Comparison and Validation of TROPOMI and OMI NO$_2$ Observations over China

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Abstract: The new-generation sensor TROPOspheric Monitoring Instrument (TROPOMI) onboard the Sentinel 5 precursor (S5P) satellite is promising for monitoring air pollutants with greater spatial resolution, especially for China, which suffers from severe pollution. As tropospheric NO$_2$ vertical column densities (VCDs) from TROPOMI have become available since February 2018, this study presents the comparisons of NO$_2$ data measured by TROPOMI and its predecessor Ozone Monitoring Instrument (OMI) over China, together with validation against ground Multi-axis differential optical absorption spectroscopy (MAX-DOAS) measurements. At the nationwide scale, we used two different filters performed for the TROPOMI data (named TROPOMI50 and TROPOMI75), and the TROPOMI50 yielded larger values than TROPOMI75. The TROPOMI NO$_2$ datasets from different filters show consistent spatial patterns with OMI, and the correlation coefficient values were both above 0.93. However, linear regression indicates that NO$_2$ loadings in TROPOMI is about 2/3 to 4/5 of those in OMI, which is presumably due to a different cloud mask and uncertainties of air mass factors. The absolute difference is prominent over the high pollution areas such as Jing-Jin-Ji region and during winter and autumn, exceeding $0.6 \times 10^{16}$ molecules cm$^{-2}$ (molec cm$^{-2}$). However, the NO$_2$ concentrations retrieved from TROPOMI50 in the southern China may be somewhat higher than OMI. When it comes to the local-scale Jing-Jin-Ji hotspot, the analysis focuses on a comparison to TROPOMI75. TROPOMI manifests high quality and exhibits a significantly better performance of representing spatial variability. In contrast, OMI shows fewer effective pixels and does a poor job of capturing local details due to its row anomaly and low resolution. The absolute difference between two datasets shows the same seasonal behavior with NO$_2$ variation, which is most striking in the winter ($0.31 \times 10^{16}$ molec cm$^{-2}$) and is lowest in the summer ($0.05 \times 10^{16}$ molec cm$^{-2}$). Furthermore, the ground MAX-DOAS instrument in Xianghe station, the representative site in Jing-Jin-Ji, is used to assess the skill of satellite retrievals. It turns out that both OMI and TROPOMI underestimate the observations, ranging from 30% to 50%, with OMI being less biased. In spite of the negative drift, the temporal structures of changes derived from OMI and TROPOMI closely match the ground-based records, since the correlation coefficients are above 0.8 and 0.95 for daily and monthly scales, respectively. Overall, TROPOMI NO$_2$ retrievals are better suited for applications in China as well as the Jing-Jin-Ji hotspot due to its higher spatial resolution, although some improvements are also needed in the near future.

Keywords: tropospheric nitrogen oxides; comparison; validation; TROPOMI; OMI; MAX-DOAS
**1. Introduction**

Nitrogen dioxide (NO$_2$) is an important trace gas in the atmosphere. It is recognized as a pollutant that is toxic at high concentrations and can participate in various chemical reactions to adversely affect air quality. Nitrogen oxides (NO$_x$ = NO + NO$_2$) as a precursor could combine with hydrocarbons and lead to the regional-scale formation of ozone (O$_3$) under the sunlight. NOx could also be oxidized to nitric acid (HNO$_3$), and then in the presence of ammonia (NH$_3$), can lead to ammonium nitrate (NH$_4$NO$_3$). It is known that high concentrations of NO$_2$ cause respiratory problems [1]. The emission of tropospheric NO$_2$ can be divided into anthropogenic and natural sources. For example, fossil fuels are by far the dominant source of NOx, especially near major cities. In addition, biomass burning, lightning and soil emissions are also sources of NO$_2$ formation [2,3]. The distribution of tropospheric NO$_2$ is inhomogeneous due to the heterogeneous NO$_x$ sources and the relatively short lifetime of NOx. Therefore, better understanding of the physical and chemical characteristics of NO$_2$ as well as its variations at various time and spatial scales are indispensable for air pollution control and sustainable development.

