



Article

Analysis of Tropospheric Nitrogen Dioxide Using Satellite and Ground Based Data over Northern Thailand

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Abstract. Tropospheric NO₂ columns over northern Thailand were analyzed using monthly satellite products of the SCIAMACHY, OMI, GOME-2A, and GOME-2B sensors for the 14-year period 2003–2016. The comparative results of the four pairs of different satellite datasets within overlapped years showed that they were well correlated with correlation coefficients (r) of 0.85–0.90 when the analysis was considered only during the dry period. Ground measurements of NO₂ concentrations were also obtained for comparative analysis with the satellite NO₂ columns. The results revealed relatively good agreement between these two parameters for a seasonal pattern. High levels of NO₂ were detected during January–April. Overall, the r -values of the satellite and ground datasets during the dry period were in the range 0.54–0.81. Moreover, most satellite and ground datasets recorded greater levels of NO₂ in the afternoon than in the morning corresponding with the number of fire hotspots. Satellite and ground measurements showed slightly increasing trends of NO₂ levels during dry months for 2010–2016 with values of 7.23% and 0.48%, respectively, over the 6-year period. The results in this study suggest that integrating satellite and ground data would provide valuable information for air quality managers to better understand the spatio-temporal distribution of NO₂ which is essential for setting air quality policy and mitigation plan in Thailand.

Keywords: NO₂, GOME-2, OMI, SCIAMACHY, MODIS, fire hotspots.

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1. Introduction

Nitrogen dioxide (NO_2) is one of the important trace gases playing significant roles in atmospheric chemistry and climate change. In terms of greenhouse gas, NO_2 contributes significantly to the local radiative forcing of climate over polluted regions (such as large urban areas or areas with extensive biomass burning) by perturbing methane (CH_4) and ozone (O_3) concentrations [1]. In the troposphere, when NO_2 is present in sunlight, O_3 is formed as a result of the photolysis of NO_2 [2]. NO_2 can also generate secondary inorganic aerosol and secondary organic aerosol through a series of chemical reactions and physical processes [3]. In addition, the formation of nitric acid (HNO_3 the main component of acid rain) due to the chemical reaction of NO_2 and hydroxyl radicals (OH) is the dominant sink of NO_2 in the troposphere during daytime and summertime [4]. The presence of NO_2 in the troposphere also causes adverse impacts on human health, crops and ecosystems [5–6]. The main sources of NO_2 include anthropogenic activities such as fossil-fuel combustion, biomass burning and fertilizer application, as well as natural sources such as wildfires, lightning and microbiological processes in soil [7–9]. The chemical lifetime of tropospheric NO_2 is relatively short varying from hours to days in the atmospheric boundary layer depending on the season [10].

Advances in satellite instruments have allowed access to information on the global distribution of atmospheric NO_2 over the past decades. Many researchers [11–15] have applied satellite-retrieved data of tropospheric NO_2 columns for air quality study since the Global Ozone Monitoring Experiment (GOME) onboard the ERS-2 platform was launched in 1995. The next generation of satellite NO_2 instruments was the SCanning Imaging Absorption spectroMeter for Atmospheric CHartographyY (SCIAMACHY) onboard Envisat in 2002, followed by the Ozone Monitoring Instrument (OMI) onboard Aura in 2004, and GOME-2 onboard MetOp-A (GOME-2A) and MetOp-B (GOME-2B) in 2006 and 2012, respectively. Boersma et al. [16] validated urban NO_2 concentrations retrieved from OMI and SCIAMACHY using ground in situ measurements in Israeli cities in 2006. Both satellite and ground observations showed higher winter NO_2 levels than for summer. In addition, SCIAMACHY recorded higher NO_2 levels than OMI in summer, but lower in winter. Hilboll et al. [17] investigated long-term trends (1996–2011) of tropospheric NO_2 over megacities using multisatellite observations. They found significantly decreasing trends in the developed world such as Western Europe, the United States, and Japan. However, strongly increasing trends of 5–10% per year were found over China, India, and the Middle East. Similar results were reported in Schneider et al. [18] using SCIAMACHY to analyze the trends of tropospheric NO_2 over 66 large urban agglomerations worldwide in 2002–2012. Chen et al. [6] compared products from different satellites (GOME, OMI, SCIAMACHY, GOME-2A and GOME-2B) across China for 1995–2015. The NO_2 products generally had good agreement regarding the spatial pattern. They also found an increasing trend of tropospheric NO_2 columns for 1997–2011 but a decreasing trend for 2011–2015. The most important sources of error in retrievals of tropospheric NO_2 columns are associated with cloud parameters, surface albedo and NO_2 vertical profiles [19]. Irie et al. [20] estimated the biases in the tropospheric NO_2 column data from SCIAMACHY, OMI, and GOME-2A in East Asia from 2006–2011. They concluded that the biases are all small (less than about 10%) and insignificant. They also suggested that these small biases allow for analyses combining these satellite data for air quality studies. Because of the short lifetime of NO_2 in the troposphere, NO_2 has its largest concentrations close to the emission sources, making satellite measurements of NO_2 columns closely correlated to ground-level NO_2 concentrations or NO_x emissions [17, 21]. Han et al. [22] evaluated NO_x emission fluxes from three inventories (INTEX-B, CAPSS, and REAS) by comparing modeling (CMAQ) results with OMI observations over East Asia in 2006. Many studies [21, 23–26] have also applied satellite observations for model validation. Bechle et al. [27] applied modeled NO_2 vertical profiles to convert OMI tropospheric NO_2 columns to ground-level NO_2 concentrations. They found that using modeled NO_2 vertical profiles provided little improvement in ground-level NO_2 estimates.

In Thailand, the northern region experiences air pollution haze episodes from forest fires and vegetation fires for land clearing each year during the dry season (January–April). During this period (especially during March–April), air pollutants such as particulate matter with diameter less than 10 micron (PM_{10}), particulate matter with diameter less than 2.5 micron ($\text{PM}_{2.5}$), carbon monoxide (CO), and NO_2 , have been reported at higher levels compared to other months [26, 28–31]. The mountain ranges surrounding the northern region create a narrow valley which is perfect for trapping air pollutants over the cities causing even higher levels of air pollutants. Moreover, a larger urbanized city such as Chiang Mai is also affected by air pollution because of its increasing traffic congestion [30, 32]. Lalitaporn et al. [26] found high concentrations of NO_2 in Chiang Mai during biomass burning season of March–April which agreed well with PM_{10} and CO concentrations. Similarly, Pimpunchat et al. [33] presented high concentrations of PM_{10} during March–April in Chiang Mai

owing to haze situation. They also reported high correlation of PM₁₀ concentrations versus NO₂, CO, and O₃ concentrations. Wiwatanadate [34] studied the acute effects of open burning-related air pollution in Chiang Mai during January–April of 2008. They found that NO₂ was significantly associated with many symptoms including nosebleed, larynx symptoms, dry cough, lower lung symptoms, heart symptoms, and eye irritation. Ground monitoring stations for NO₂ and other criteria air pollutants were setup in the northern Thailand by the Pollution Control Department (PCD), Thailand. However, most of them are located in the center of the cities, while they are still sparse in the remote areas where forest fire events generally occur. Therefore, satellite observed data could be useful to assess air pollution information for the areas that lack of ground monitoring stations.

Since NO₂ is one of the criteria air pollutants and can be measured globally by satellite sensors, the current study used monthly products of four satellites (SCIMACHY, OMI, GOME-2A, and GOME-2B) to analyze the spatio-temporal variability of tropospheric NO₂ columns over northern Thailand for 2003–2016. The comparison between different satellite products was conducted to investigate diurnal variation and its potential use for the long-term study of tropospheric NO₂. Ground measurements of NO₂ concentrations and satellite-observed fire hotspots were also compared with satellite NO₂ data to assess the consistency between them.

2. Materials and Methods

2.1. Ground Monitoring Data

This study focused on the analysis of NO₂ levels in the upper part of northern Thailand. Ground monitoring data was based on hourly NO₂ concentrations collected by PCD, Thailand over 10 stations in six cities (Chiang Mai, Lamphun, Lampang, Nan, Phrae, and Phayao) in northern Thailand for 2003–2016. Table 1 and Fig. 1 present the locations of the PCD monitoring stations considered in this study. The PCD reference method used for measuring NO₂ concentrations is chemiluminescence.

Table 1. Locations of ground monitoring NO₂ stations and satellite grid boxes.

