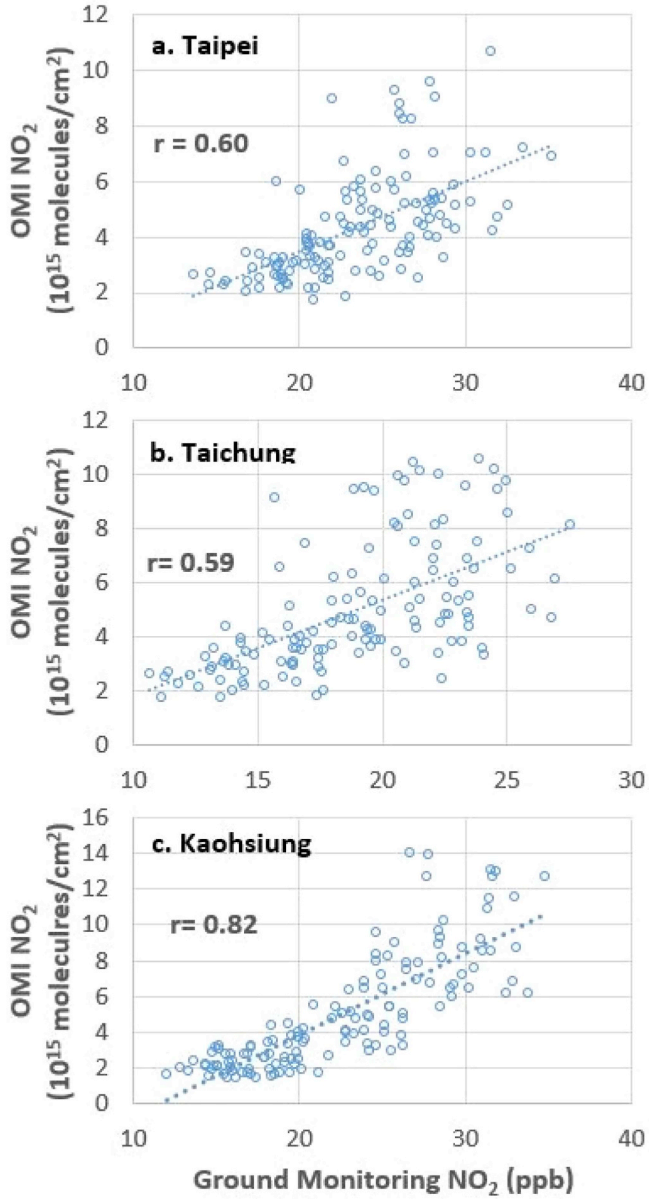

Fig. 6. Regression analysis of original monthly OMI tropospheric NO_2 column values and ground monitoring NO_2 concentrations for the three cities.

Table 3
Cross-correlation of NO_2 for the studied cities.

	Taipei	Taichung	Kaohsiung	Douliu	Taiwan
Taipei	1.00	0.71	0.70	0.77	0.84
Taichung	0.71	1.00	0.93	0.91	0.95
Kaohsiung	0.70	0.93	1.00	0.93	0.93
Douliu	0.77	0.91	0.93	1.00	0.96
Taiwan	0.84	0.95	0.93	0.96	1.00

$p < 0.05$.

Hassoon, 2015), whereas negative correlation was noted (Sarnat et al., 2001; Bralić et al., 2012).

4. Conclusion remarks

The present study used the retrieved OMI NO_2 column to evaluate changes for the entire Taiwan as well as 5 cities. For the 10-year span, the OMI tropospheric NO_2 VCDs observed in Taiwan and in 5 cities all indicate decreasing trend with following order: Kaohsiung ($0.19 \pm 0.03 \times 10^{15}$ molecules/cm^2/yr) > Taichung > Taipei > Douliu > Taiwan > Hualien ($0.013 \pm 0.013 \times 10^{15}$ molecules/cm^2/yr). The distribution of OMI NO_2 VCDs in 2015 follows the order of Taichung ($4.4 \pm 2.4 \times 10^{15}$ molecules/cm^2) > Kaohsiung > Taipei > Douliu > Taiwan > Hualien ($1.7 \pm 0.7 \times 10^{15}$ molecules/cm^2). The significant correlation of original monthly NO_2 pollutant among 4 cities (3 metropolitan cites and Douliu city) and the entire Taiwan indicates similar NO_x emission sources in the western part of Taiwan.

The significant decrease in OMI NO_2 VCDs during the 10-yr duration is due to effective control strategies for both stationary and mobile sources. This study also indicates that original monthly satellite-based OMI NO_2 and ground-based NO_2 levels exhibit high/moderate correlation in 4 cities and in Taiwan. Although this paper only addresses control policy for NO_2, the adequate and appropriate approaches towards enhancing overall air quality should be the key with cost effectiveness for all other pollutants (NO_2, SO_2, VOCs, NH_3, CO, etc.).

Banning 2-stoke motorcycles is the sensible way to reduce mobile emission sources. Further, use of electric vehicles can reduce ambient NO_2 level but it requires governmental policy regarding the installation of convenient charging stations. The emission standards for commercial vehicles should be further upgraded such as those used in developed countries (e.g. emission factor below 1 g NO_x/kWh; Zhu et al., 2015). Recently, government has announced the policies/measures including different incentives to reduce annual $PM_{2.5}$ level from 22 to 18 μg/m^3 by 2019. In doing so, the ambient NO_2 concentration will be also reduced. In addition, use of clean fuel in lieu of conventional bituminous coal and petroleum coke at factories and power plants is the policy that the central and local governments are now actively pursuing. Lastly, monitoring stations near roadside and power plants should be expanded to identify hot spots and to see the impact of mobile/stationary sources towards overall ambient NO_2 concentration.

One step further is setting emission ceiling and/or minimum emission reduction goal in 2020 for NO_x emission in air pollution sensitive areas, as in the case of EU policy. Although cost associated with further reduction is high, but the end results for achieving much better air quality is worthwhile for pursuing. The government should provide financial incentives to assist industry to install and update energy-saving, low-pollution facilities to achieve the necessary emission reduction.

The satellite data can be easily used for evaluating long term trend analysis and extent of any reduction or increase. Furthermore, the data can be used for observing any changes in pollutants in hot spots, e.g., NO_2 near power plants and high volume traffic areas. The data can also be used along with meteorological information for tracking episode events – its plume formation, pathway and eventual disappearance and their effect on the formation of nitrate in the size-resolved particles. Since Taiwan is surrounded by sea, the LRT of pollutants from outside and the extent of local emission been transported out of the island can be also evaluated via easily retrievable satellite data.

Conflicts of interest

The authors declare no conflict of interest.

Acknowledgments

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.apr.2019.01.004.

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