In recent years, satellite remote sensing has become one of the important techniques of NO$_2$ observation, including the monitoring of NO$_2$ variations at global and regional scales [4–6], the NO$_2$ transport phenomena [7,8], estimation of NO$_x$ emissions [3,9–11] and inference of surface NO$_2$ concentration [12,13]. Compared with ground-based observations and situ monitors, satellite measurements could provide data with higher spatial-temporal resolution and global coverage. NO$_2$ products are now available from the satellite instruments such as Global Ozone Monitoring Experiment (GOME, 1995–2011), Scanning Imaging Absorption spectroMeter for Atmospheric ChartographY (SCIAMACHY, 2002–2012), Ozone Monitoring Instrument (OMI, since 2004) and TROPOspheric Monitoring Instrument (TROPOMI, since 2017).

However, atmospheric trace gas inversion based on satellite data, especially the tropospheric NO$_2$ column retrievals, are subject to several factors, such as the presence of clouds and aerosols, the uncertainty of the data quality, the accuracy of surface albedo and the influence of the priori assumed NO$_2$ profile [2]. Therefore, evaluating the accuracy of satellite data by validation and error analysis is of great importance, which is a significant part for the next-generation product improvement and application. A number of efforts about NO$_2$ validation have been conducted and the approaches to verify the quality of satellite products have become increasingly diverse and mature, e.g., airborne in situ profile measurements [14], ground-based and airborne observations [15–17] and model validation [18]. For example, Konovalov et al. [19] presented the results of a first comparison of the tropospheric NO$_2$ column amounts derived from GOME with the simulated data from a European-scale chemistry transport model (CTM). The analysis was performed separately for Western and Eastern Europe using the data for summer months of 1997 and 2001. It was found that the total random errors of NO$_2$ columns over Eastern Europe were larger than Western. Heue et al. [20] validated the SCIAMACHY tropospheric NO$_2$ vertical and slant columns data with the Airborne Multi AXis DOAS (AMAXDOAS) instrument in February 2003 over the Alps, the Po-Valley and the Mediterranean. The result suggested that the tropospheric NO$_2$ between the two datasets showed a good agreement and linear correlation was in a slope of 0.93 over less polluted areas. Irie et al. [21] compared tropospheric NO$_2$ vertical column densities (VCDs) from three satellite datasets (SCIAMACHY, OMI and GOME-2) with Multi-axis differential optical absorption spectroscopy (MAX-DOAS) measurements, respectively. The biases estimated from three datasets against the MAX-DOAS observations were $-5 \pm 14\%$, $-10 \pm 14\%$ and $1 \pm 14\%$ respectively, over China, which were all small and insignificant. Moreover, many studies have validated the quality of different versions of OMI NO$_2$ products. Celarier et al. [22] comprehensively validated the OMI standard NO$_2$ product (Level 2) derived from the National Aeronautics and Space Administration (NASA), including the stratospheric, tropospheric and total NO$_2$ VCD. Several different instruments (ground- and aircraft-based measurements) have been used in this experiment from November 2004 through December 2006. Correlations between OMI NO$_2$ and nearby ground-based measurements are about 0.8–0.9 on a global scale, with OMI stratospheric NO$_2$ underestimated by
about 14% and total and tropospheric columns underestimated by 15–30%. Zheng et al. [23] presented a comparison between the DP (DOMINO) and SP (Standard Product) NO\textsubscript{2} products from different OMI algorithms. The result showed that DP was higher than SP by 13% in winter and lower by 9% in summer in east China. Krotkov et al. [24] introduced the new version 3.0 NASA OMI standard NO\textsubscript{2} product (SPv3) and its major improvements, including a new spectral fitting algorithm for NO\textsubscript{2} slant column density (SCD) retrieval and an updated air mass factors (AMFs). The research also demonstrated that SPv3 products agreed better with independent satellite- and ground-based Fourier transform infrared (FTIR) measurements, while further evaluation was needed over polluted areas. The assessment performed by Choi et al. [25] also found that OMI NO\textsubscript{2} values were lower compared with Pandora measurement over the highly polluted environments in four US states and South Korea, due in part to inaccurate retrieval assumptions (e.g., priori profiles), but mostly to OMI’s areal (>312 km\textsuperscript{2}) averaging.