Satellite grid box		Ground monitoring station			
ID	0.5×0.5° box (latitude, longitude)	ID	Station name	City	Available year
1	18.50–19.00°N, 98.75–99.25°E	1	Chiang Mai City Hall	Chiang Mai	1996–present
		2	Yupparaj Wittayalai School	Chiang Mai	1996–present
		3	Provincial Administrative Stadium	Lamphun	2009–present
2	18.00–18.50°N, 99.50–100.00°E	4	City Pillar Shrine	Lampang	1996–present
		5	Health Promotion Hospital Sob Pad	Lampang	1996–present
		6	Health Promotion Hospital Ta See	Lampang	1996–present
		7	Provincial Waterworks Authority Mae Moh	Lampang	1996–present
3	18.50–19.00°N, 100.50–101.00°E	8	Municipality Office	Nan	2009–present
4	18.00–18.50°N, 100.00–100.50°E	9	Meteorological Station	Phrae	2010–present
5	19.00–19.50°N, 99.75–100.25°E	10	Knowledge Park	Phayao	2010–present

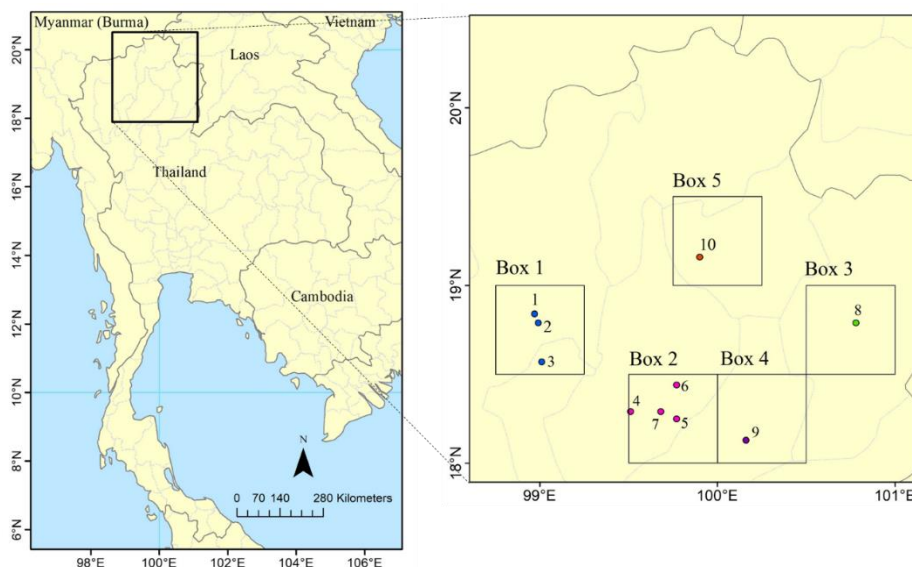


Fig. 1. Locations of 10 PCD stations and 5 satellite grid boxes.

2.2. Satellite Observation Data

The satellite NO_2 data were retrieved as monthly mean tropospheric NO_2 columns (unit: molecule per cm^2) from level-2 products of SCIAMACHY, OMI, GOME-2A, and GOME-2B for 2003–2016. OMI [35–36] and GOME-2A/B [19, 37] provide nearly global coverage in one day with a spatial resolution of $13 \times 24 \text{ km}^2$ and $80 \times 40 \text{ km}^2$, respectively. For SCIAMACHY [19, 38], global coverage requires 6 days with a spatial resolution of $60 \times 30 \text{ km}^2$. The original satellite retrievals are regridded on a regular $0.125^\circ \times 0.125^\circ$ grid for OMI and on a regular $0.25^\circ \times 0.25^\circ$ grid for SCIAMACHY, GOME-2A, and GOME-2B by averaging the satellite observations weighted by the size of the overlapping surface area [18]. SCIAMACHY, GOME-2A, and GOME-2B have a satellite overpass local time (LT) in the morning of approximately 9:30 LT for SCIAMACHY and 10:30 LT for GOME-2A/B. While, OMI overpasses in the afternoon of approximately 13:45 LT. Table 2 summarizes the characteristics of the satellite products used in this study. All satellite NO_2 data were downloaded as monthly products from the website of the Tropospheric Emission Monitoring Internet Service (TEMIS) of the European Space Agency (ESA) (<http://www.temis.nl/index.php>).

Data for tropospheric NO_2 columns in this study were collected for the following periods:

- SCIAMACHY: 01/2003–03/2012
- OMI: 10/2004–12/2016
- GOME-2A: 01/2007–12/2012
- GOME-2B: 01/2013–12/2016

Two-year data of fire hotspots retrieved from the Moderate Resolution Imaging Spectroradiometer (MODIS) onboard the Terra (10:30 LT) and Aqua (13:30 LT) platforms were collected for analysis in this study for 2015–2016. The MODIS fire products of Terra and Aqua detect fires in 1-km pixels that are burning at the time of satellite overpasses. The data are available at the website of Geo-Informatics and Space Technology Development Agency (GISTDA) (<http://fire.gistda.or.th/>). More details of MODIS fire products are available in Justice et al. [39].

Table 2. Characteristics of satellite NO_2 products.

Satellite sensor	SCIAMACHY	OMI	GOME-2
Platform	Envisat	Aura	MetOp-A/B
Spectral range (μm)	0.23–2.38	0.27–0.50	0.24–0.79
Ground resolution (km^2)	60×30	13×24	80×40
Overpass local time	10:00	13:45	09:30
Operating time	Mar 2002–Apr 2012	Jul 2004–present	Oct 2006–present/ Sep 2012–present

2.3. Data Analysis

First, the comparison between four satellite products of tropospheric NO₂ columns was conducted based on simple linear regression analysis to investigate the consistency between them. Since the four satellites considered in this study have different ground resolutions (regridged on a 0.125°×0.125° grid for OMI and on a 0.25°×0.25° grid for SCIAMACHY, GOME-2A, and GOME-2B), five 0.5°×0.5° grid boxes were set up around ground monitoring stations of interest. All satellite pixels falling within the decided grid boxes were then extracted using Python version 2.7 for the comparison among satellite products. Table 1 and Fig. 1 present the locations and details of the five grid boxes used in this study. Furthermore, fire hotspots within the considered area were also retrieved from MODIS-Terra and -Aqua and compared with each other.

The comparative analysis between satellite and ground NO₂ data was performed based on 3-hour averages of ground NO₂ concentrations covering the satellite overpass times. Hourly ground NO₂ concentrations were collected for 9:00–12:00 LT and for 12:00–15:00 LT corresponding to the satellite overpass times in the morning (9:30 LT for GOME-2A/B and 10:00 LT for SCIAMACHY) and afternoon (13:45 LT for OMI), respectively. Collected ground data were then averaged as monthly and yearly means for comparison with the satellite data. All ground data falling within the same 0.5°×0.5° grid boxes as satellite data were grouped and averaged for comparison with satellite data for concurrent locations and time periods. It is noteworthy that ground measurements of NO₂ concentrations represent point-based data at a single location whereas satellite data represent spatial averages of NO₂ over a specific land area. In addition, ground NO₂ data are time averaged (in this case are 3-hour averaged) whereas satellite NO₂ data are measured at a single time when the satellite overpass. Therefore, even if both measurements are error-free, there could be uncertainty in the comparison between ground and satellite datasets owing to the mismatch of spatial and temporal averaging [27, 40]. Finally, spatial distributions of tropospheric NO₂ columns from four satellites were also depicted for the northern region of Thailand by using ArcMap version 10.3.1.

During the study period, satellite monthly products of NO₂ included more data gaps than monthly data of PCD NO₂. According to data collection periods, SCIAMACHY, OMI, GOME-2A, and GOME-2B had missing data (calculated from total possible data of all boxes) of 19.82% (from 555), 0.54% (from 735), 5.00% (from 360), and 1.67% (from 240), respectively. Monthly PCD data averaged for each box in the morning (9:00–12:00) had no missing data, and in the afternoon (12:00–15:00) had missing data of 0.34% (from 586). Outlier treatment for satellite and ground data was also considered before performed the data analysis. However, satellite and ground data that had high values generally revealed together during haze episodes which implied large emissions from fires. Thus, these data were kept for the analysis.