More recently, NO\textsubscript{2} data derived from the new satellite instrument TROPOMI could be acquired. In addition to the high spatial resolution, the quality of the data is also unprecedentedly improved compared to previous instruments [26]. With regards to the TROPOMI-derived tropospheric NO\textsubscript{2} columns, only a few studies dealing with validation against in situ aircraft and ground-based measurements have been reported. For example, Griffin et al. [26] compared TROPOMI tropospheric NO\textsubscript{2} data with aircraft, surface in situ NO\textsubscript{2} and ground-based direct sun Pandora measurements respectively, over the Canadian oil sands. The results suggested that good agreement was generally seen between the TROPOMI NO\textsubscript{2} VCD and in situ measurements. As China is one of the regions suffering severe NO\textsubscript{2} pollution in the world, especially the densely populated areas over the Eastern China [27–29], fully validating the quality of TROPOMI NO\textsubscript{2} data in these areas are necessary, the results of which benefit the relevant application and algorithm development. Therefore, the main goal of this study is to evaluate the new TROPOMI tropospheric NO\textsubscript{2} product (reprocessed and offline, v1.2.0) over China, involving its performance validation using the ground MAX-DOAS technique and comparison with its predecessor, the OMI dataset.

In Section 2, we present a brief description of the TROPOMI and OMI NO\textsubscript{2} products as well as the MAX-DOAS measurement at Xianghe site. Section 3 discusses the comparisons between TROPOMI and OMI NO\textsubscript{2} observation, over the entire China (Section 3.1.1) and Jing-Jin-Ji region (Section 3.1.2). Moreover, evaluation of TROPOMI and OMI products against ground MAX-DOAS measurements are also described (Section 3.2). Finally, the conclusions are presented in Section 4.

2. Data

2.1. TROPOMI NO\textsubscript{2} Data

On 13 October 2017, the Sentinel 5 precursor (S5P) satellite, a sun-synchronous orbit satellite, was launched. The S5P is dedicated to monitoring air quality and acquiring atmospheric composition, including O\textsubscript{3}, NO\textsubscript{2}, SO\textsubscript{2}, carbon monoxide (CO), CH\textsubscript{4}, formaldehyde (HCHO) and aerosol properties, with daily global coverage. The single payload of the S5P is TROPOMI, which is a push-broom imaging spectrometer with eight spectral bands covering ultraviolet (UV) to shortwave infrared (SWIR) wavelengths. Specifically, the spectral bands contain the UV-visible band at 270–500 nm, the near-infrared (NIR) band at 710–770 nm and the SWIR band at 2314–2382 nm. The instrument images a stretch of land on a two-dimensional detector in 1 s, during which the satellite moves about 7 km. After the 1 s, the satellite moves again and continues to scan the earth. The two-dimensional detector is used to scan different ground pixels with different wavelengths and across track directions. Moreover, the several spectral bands are detected by the four different detectors. TROPOMI has a heritage to both the OMI as well as the SCIAMACHY and aims to continually provide the time series of tropospheric data products [30]. In the UV wavebands, its radiometric performance and spectral resolution are the same as its predecessor OMI, but the horizontal resolution at true nadir is almost raised one order of magnitude [31]. The TROPOMI NO\textsubscript{2} data product was developed by the Royal
Netherlands Meteorological Institute (KNMI) and is based on the Differential Optical Absorption Spectroscopy (DOAS) retrieval method in the 405–465 nm spectral range, which was similar to the QA4ECV OMI NO$_2$ retrieval algorithm (a new DOAS retrieval performed for NO$_2$ within the EU FP7-project Quality Assurance for Essential Climate Variables). The product provides the tropospheric VCD, which describes the vertically integrated number of NO$_2$ molecules per unit area from the surface to the tropopause. The TROPOMI NO$_2$ retrieval algorithm is introduced in detail by van Geffen et al. [32].