3. Results and Discussion

3.1. Comparisons among Satellite Products

Data consistency among the tropospheric NO₂ columns from the four satellite products was analyzed. Four pairs of different monthly satellite datasets (OMI versus SCIAMACHY, GOME-2A versus SCIAMACHY, OMI versus GOME-2A, and OMI versus GOME-2B) within overlapping years at the concurrent locations (box ID 1–5) were compared based on simple linear regressions and the results are presented in Fig. 2(a). The results showed a relatively good correlation between the different datasets with the *r*-values in the range 0.82–0.88 (at the significance level (*p*) < 0.001). The results were slightly better when the analysis was focused only on the 7 months of the year (October–April) covering the dry period in Thailand, with the *r*-values improving to 0.85–0.90 (*p* < 0.001) as presented in Fig. 2(b). The slope of the comparison between tropospheric NO₂ columns retrieved from SCIAMACHY and GOME-2A was close to 1.00 (slope = 1.05) showing good agreement between their datasets. This can be explained by their similarities in satellite overpass times and spatial resolutions. Chen et al. [6] also reported similar results across China with slightly higher levels of tropospheric NO₂ columns from SCIAMACHY compared to GOME-2A (slope = 1.18). The regression analysis between OMI and other satellite datasets indicated higher levels of tropospheric NO₂ columns from OMI than those from SCIAMACHY, GOME-2A, and GOME-2B with slopes in the range 0.69–0.70. The discrepancy in this case was caused mainly by the differences in satellite overpass times as OMI collects data in the afternoon (13:45 LT) while SCIAMACHY, GOME-2A, and GOME-2B collect data in the morning (9:30–10:30 LT). According to previous studies [41–42], the major source of nitrogen oxide or NO_x (mainly NO₂ and NO) emissions in Thailand and Southeast Asia is road transportation. High

concentrations of NO_2 are generally produced during the morning rush hour from transportation, with the concentration reducing in the afternoon due to the higher photolysis rate [21]. However, in this case, OMI recorded higher levels in the tropospheric NO_2 columns, implying that there were higher emissions of NO_2 from activities in the afternoon in the northern Thailand. Figure 3 displays the number of monthly total fire hotspots retrieved from MODIS-Terra and -Aqua in the northern region for 2015–2016. During January–April, specifically in March, notably larger numbers of fire hotspots were detected by MODIS-Aqua (13:30 LT) than MODIS-Terra (10:30 LT). A similar study [43] used the MODIS fire hotspots from 2002–2010 for analysis of fire events in the Asian region. They reported that MODIS-Aqua captured 70% of the fires relative to MODIS-Terra. Arunrat et al. [28] presented times of the day for burning crop residues in Chiang Mai. They found that most of the farmers burned the crop residues (67% of grain maize and seed maize residues and 86% of crop residue from the integrated farming system) in the afternoon (12:01–18:00 LT) compared to in the morning (06:01–12:00 LT) and in the evening (18:01–06:00 LT). A previous study [44] also demonstrated that over tropical biomass-burning regions, SCIAMACHY recorded lower tropospheric NO_2 columns than OMI (up to 40%). This suggests that biomass-burning activities in the afternoon can be an important cause of the greater levels of tropospheric NO_2 columns observed by OMI in the northern Thailand. However, further analysis on diurnal variation in biomass burning emission in each area of the northern region would provide stronger conclusion. The discrepancy among these satellite products can also be from the biases in satellite retrieved data. However, in accordance with Irie et al. [20], the biases in these datasets (SCIAMACHY, OMI and GOME-2A products) are small and insignificant. Furthermore, from Fig. 2(b), it is noticeable that the highest levels of the tropospheric NO_2 columns were recorded in March which were related to the number of fire hotspots. Besides source emission from biomass-burning activities, meteorology also plays a key role in contributing to high NO_2 levels during February–April. In particular, stagnant air condition, radiative inversion (associated with calm wind, clear sky and low mixing height) and subsidence inversion (influenced by the northeast monsoon with cold dry air) are prevalent in the northern region during these months [32]. These weather conditions can restrict the dispersion of air pollutants and then subsequently enhance the built-up of high NO_2 levels as well.

For the wet period of May to September, the correlation between the four satellites datasets were poor with low r -values of 0.29–0.49 as presented in Fig. 2(c). In general, the presence of clouds can introduce uncertainties in satellite observations of tropospheric trace gases [19, 45]. Kim Oanh and Leelasakultum [32] showed that it is less cloudy during the dry months in the northern region of Thailand. This perhaps causes better correlation of tropospheric NO_2 columns between different satellites datasets in the dry period than in the wet period. In addition, the levels of tropospheric NO_2 columns during the wet period were lower than in the dry period which also matched with the seasonal trend of the number of fire hotspots. The monthly mean tropospheric NO_2 columns of all considered boxes for each satellite at different periods are illustrated in Fig. 4. Overall, all four satellites recorded higher levels of tropospheric NO_2 columns during the dry period with the peak in March. For the morning satellites, the analysis showed that GOME-2B (2013–2016) recorded the highest levels of tropospheric NO_2 columns followed by GOME-2A (2007–2012) and SCIAMACHY (2003–2011), respectively. This suggests an increasing trend in NO_2 in the northern region. For the afternoon observations, the levels of tropospheric NO_2 columns from OMI (2005–2016) were clearly higher than those from the other satellites in March. This highlights the intensive biomass-burning activities in the afternoon during this month.

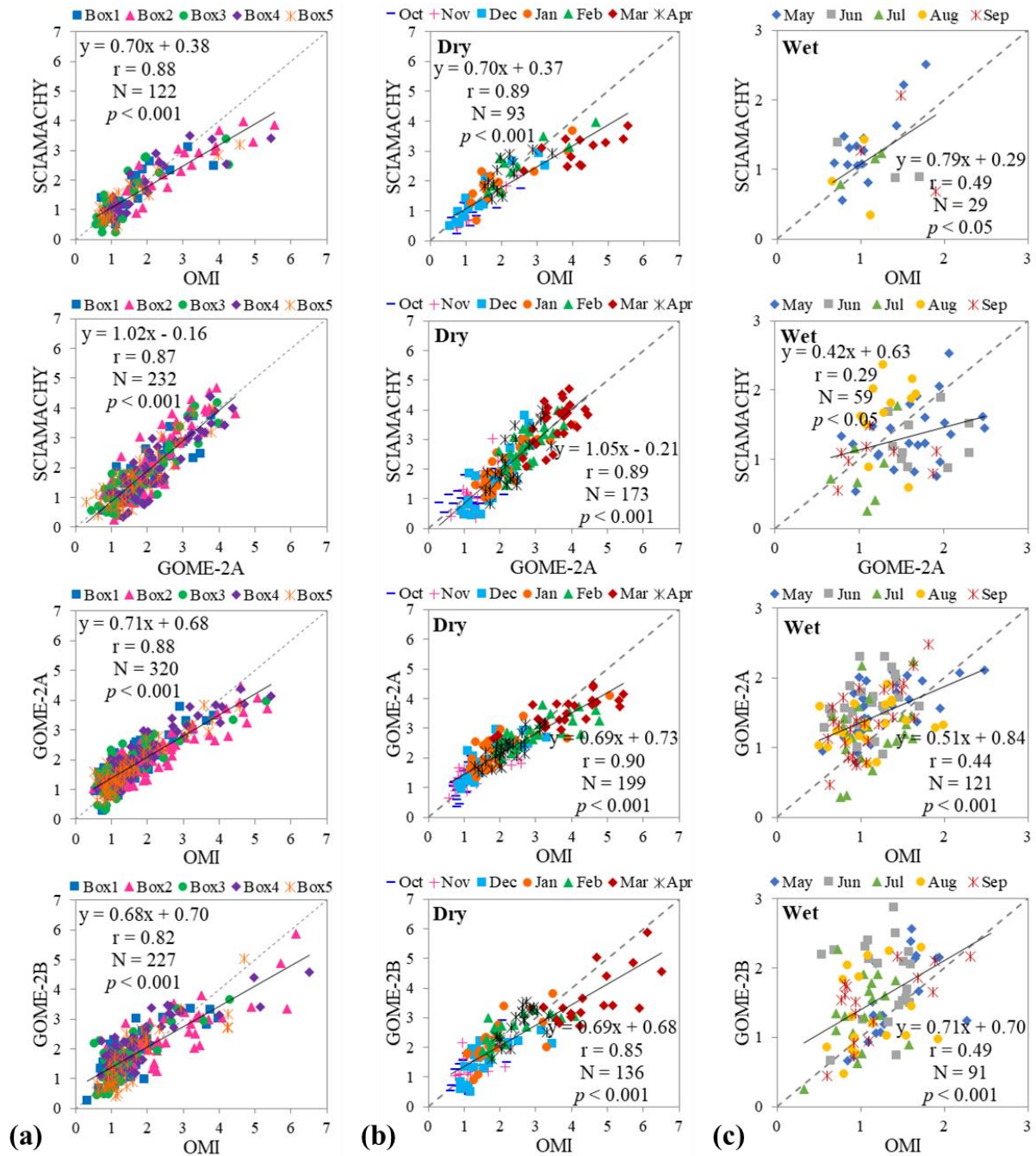


Fig. 2. Scatter plots of tropospheric NO₂ columns (10¹⁵ molecule per cm²) extracted from 5 satellite grid boxes (box ID 1–5) of OMI versus SCIAMACHY, GOME-2A versus SCIAMACHY, OMI versus GOME-2A, and OMI versus GOME-2B for all months (a), for dry months (b), and for wet months (c).