In this study, we use the reprocessed (RPRO, from February to April 2018) and offline (OFFL, from May 2018 to January 2019) TROPOMI Level 2 NO$_2$ products compiled by the KNMI (http://www.temis.nl). Spatial resolution varies with the across track position, and in this study, the average pixel size of NO$_2$ data is $3.5 \times 7$ km$^2$ (its spatial resolution has been changed to $3.5 \times 5.5$ km$^2$ since 6 August 2019 [33]). For TROPOMI, quality-control has been performed before the analysis. A flag, namely quality assurance value (qa_value), for each ground pixel indicates the status and quality of the retrieval result, ranging from 0 (no output) to 1 (all is well) [32]. We selected the valid pixel, for which the qa_value was above 0.75, to exclude part of the scenes covered by snow/ice, errors and problematic retrievals. Cloud-covered records (cloud radiance fraction >0.5) were also excluded during the data processing (across each pixel). We used this highest quality data filtered from TROPOMI and named it “TROPOMI75” in the analysis. In addition, to make a further comparison with OMI, a “moderate” filter (qa_value > 0.5) was also performed for TROPOMI. This adds the good-quality retrievals over clouds and over scenes covered by snow/ice and filters out the pixels with errors and problematic retrievals. TROPOMI data with a “moderate” filter (named TROPOMI50) is also used and compared with OMI, and the results are presented in Section 3.1.1.

2.2. OMI NO$_2$ Data

We used tropospheric NO$_2$ columns retrieved from OMI onboard NASA’s EOS-Aura satellite (launched in July 2004). This satellite orbits the Earth in a near-polar, sun-synchronous way, whose orbital cycle is 99 min, and crosses the equator at 13:40 local time. OMI has three spectral channels from 264 to 504 nm at an average spectral resolution of 0.5 nm [16]. The spatial resolution is about $13 \times 24$ km$^2$ at nadir. Under normal situations, OMI could scan the entire earth’s surface once per 24 h. Before the launch of TROPOMI, OMI had been used extensively to study the distribution and characteristics of NO$_2$ at global and regional scales and accumulated more than 15 years of data. In this study, the QA4ECV OMI NO$_2$ dataset (Level 2 orbit data) retrieved by KNMI (http://www.temis.nl) was taken into account to compare against TROPOMI, for the entire research period of February 2018 to January 2019. The improved QA4ECV NO$_2$ data record provides valuable information to speculate emissions and explore trends in NO$_2$ on a global scale. At the same time, QA4ECV NO$_2$ data has been proven to reduce and better quantify the uncertainty of the NO$_2$ retrieval. A detailed description of the OMI NO$_2$ vertical columns retrieval algorithm is available from Boersma et al. [34]. Starting in 2007, some track positions were affected by row anomaly. Therefore, affected pixels have been removed before statistical analysis. At the same time, only clear sky data, defined as having a cloud radiance fraction less than 50% for each pixel, and satellite zenith angle less than 70° were collected. In addition, quality-control (processing_error_flag = 0) has also been performed for OMI to ensure that the pixels are valid in analysis.

2.3. MAX-DOAS Measurements

The MAX-DOAS instrument based on the zenith-sky DOAS technique exhibits high sensitivity in the lower troposphere. The instrument used in this study was set up at the Xianghe station (39.75° N, 116.96° E), a representative observatory in the Jing-Jin-Ji region, about 50 km southeast of Beijing (marked in red in Figure 1). It consists of three components: a thermo-regulated box equipped with two spectrometers, an optical head fixed on a sun tracker and two computers for instrument control and data storage [35]. The instrument can measure both scattered and direct sunlight. Sunlight collected
by telescopes is transmitted and separated by two-way splitter optical fibers to the spectrometers for spectral analysis. One spectrometer operates in the UV region (300–390 nm) and the other works in the visible band (400–720 nm). The full width at half maximum (FWHM) spectral resolutions of two spectrometers are 0.4 and 0.9 nm, respectively [36]. In the current observation, the telescope points towards a fixed azimuth direction to the north. A full MAX-DOAS scan comprises 9 elevation viewing angles (2, 4, 6, 8, 10, 12, 15, 30 and 90°) and requires about 15 min [37]. The observations investigated in this study cover five consecutive months, from February to June 2018.