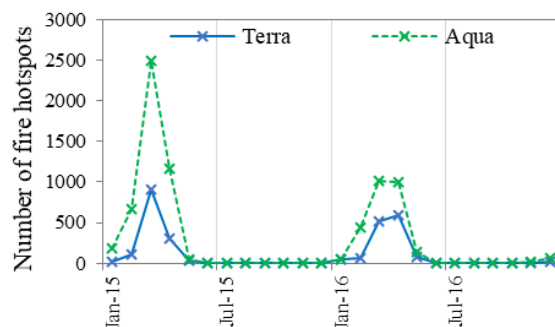


Fig. 3. Monthly total fire hotspots of MODIS-Terra and MODIS-Aqua.

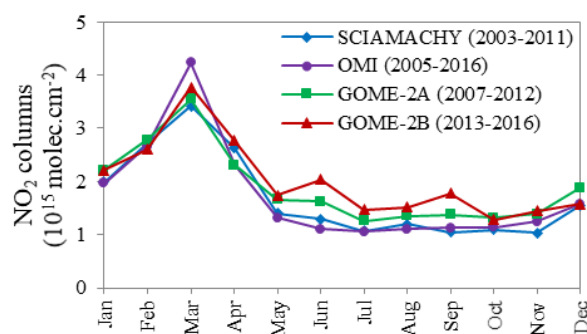


Fig. 4. Monthly mean satellite observations of tropospheric NO₂ columns.

3.2. Monthly Analysis of Satellite and Ground NO₂

Tropospheric NO₂ columns were compared with ground NO₂ concentrations for the 14-year period 2003–2016. Figure 5 presents the monthly series of averaged tropospheric NO₂ columns retrieved from the three morning satellites (SCIAMACHY, GOME-2A, and GOME-2B) and the averaged ground NO₂ concentrations (9:00–12:00 LT). Figure 5(a–e) present the comparisons for boxes with ID 1–5, respectively, and Fig. 5(f) presents the average of all five boxes. Overall, the morning satellite observations were able to capture the seasonal variations of ground NO₂ concentrations. The highest levels of NO₂ from both satellites and ground monitoring stations were from January–April with the maximum peak in March for all five boxes. Similar results are presented in Fig. 6(a–f) for the monthly series of tropospheric NO₂ columns from the afternoon satellite (OMI) plotted with ground NO₂ concentrations (12:00–15:00 LT). Table 3 summarizes the results of the correlation analysis between satellite and ground data for all boxes for both the morning and afternoon cases. Overall results showed the *r*-values in the range 0.43–0.80 ($p < 0.001$). When the analysis was restricted to only on dry months (October–April), the *r*-values ranged from 0.54–0.81 ($p < 0.001$). Most of the *r*-values in all boxes improved, except for Box 2 for both the morning and afternoon cases. As mentioned in the previous section, one of the main sources of error in determining of tropospheric NO₂ columns is cloud parameters and low cloudiness was generally observed during dry months in the northern Thailand. The reason for the better results during the dry months probably due to the less error in the satellite tropospheric NO₂ columns in the dry months than in the wet months. Moreover, low mixing height during dry period, especially during February–April [32] can also cause better correlation since when the mixing height is low almost all NO₂ are present close to the ground and the satellites will measure almost the same amount of NO₂ as measured by ground instruments.

Both satellite and ground NO₂ datasets had minimum levels during the wet period. A previous study [46] reported high relative humidity in the northern Thailand during May to October. During these months, more rain and higher wind speed dominated by the southwest monsoon enhance the dilution and dispersion of air pollutants. With the presence of sunlight, water vapor in the atmosphere will produce OH radicals which subsequently react with NO₂ and cause a sink of NO₂ in terms of HNO₃ [4]. Furthermore, biomass-burning activities during this period are also commonly at a reduced level. For these reasons, the NO₂ levels measured during the wet months were lower than in other months.

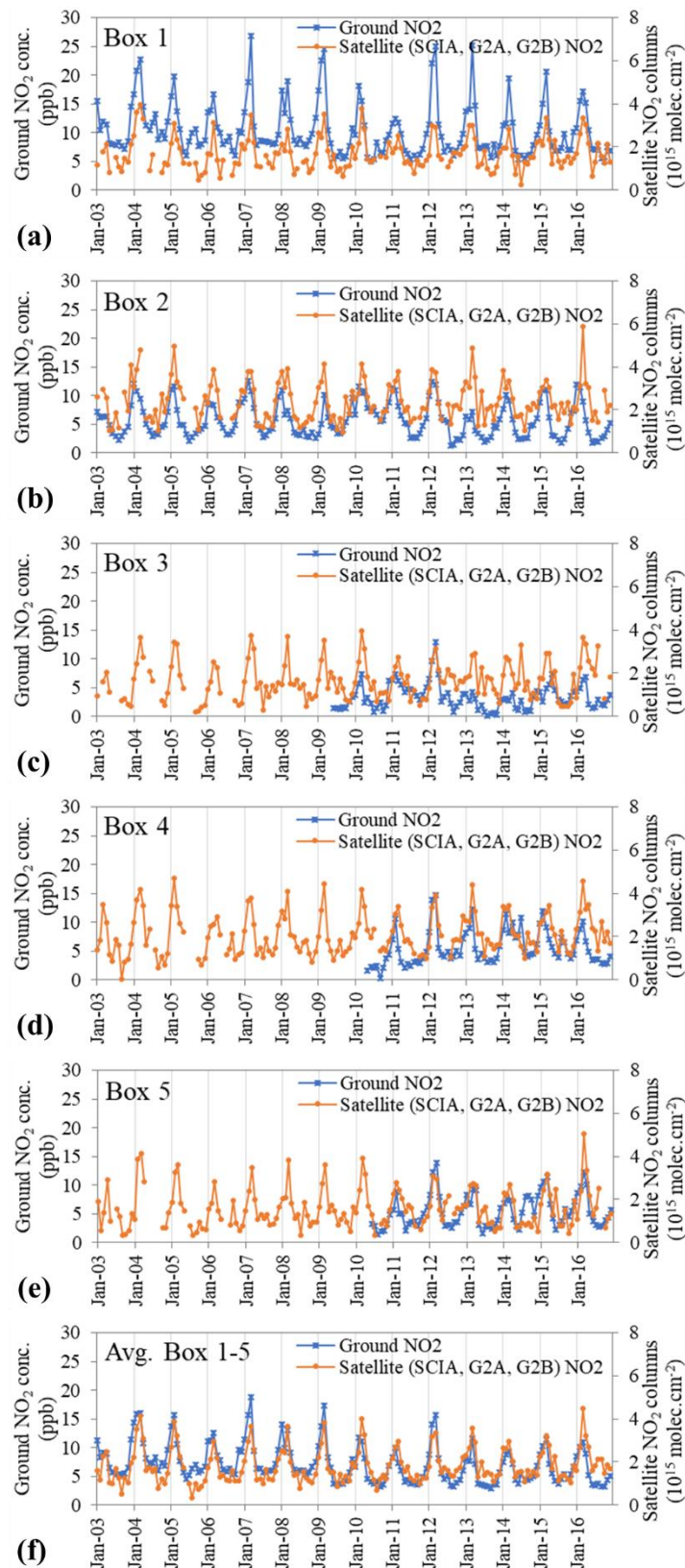


Fig. 5. Time series of morning satellite (SCIAMACHY, GOME-2A, and GOME-2B) NO_2 columns and ground NO_2 concentrations (9:00-12:00 LT) for box ID 1 (a), box ID 2 (b), box ID 3 (c), box ID 4 (d), box ID 5 (e), and all 5 boxes average (f).

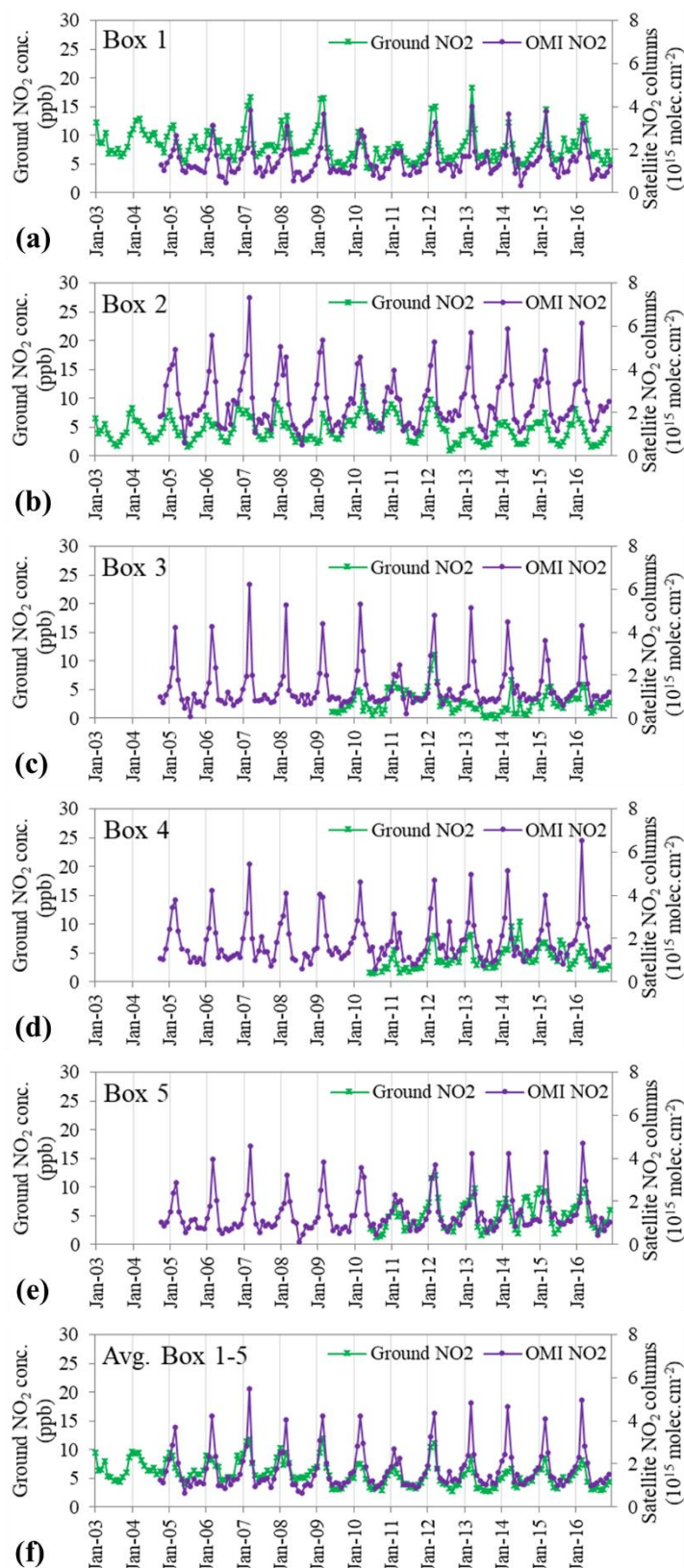
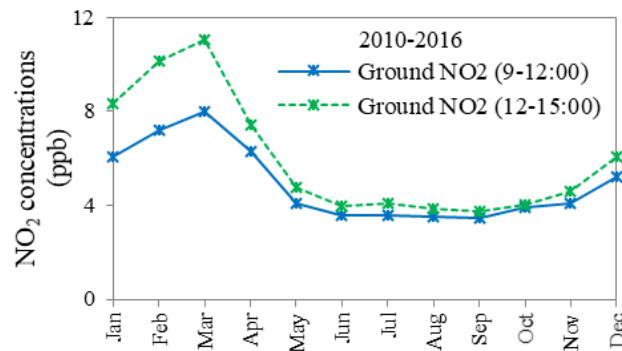


Fig. 6. Time series of afternoon satellite (OMI) NO₂ columns and ground NO₂ concentrations (12:00-15:00 LT) for box ID 1 (a), box ID 2 (b), box ID 3 (c), box ID 4 (d), box ID 5 (e), and all 5 boxes average (f).

Table 3. Correlation coefficients of satellite and ground NO₂ data.

All months						
The morning analysis	Box1	Box2	Box3	Box4	Box5	Avg. Box1-5
r	0.79	0.69	0.43	0.64	0.61	0.73
N	155	156	88	76	75	172
p	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001
The afternoon analysis	Box1	Box2	Box3	Box4	Box5	Avg. Box1-5
r	0.80	0.67	0.64	0.60	0.65	0.74
N	146	147	91	77	79	147
p	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001
Dry months						
The morning analysis	Box1	Box2	Box3	Box4	Box5	Avg. Box1-5
r	0.79	0.56	0.54	0.73	0.72	0.74
N	96	96	51	45	45	98
p	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001
The afternoon analysis	Box1	Box2	Box3	Box4	Box5	Avg. Box1-5
r	0.81	0.58	0.72	0.71	0.70	0.75
N	87	80	51	43	45	87
p	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001

Ground NO₂ data in the morning (9:00–12:00 LT) and in the afternoon (12:00–15:00 LT) were compared together to investigate diurnal variation between these two periods. Figure 7 shows the monthly mean ground NO₂ concentrations averaged from the five boxes considered using the data for 2010–2016. For May–December, the levels of NO₂ concentrations in the morning and in the afternoon were not much different. However, for January–April, the levels of NO₂ concentrations in the afternoon were much higher than in the morning which matched with the results of satellite observations and fire hotspots in the previous section.

Fig. 7. Monthly mean ground NO₂ concentrations in the morning and in the afternoon.

3.3. Yearly Analysis of Satellite and Ground NO₂

Six-hourly (9:00–15:00) ground NO₂ concentrations covering morning and afternoon periods from all 10 stations were yearly averaged for 2010–2016 (ground monitoring NO₂ data of station ID 9 and 10 are available from 2010). The averaged ground NO₂ data were then compared with tropospheric NO₂ columns averaged from four satellites (SCIAMACHY, OMI, GOME-2A, and GOME-2B) from all five boxes. Figure 8 (a) presents the inter-annual variations (solid lines) and linear trends (dot lines) of the satellite and ground NO₂ datasets. During this 6-year period, both satellite and ground NO₂ had slightly increasing trends of 8.40% and 1.18% per 6 years, respectively. For 2010–2013, the inter-annual variation of tropospheric NO₂ columns and ground NO₂ concentrations correlated well. However, the annual level of tropospheric NO₂ columns slightly decreased for 2013–2015 and then increased after 2015, while the annual level of ground NO₂

concentrations showed the opposite direction after 2013. Figure 8 (b) presents the comparison considered only during dry months (October–April). The inter-annual variation of satellite and ground NO₂ were similar to the first case when the analysis was considered all months of the year. However, the results of the dry months presented higher levels of both satellite and ground NO₂ data than the results in the first case. The 6-year trends of satellite and ground NO₂ considered during dry months were 7.23% and 0.48% per 6 years, respectively.

Spatial distributions of annual mean tropospheric NO₂ columns from the morning satellites of SCIAMACHY in 2003, GOME-2A in 2008, GOME-2A in 2012, and GOME-2B in 2016 are presented in Fig. 9(a). The overall results clearly illustrated the increase in tropospheric NO₂ columns in the northern region. The afternoon results of OMI in 2008, 2012, and 2016 in Fig. 9(b) also revealed a similar increasing trend. The highest levels of annual mean tropospheric NO₂ columns in both the morning and the afternoon were notable in Lampang (box ID 2) and Phrae (box ID 4) compared to the other cities.