![Figure 1](image)

**Figure 1.** Terrain elevation and location of the Xianghe Station (red triangle) and other major cities.

### 3. Results and Discussion

3.1. Comparison of TROPOMI and OMI Tropospheric NO₂

3.1.1. Spatial Analysis at the National Scale

To compare the spatial and temporal characteristics of NO₂ products from TROPOMI and OMI at the nationwide scale, the NO₂ concentrations from two datasets between February 2018 and January 2019 have been used in this section. The TROPOMI data is divided into TROPOMI75 and TROPOMI50 due to different filtering conditions. To facilitate the comparison, the TROPOMI L2 daily data is resampled to a resolution of 0.25° × 0.25° grid based on inverse distance weighted interpolation (IDW), identical to that of OMI. For the spatial colocation, a geographical grid is established around the study region of 0.25° × 0.25°. Assuming that the points within every grid have impacts on the grid value, then the impact of these points is inversely proportional to the distance between the center pixel value. The distance \( D_i \) from the discrete point \((x_i, y_i)\) to the established grid center point \((x_0, y_0)\) can be defined as:

\[
D_i = \sqrt{(x_0 - x_i)^2 + (y_0 - y_i)^2}
\]

and the estimated value on grid point \(Z_{(x_0,y_0)}\) can be defined by:

\[
Z_{(x_0,y_0)} = \sum_{i=1}^{n} \frac{1}{(D_i)^p} Z_i / \sum_{i=1}^{n} \frac{1}{(D_i)^p}
\]

where \(Z_i\) is the observation on discrete point \(i\), \(N\) is the number of pixels involved in the calculation and \(P\) is the weight of distance (set 2 in this calculation). Note that invalid records, cloud- and snow-covered
pixels have been eliminated. The further calculation of monthly averages only includes days when both instruments observed the valid NO\textsubscript{2} information.

Figure 2 shows the monthly mean NO\textsubscript{2} fields of TROPOMI and OMI retrievals and their absolute difference over China (4° N–60° N, 70° E–140° E), which were exemplary for November 2018. The absolute difference (AD) is defined as in Equation (3):

\[
AD = VCD_{OMI} - VCD_{TROPOMI}
\]

Evidently, both retrievals from TROPOMI and OMI exhibit a distinct spatial pattern, with high NO\textsubscript{2} loadings in Eastern China in November and especially in the North China Plain (NCP). The TROPOMI produces smoother fields than OMI because it has more pixels. In contrast, the OMI product shows more missing values over Western China. Furthermore, the NO\textsubscript{2} is generally higher in OMI than in TROPOMI over the NCP, both in TROPOMI75 and TROPOMI50, as reflected in Figure 2d,e, with the absolute differences being greater than 0.6 \times 10^{16} \text{molec cm}^{-2}. Notably, obvious differences occurred in Southern China between TROPOMI75 and TROPOMI50 (Figure 2f), where higher values were observed in TROPOMI50 than TROPOMI75, indicating that strict filtering conditions may eliminate some low NO\textsubscript{2} values. When considering TROPOMI50 and OMI, there were significant negative differences in Southern China, corresponding to high NO\textsubscript{2} values measured by TROPOMI. In general, the TROPOMI shows lower tropospheric NO\textsubscript{2} values than OMI at the national scale in November, especially in the NCP, meaning that TROPOMI may underestimate the NO\textsubscript{2} VCDs when NO\textsubscript{2} enhancements occur. However, the concentrations retrieved from TROPOMI away from highly polluted areas may be somewhat higher than OMI.