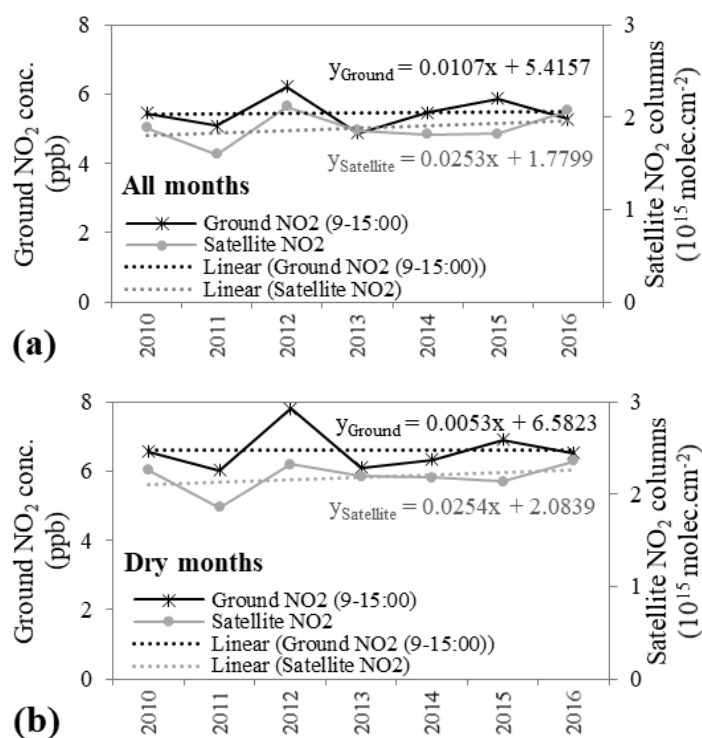


Fig. 8. Annual trends of satellite NO₂ columns and ground NO₂ concentrations; considered for all months (a) and for dry months (b).

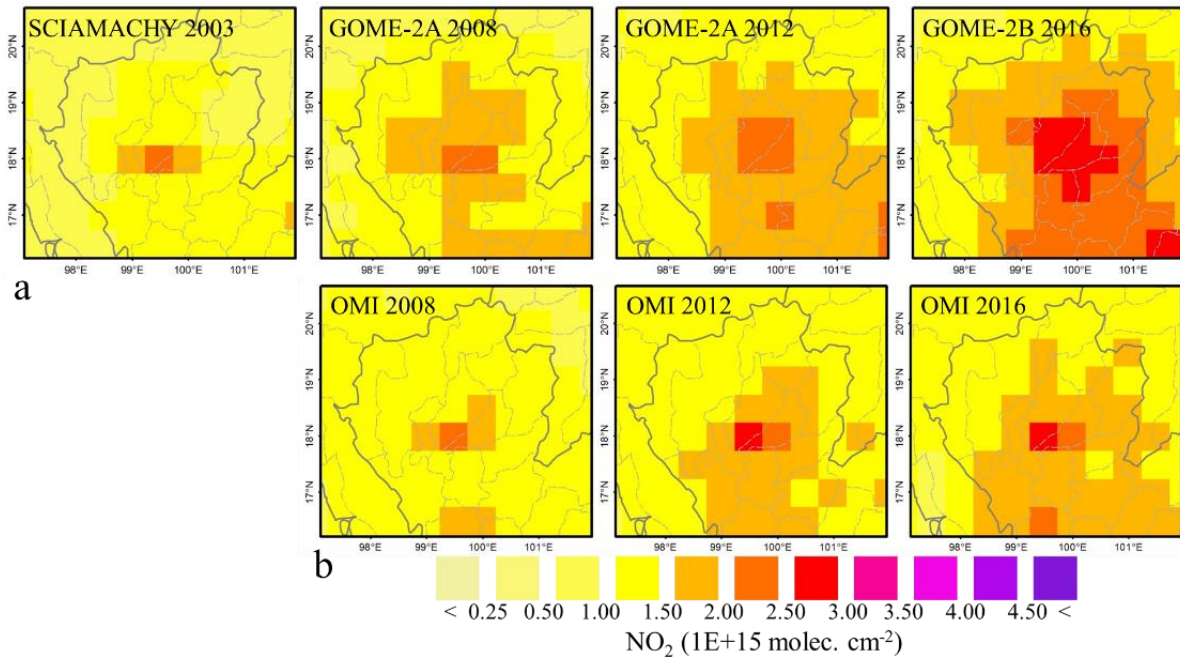


Fig. 9. Spatial distributions of annual mean tropospheric NO₂ columns from the morning satellites (a) and the afternoon satellite (b).

Figure 10 shows the comparison of 7-year (2010–2016) averaged data of satellite and ground NO₂ levels for each box. The analysis was separated into four cases. For the first and second cases, the NO₂ data were obtained from all months of the year (Fig. 10 (a)) and only on dry months (October–April) (Fig. 10 (b)), respectively. The results from both cases showed that the tropospheric NO₂ columns averaged from four satellites (SCIAMACHY, OMI, GOME-2A, and GOME-2B) had the highest levels in box ID 2 and the lowest levels in box ID 3. Ground NO₂ concentrations averaged for the time 9:00–15:00 LT had the highest levels in box ID 1 and the lowest levels in box ID 3. The NO₂ levels from both satellite and ground measurements were not much different between the other boxes. For the third case, the NO₂ data were collected for the analysis only on wet months (May–September) (Fig. 10 (c)). The result were similar to the first two cases with the highest levels of tropospheric NO₂ columns in box ID 2 and the highest levels of ground NO₂ concentrations in box ID 1. It is noteworthy that the satellite data represent the mean NO₂ levels covering all the areas of the 0.5°×0.5° boxes, whereas the ground data represent the NO₂ levels at the points where the ground monitoring stations are located. For box ID 1, the ground monitoring stations were in Chiang Mai and Lamphun cities. Chiang Mai is the most populated city in northern Thailand. Eighty percent of the area is mountains covered with forests. However, the ground monitoring stations are clustered in the urban area where high levels of air pollutants are generally emitted from road traffic, domestic, and industrial sources, which explains the highest levels of NO₂ collected from the ground monitoring stations being in box ID 1. On the contrary, the satellites recorded NO₂ data from both urban and rural areas and this was probably the cause of the discrepancy between the satellite and ground datasets, since NO₂ in rural areas is commonly emitted from biomass-burning activities. Further collection of ground NO₂ data in rural areas would clarify this hypothesis. Overall, NO₂ levels in the dry months were higher than in the wet months in all boxes. The analysis in the fourth case determined the ratios of NO₂ levels in the dry months to in the wet months (Fig. 10 (d)). The results showed that NO₂ levels in the dry months were approximately 1.30–2.00 times higher than NO₂ levels in the wet months for both satellite and ground measurements.

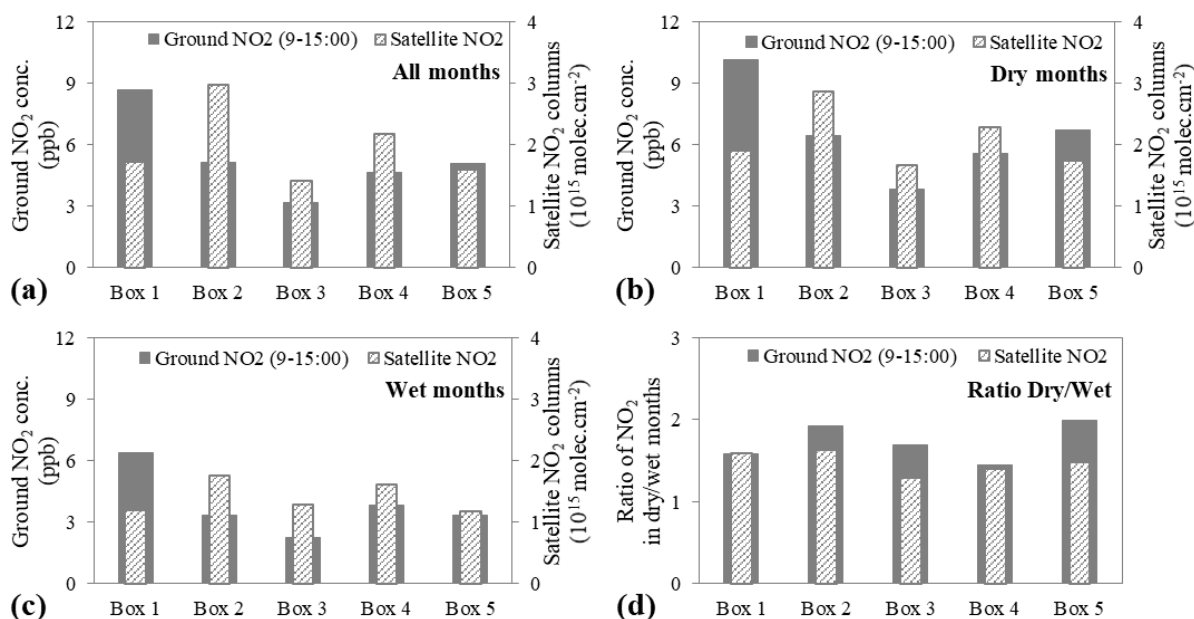


Fig. 10. Comparison of averaged data (2010–2016) of satellite and ground NO₂ for the five boxes; considered for all months (a), for dry months (b), for wet months (c), and ratios of NO₂ in dry months to in wet months (d).

4. Conclusions

Four satellite products of SCIMACHY, OMI, GOME-2A, and GOME-2B were used to analyze temporal and spatial variations of the tropospheric NO₂ columns over northern Thailand for 2003–2016. The comparisons among the satellite products showed better correlation during the dry period than the wet period. The afternoon satellite (OMI) generally recorded higher levels of NO₂ columns than morning satellites (SCIMACHY, GOME-2A, and GOME-2B), particularly during the biomass-burning period. The results correlated with the number of fire hotspots, highlighting intensive biomass-burning activities in the afternoon. Compared with ground monitoring data, all four satellites were able to detect the seasonal variations in the ground NO₂ concentrations. Both satellite and ground measurements recorded maximum levels of NO₂ during the peak biomass-burning period of January–April and minimum levels during the wet period of June–September. When the analysis was restricted to on dry months, the *r*-values between the satellite and ground datasets were found in the range 0.54–0.81. Moreover, there were slightly increasing trends in both the satellite and ground NO₂ datasets during dry months with values of 7.23% and 0.48% per 6 years, respectively, for 2010–2016. Satellite observations indicated that the maximum levels of NO₂ were in Lampang, while the ground measurements indicated this occurred in Chiang Mai and Lamphun. This discrepancy was perhaps due to the limited spatial coverage of ground monitoring data. Further collection data of ground NO₂ at rural or remote sites will provide more reliable results. However, both satellite and ground measurements showed approximately 1.30–2.00 times higher in NO₂ levels in the dry months than in the wet months. In conclusion, this study demonstrated that satellite observations were capable of providing valuable information to supplement existing ground-based data, especially in rural or remote areas where there is still a lack of a ground-based monitoring network. Satellite data can be used to assess spatial and temporal variations of NO₂. Integrating satellite measurements with ground-based data would provide a big picture view of NO₂ in the atmosphere which is an essential tool for air quality managers to understand the spatial distribution of air pollution for setting air quality policy and mitigation plan in Thailand. Further studies should extend the study areas to other regions of Thailand. Moreover, in order to determine ground-level of NO₂ concentrations from satellite columns, further analysis of daily data of satellite and ground NO₂ measurements with NO₂ vertical profiles will provide a better understanding of the relationship of satellite NO₂ columns and ground NO₂ concentrations.

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References

- [1] S. Solomon, R. W. Portmann, R. W. Sanders, J. S. Daniel, W. Madsen, B. Bartram, and E. G. Dutton, "On the role of nitrogen dioxide in the absorption of solar radiation," *J. Geophys. Res. Atmos.*, vol. 104, no. D10, pp. 12047–12058, 1999.
- [2] J. H. Seinfeld and S. N. Pandis, "Chemistry of the troposphere," in *Atmospheric Chemistry and Physics: From Air Pollution to Climate Change*, 2nd ed., New York, John Wiley & Sons, 2006, ch. 6, sec. 6.2, pp. 209–211.
- [3] S. Squizzato, M. Masiol, A. Brunelli, S. Pistollato, E. Tarabotti, G. Rampazzo, and B. Pavoni, "Factors determining the formation of secondary inorganic aerosol: A case study in the Po Valley (Italy)," *Atmos. Chem. Phys.*, vol. 13, pp. 1927–1939, 2013.
- [4] T. Stavrou, J.-F. Müller, K. F. Boersma, R. J. van der A, J. Kurokawa, T. Ohara, and Q. Zhang, "Key chemical NO_x sink uncertainties and how they influence top-down emissions of nitrogen oxides," *Atmos. Chem. Phys.*, vol. 13, pp. 9057–9082, 2013.
- [5] J. Ma, A. Richter, J. P. Burrows, H. Nüß, and J. A. van Aardenne, "Comparison of model-simulated tropospheric NO₂ over China with GOME-satellite data," *Atmos. Environ.*, vol. 40, pp. 593–604, 2006.
- [6] D. Chen, Y. Feng, and X. Zhang, "Comparison of variability and change rate in tropospheric NO₂ column obtained from satellite products across China during 1997–2015," *Int. J. Digit. Earth*, vol. 10, no. 8, pp. 814–828, 2017.
- [7] L. M. David and P. R. Nair, "Diurnal and seasonal variability of surface ozone and NO_x at a tropical coastal site: Association with mesoscale and synoptic meteorological conditions," *J. Geophys. Res.*, vol. 116, no. D10303, pp. 1–16, 2011.
- [8] H. Akiyama and H. Tsuruta, "Nitrous oxide, nitric oxide, and nitrogen dioxide fluxes from soils after manure and urea application," *J. Environ. Qual.*, vol. 32, no. 2, pp. 423–431, 2003.
- [9] L. Jaeglé, L. Steinberger, R. V. Martin, and K. Chance, "Global partitioning of NO_x sources using satellite observations: Relative roles of fossil fuel combustion, biomass burning and soil emissions," *Faraday Discuss.*, vol. 130, pp. 407–423, 2005.
- [10] S. Beirle, U. Platt, M. Wenig, T. Wagner, "Weekly cycle of NO₂ by GOME measurements: A signature of anthropogenic sources," *Atmos. Chem. Phys.*, vol. 3, no. 6, pp. 2225–2232, 2003.
- [11] A. Richter, J. P. Burrows, H. Nüß, C. Granier, and U. Niemeier, "Increase in tropospheric nitrogen dioxide over China observed from space," *Nature*, vol. 437, pp. 129–132, 2005.
- [12] Y. He, I. Uno, Z. Wang, T. Ohara, N. Sugimoto, A. Shimizu, A. Richter, and J. P. Burrows, "Variations of the increasing trend of tropospheric NO₂ over central east China during the past decade," *Atmos. Environ.*, vol. 41, pp. 4865–4876, 2007.
- [13] R. J. van der A, H. J. Eskes, K. F. Boersma, T. P. C. van Noije, M. Van Roozendaal, I. De Smedt, D. H. M. U. Peters, and E. W. Meijer, "Trends, seasonal variability and dominant NO_x source derived from a ten year record of NO₂ measured from space," *J. Geophys. Res.*, vol. 113, no. D04302, pp. 1–12, 2008.
- [14] L. N. Lamsal, R. V. Martin, A. van Donkelaar, E. A. Celarier, E. J. Bucsela, K. F. Boersma, R. Dirksen, C. Luo, and Y. Wang, "Indirect validation of tropospheric nitrogen dioxide retrieved from the OMI satellite instrument: Insight into the seasonal variation of nitrogen oxides at northern midlatitudes," *J. Geophys. Res.*, vol. 115, no. D05302, pp. 1–15, 2010.
- [15] H.-J. Lee, S.-W. Kim, J. Brioude, O. R. Cooper, G. J. Frost, C.-H. Kim, R. J. Park, M. Trainer, and J.-H. Woo, "Transport of NO_x in East Asia identified by satellite and in situ measurements and Lagrangian particle dispersion model simulations," *J. Geophys. Res. Atmos.*, vol. 119, pp. 2574–2596, 2014.
- [16] K. F. Boersma, D. J. Jacob, M. Trainic, Y. Rudich, I. DeSmedt, R. Dirksen, and H. J. Eskes, "Validation of urban NO₂ concentrations and their diurnal and seasonal variations observed from the SCIAMACHY and OMI sensors using in situ surface measurements in Israeli cities," *Atmos. Chem. Phys.*, vol. 9, pp. 3867–3879, 2009.