![Figure 2](image_url)

**Figure 2.** Exemplary monthly mean maps (0.25° × 0.25° grid) of (a) TROPOMI75, (b) TROPOMI50, (c) OMI tropospheric NO\textsubscript{2} VCDs as well as (d–f) their absolute differences (AD) (units: 10^{16} \text{molec cm}^{-2}) for November 2018.

In the same way, to better explore NO\textsubscript{2} seasonal variation and product discrepancy, the months have been grouped into four seasons, including spring (March, April and May), summer (June, July and August), autumn (September, October and November) as well as winter (December, January and February). Figure 3 illustrates the seasonal cycle of NO\textsubscript{2} based on TROPOMI75 (a-1 to a-4) and OMI (b-1 to b-4). The NO\textsubscript{2} concentration has a pronounced seasonal cycle over China, which is highest in winter and lowest in summer. Previous studies concluded that the seasonal variation in the lifetime of NO\textsubscript{x} depends strongly on the photolysis rates, which is a function of
sun angle [38,39]. In reality, the seasonal cycle in NO\textsubscript{2} is also affected by seasonal variations in anthropogenic emissions and meteorological conditions, but these are assumed to be of secondary importance [4,40]. At the same time, the two products show very similar NO\textsubscript{2} spatial structure and seasonal evolution and both can highlight highly contaminated regions. Furthermore, a substantial difference is found in winter and autumn compared to the other two seasons. The significant discrepancies exceeding $0.6 \times 10^{16}$ molec cm\textsuperscript{-2} are found over Jing-Jin-Ji region centered at Beijing, Tianjin and Tangshan. Besides, it is also the case in the Yangtze River Delta and Chengdu–Chongqing district during wintertime.

In what follows, the annual means of the NO\textsubscript{2} VCDs over China from February 2018 to January 2019, as observed by TROPOMI and OMI respectively, are shown in Figure 4a–c. In terms of annual average, the spatial pattern from TROPOMI bears a great resemblance to that from OMI, with NO\textsubscript{2} VCDs varying from 0.02 to 2.20 ($\times 10^{16}$ molec cm\textsuperscript{-2}) for the whole of China. High NO\textsubscript{2} loadings are mainly located over the North China and Yangtze River Delta as a consequence of the combined effects of both human activities and natural causes [28,41]. However, the OMI measures more
NO₂ than TROPOMI over the high pollution areas such as the NCP, where the difference is about 0.40 (×10^{16} \text{molec cm}^{-2}) with TROPOMI75 and about 0.20 (×10^{16} \text{molec cm}^{-2}) with TROPOMI50, as shown in Figure 4d–e. This result based on an annual basis is similar to that on a seasonal basis mentioned above. Moreover, the NO₂ VCD retrieved from TROPOMI50 in southern China is higher than OMI, with the absolute difference of −0.4 (×10^{16} \text{molec cm}^{-2}). Explanations for this difference may result from the restrictions in the different satellite sensors and algorithms. For instance, the instrument issues and the quality of the level 1 data may affect the retrieval of NO₂ SCDs and their uncertainties [34], such as available spectral coverage and wavelength calibration. For another example, satellite remote sensing of NO₂ is frequently hindered by the cloud contamination, and thereby, cloud-screening is one of crucial steps in NO₂ retrieval. TROPOMI NO₂ retrieval is prone to be influenced by strict cloud mask (called FRESCO-S), which presumably exclude some heavy pollution events in the retrievals and leads to the underestimation of inversion results [32]. As shown by Boersma et al. [42], exclusion of cloudy scenes in the validation dataset between OMI and SCIAMACHY decreases NO₂ concentrations by 15% on average, but does not affect the seasonal variation. In the TROPOMI processor, clouds are characterized by using cloud fraction and cloud top height parameters, which are both derived from radiance observations in the O₂ A-band and used as an input in a cloud-correction scheme in NO₂ retrieval [43]. Therefore, the presence of clouds can strongly affect the retrieval results. Like clouds, aerosol is also a potential influence which can affect the accuracy of tropospheric NO₂ retrieval. Aerosol-related errors are intimately coupled to cloud parameter errors. The O₂ A-band cloud algorithm currently does not correct for the presence of aerosols, which means aerosols are not explicitly treated in the TROPOMI NO₂ algorithm (all AMF calculations being performed for a Rayleigh atmosphere) [43]. Furthermore, Figure 4f shows the scatter diagram of NO₂ annual means between the two products over the study region. The linear regression yields a slope of 0.8 and spatial correlation coefficient (R) of about 0.97 between TROPOMI50 and OMI. For the comparison between TROPOMI50 and OMI, it is also well-correlated with the slope of 0.8 and R of 0.93. Note that the dots denote the pixels excluded invalid values over the study area in Figure 4a–c. Therefore, TROPOMI and OMI show good agreement in terms of temporal pattern, but the magnitudes in the TROPOMI are generally smaller than in the OMI data.

![Figure 4](image_url)

**Figure 4.** Tropospheric NO₂ distributions averaged over a period from February 2018 to January 2019 (units: 10^{16} \text{molec cm}^{-2}) (a) from TROPOMI75, (b) from TROPOMI50 and (c) from OMI, along with (d–e) absolute difference maps (OMI minus TROPOMI). (f) Scatterplot of TROPOMI versus OMI annual mean tropospheric NO₂ columns with regression line superimposed. The slope and correlation coefficient (R) are also shown in the plot.
3.1.2. Analysis over the Jing-Jin-Ji Region

As mentioned before, Jing-Jin-Ji region is an economic mega-urban zone in East China, but at the same time, it suffers from high environmental pollution as well. Thus, the hotspot has been highlighted and will be specifically examined in this section. Specifying that the rest of the analysis focuses on a comparison to TROPOMI75, Figure 5 depicts the daily and monthly NO2 measured by TROPOMI and OMI over the Jing-Jin-Ji region. Observations for a single day, 22 January 2019, are shown in Figure 5a,b. TROPOMI offers higher quality and finer details of NO2. OMI, by contrast, shows fewer effective pixels and does a poor job of capturing local details due to its row anomaly and low resolution. As regards OMI, therefore, multiple days’ or months’ average is essential to obtain reliable estimates. To this end, we average TROPOMI and OMI tropospheric NO2 fields over a 0.25° × 0.25° grid cell and depict the monthly mean NO2 VCD for January 2019 (Figure 5c,d). It is noteworthy that TROPOMI exhibits clear and structural features of NO2 distribution, with elevated NO2 column densities detected in the southwest part and some urban centers. However, OMI provides more uniform spatial distribution and contains lots of noise grids, which to some extent is unable to unravel spatial diversity. Moreover, the absolute difference between the two datasets is apparent in Figure 5e. The difference between mean NO2 value of OMI (1.38) and TROPOMI (1.00) reaches 0.38 (×1016 molec cm−2). NO2 VCDs derived from TROPOMI are obviously lower than OMI over this region, especially in the southern area with high NO2 loadings (blue box highlighted in Figure 5e). If the domain turns from Jing-Jin-Ji to the area marked in the blue box, the correlation coefficient deteriorates substantially from 0.87 to 0.58. Therefore, the smaller the spatial scale, the more significant the difference between TROPOMI and OMI.