- [17] A. Hilboll, A. Richter, and J. P. Burrows, “Long-term changes of tropospheric NO₂ over megacities derived from multiple satellite instruments,” *Atmos. Chem. Phys.*, vol. 13, pp. 4145–4169, 2013.
- [18] P. Schneider, W. A. Lahoz, and R. van der A, “Recent satellite-based trends of tropospheric nitrogen dioxide over large urban agglomerations worldwide,” *Atmos. Chem. Phys.*, vol. 15, pp. 1205–1220, 2015.
- [19] K. F. Boersma, H. J. Eskes, and E. J. Brinksma, “Error analysis for tropospheric NO₂ retrieval from space,” *J. Geophys. Res. Atmos.*, vol. 109, pp. D04311, 2004.
- [20] H. Irie, K. F. Boersma, Y. Kanaya, H. Takashima, X. Pan, and Z. F. Wang, “Quantitative bias estimates for tropospheric NO₂ columns retrieved from SCIAMACHY, OMI, and GOME-2 using a common standard for East Asia,” *Atmos. Meas. Tech.*, vol. 5, pp. 2403–2411, 2012.
- [21] S. Itahashi, I. Uno, H. Irie, J.-I. Kurokawa, and T. Ohara, “Regional modeling of tropospheric NO₂ vertical column density over East Asia during the period 2000-2010: Comparison with multisatellite observations,” *Atmos. Chem. Phys.*, vol. 14, pp. 3623–3635, 2014.
- [22] K. M. Han, S. Lee, L. S. Chang, and C. H. Song, “A comparison study between CMAQ-simulated and OMI-retrieved NO₂ columns over East Asia for evaluation of NO_x emission fluxes of INTEX-B, CAPSS, and REAS inventories,” *Atmos. Chem. Phys.*, vol. 15, pp. 1913–1938, 2015.
- [23] I. Uno, Y. He, T. Ohara, K. Yamaji, J.-I. Kurokawa, M. Katayama, Z. Wang, K. Noguchi, S. Hayashida, A. Richter, and J. P. Burrows, “Systematic analysis of interannual and seasonal variations of model-simulated tropospheric NO₂ in Asia and comparison with GOME-satellite data,” *Atmos. Chem. Phys.*, vol. 7, pp. 1671–1681, 2007.
- [24] K. M. Hana, C. K. Lee, J. Lee, J. Kim, and C. H. Song, “A comparison study between model-predicted and OMI-retrieved tropospheric NO₂ columns over the Korean peninsula,” *Atmos. Environ.*, vol. 45, pp. 2962–2971, 2011.
- [25] H. Irie, K. Yamaji, K. Ikeda, I. Uno, S. Itahashi, T. Ohara, and J. Kurokawa, “An evaluation of the CMAQ reproducibility of satellite tropospheric NO₂ column observations at different local times over East Asia,” *Atmos. Chem. Phys. Discuss.*, vol. 13, pp. 14037–14067, 2013.
- [26] P. Lalitaporn, G. Kurata, Y. Matsuoka, N. Thongboonchoo, and V. Surapipith, “Long-term analysis of NO₂, CO, and AOD seasonal variability using satellite observations over Asia and intercomparison with emission inventories and model,” *Air Qual. Atmos. Health.*, vol. 6, pp. 655–672, 2013.
- [27] M. J. Bechle, D. B. Millet, and J. D. Marshall, “Remote sensing of exposure to NO₂: Satellite versus ground-based measurement in a large urban area,” *Atmos. Environ.*, vol. 69, pp. 345–353, 2013.
- [28] N. Arunrat, N. Pumijumnong, and S. Sereenonchai, “Air-pollutant emissions from agricultural burning in Mae Chaem basin, Chiang Mai province, Thailand,” *Atmos.*, vol. 9, no. 4, pp. 145, 2018.
- [29] B. Mitmark and W. Jinsart, “A GIS model for PM₁₀ exposure from biomass burning in the north of Thailand,” *App. Envi. Res.*, vol. 39, no. 2, pp. 77–87, 2017.
- [30] S. Sooktawee, U. Humphries, A. Patpai, R. Kongsong, S. Boonyapitak, and N. Piemyai, “Visualization and interpretation of PM₁₀ monitoring data related to causes of haze episodes in Northern Thailand,” *App. Envi. Res.*, vol. 37, no. 2, pp. 33–48, 2015.
- [31] M. Sukitpaneemit and N. T. Kim Oanh, “Satellite monitoring for carbon monoxide and particulate matter during forest fire episodes in northern Thailand,” *Environ. Monit. Assess.*, vol. 186, no. 4, pp. 2495–2504, 2014.
- [32] N. T. Kim Oanh and K. Leelasakultum, “Analysis of meteorology and emission in haze episode prevalence over mountain-bounded region for early warning,” *Sci. Total Environ.*, vol. 409, pp. 2261–71, 2011.
- [33] B. Pimpunchat, K. Sirimangkhal, and S. Junyapoon, “Modeling haze problems in the North of Thailand using logistic regression,” *J. Math. Fund. Sci.*, vol. 46, no. 2, pp. 183–193, 2014.
- [34] P. Wiwatanadate, “Acute air pollution-related symptoms among residents in Chiang Mai, Thailand,” *J. Environ. Health*, vol. 76, no. 6, pp. 76–85, 2014.
- [35] K. F. Boersma, H. J. Eskes, R. J. Dirksen, R. J. van der A, J. P. Veefkind, P. Stammes, V. Huijnen, Q. L. Kleipool, M. Sneep, J. Claas, J. Leitão, A. Richter, Y. Zhou, and D. Brunner, “An improved tropospheric NO₂ column retrieval algorithm for the Ozone Monitoring Instrument,” *Atmos. Meas. Tech.*, vol. 4, pp. 1905–1928, 2011.
- [36] P. F. Levelt, G. H. J. van den Oord, M. R. Dobber, A. Malkki, H. Visser, J. de Vries, P. Stammes, J. O. V. Lundell, and H. Saari, “The ozone monitoring instrument,” *IEEE Trans. Geosci. Remote Sens.*, vol. 44, no. 5, pp. 1093–1101, 2006.

- [37] P. Valks, G. Pinardi, A. Richter, J.-C. Lambert, N. Hao, D. Loyola, M. Van Roozendael, and S. Emmadi, "Operational total and tropospheric NO₂ column retrieval for GOME-2," *Atmos. Meas. Tech.*, vol. 4, pp. 1491–1514, 2011.
- [38] H. Bovensmann, J. P. Burrows, M. Buchwitz, J. Frerick, S. Noël, V. V. Rozanov, K. V. Chance, and A. P. H. Goede, "SCIAMACHY: Mission objectives and measurement modes," *J. Atmos. Sci.*, vol. 56, no. 2, pp. 127–150, 1999.
- [39] C. O. Justice, L. Giglio, S. Korontzi, J. Owens, J. T. Morissette, D. Roy, J. Descloitres, S. Alleaume, F. Petitcolin, and Y. Kaufman, "The MODIS fire products," *Remote Sens. Environ.*, vol. 83, pp. 244–262, 2002.
- [40] J. Engel-Cox, A. Huff, T. Kanabkaew, and N. T. Kim Oanh, "Satellite tools for air quality management with focus on particulate matter," in *Integrated Air Quality Management: Asian Case Studies*, N. T. Kim Oanh Ed. Boca Raton, Florida: CRC Press, 2012, ch. 4, pp. 125–148.
- [41] B.-N. Zhang and N. T. Kim Oanh, "Photochemical smog pollution in the Bangkok Metropolitan Region of Thailand in relation to O₃ precursor concentrations and meteorological conditions," *Atmos. Environ.*, vol. 36, pp. 4211–4222, 2002.
- [42] S. Itahashi, I. Uno, H. Irie, J. -I. Kurokawa, and T. Ohara, "Impacts of biomass burning emissions on tropospheric NO₂ vertical column density over continental Southeast Asia," in *Land-Atmospheric Research Applications in South and Southeast Asia*, K. Vadrevu, T. Ohara, and C. Justice, Ed. Springer Remote Sensing/Photogrammetry, 2018, ch. 4, pp. 67–81.
- [43] K. P. Vadrevu and C. O. Justice, "Vegetation fires in the Asian region: Satellite observational needs and priorities," *Glob. Environ. Res.*, vol. 15, pp. 65–76, 2011.
- [44] K. F. Boersma, D. J. Jacob, H. J. Eskes, R. W. Pinder, J. Wang, R. J. van der A, "Intercomparison of SCIAMACHY and OMI tropospheric NO₂ columns: Observing the diurnal evolution of chemistry and emissions from space," *J. Geophys. Res.*, vol. 113, no. D16S26, pp. 1–14, 2008.
- [45] Y. Wang, S. Beirle, J. Lampel, M. Koukouli, I. De Smedt, N. Theys, A. Li, D. Wu, P. Xie, C. Liu, M. Van Roozendael, T. Stavrou, J. F. Müller, and T. Wagner, "Validation of OMI, GOME-2A and GOME-2B tropospheric NO₂, SO₂ and HCHO products using MAX-DOAS observations from 2011 to 2014 in Wuxi, China: Investigation of the effects of priori profiles and aerosols on the satellite products," *Atmos. Chem. Phys.*, vol. 17, pp. 5007–5033, 2017.
- [46] P. Lalitaporn, "Temporal and spatial variability of tropospheric NO₂ columns retrieved from OMI satellite data and comparison with ground based information in Thailand," *Eng. Appl. Sci. Res.*, vol. 44, no. 4, pp. 227–234, 2017.