Figure 5. (a,b) Tropospheric NO2 measured by TROPOMI75 and OMI on 22 January 2019 over the Jing-Jin-Ji region. (c,d) The same as (a,b) but for the average of January 2019. (e) The absolute difference of OMI minus TROPOMI tropospheric NO2 columns averaged in January 2019. Blue box denotes the area with high NO2 loadings. (units: 1016 molec cm−2).
Furthermore, we present a quantitative comparison between time series of averaged NO$_2$ retrievals from two datasets and the statistics are listed in Table 1. Overall, over the chosen area, tropospheric NO$_2$ retrievals illustrated the high degree of temporal consistency between the two datasets in four seasons and individual months, as suggested by R. Meanwhile, the NO$_2$ VCDs from the two datasets both indicated a pronounced seasonal fluctuation. The highest NO$_2$ concentration appears in winter, followed by autumn. The difference of the two datasets implies the same seasonal behavior, with the maximum value occurring in winter ($0.31 \times 10^{16}$ molec cm$^{-2}$) and the minimum in summer ($0.05 \times 10^{16}$ molec cm$^{-2}$). These results indicate that the discrepancy between TROPOMI and OMI measurements over the Jing-Jin-Ji region is significant during all seasons, and particularly marked during winter and autumn. Lorente et al. [44] performed an experiment and concluded that the AMF structural uncertainty is to be 42% over polluted regions and 31% over unpolluted regions, mostly driven by substantial differences in the a priori trace gas profiles, surface albedo and cloud parameters. Therefore, the discrepancy may be more evident in the season with NO$_2$ enhancement.

For the comparisons of monthly averages, differences range from 0.03 to 0.56 ($\times 10^{16}$ molec cm$^{-2}$) and correlations are above 0.85, again confirming their spatial similarity, but with deviations in magnitude. In addition, on the annual basis, the mean value of TROPOMI was about $0.57 (\times 10^{16}$ molec cm$^{-2}$), which was smaller than that of the corresponding OMI data by $0.74 (\times 10^{16}$ molec cm$^{-2}$). Meanwhile, the correlation coefficient ($R = 0.98$) was extremely high throughout the whole year from February 2018 to January 2019.

Table 1. Statistics describing the comparison of TROPOMI and OMI NO$_2$ retrievals over the Jing-Jin-Ji region.

<table>
<thead>
<tr>
<th>Period</th>
<th>OMI</th>
<th>TROPOMI</th>
<th>Difference</th>
<th>R</th>
<th>Slope</th>
<th>Intercept</th>
<th>N</th>
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<td>0.57</td>
<td>0.17</td>
<td>0.98</td>
<td>0.74</td>
<td>0.02</td>
<td>841</td>
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<tr>
<td>Spring</td>
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<td>0.52</td>
<td>0.11</td>
<td>0.96</td>
<td>0.77</td>
<td>0.03</td>
<td>841</td>
</tr>
<tr>
<td>Summer</td>
<td>0.34</td>
<td>0.29</td>
<td>0.05</td>
<td>0.95</td>
<td>0.78</td>
<td>0.02</td>
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</tr>
<tr>
<td>Autumn</td>
<td>0.86</td>
<td>0.63</td>
<td>0.23</td>
<td>0.96</td>
<td>0.67</td>
<td>0.06</td>
<td>841</td>
</tr>
<tr>
<td>Winter</td>
<td>1.15</td>
<td>0.84</td>
<td>0.31</td>
<td>0.94</td>
<td>0.65</td>
<td>0.09</td>
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</tr>
<tr>
<td>February</td>
<td>0.67</td>
<td>0.57</td>
<td>0.10</td>
<td>0.90</td>
<td>0.75</td>
<td>0.08</td>
<td>816</td>
</tr>
<tr>
<td>March</td>
<td>0.83</td>
<td>0.62</td>
<td>0.21</td>
<td>0.91</td>
<td>0.61</td>
<td>0.11</td>
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<tr>
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<td>0.11</td>
<td>0.90</td>
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<td>0.40</td>
<td>0.03</td>
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<td>0.03</td>
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<tr>
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<td>0.92</td>
<td>0.82</td>
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<tr>
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<td>0.85</td>
<td>0.59</td>
<td>0.07</td>
<td>816</td>
</tr>
<tr>
<td>August</td>
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<td>0.25</td>
<td>0.06</td>
<td>0.86</td>
<td>0.68</td>
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<td>0.87</td>
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</table>

Note: The OMI and TROPOMI represent the NO$_2$ mean value from OMI and TROPOMI75. Difference is OMI minus TROPOMI. R is the correlation coefficient between two datasets. Columns are in the unit of $10^{16}$ molec cm$^{-2}$. Slope and intercept of the regression of TROPOMI on OMI, along with the sample size, are shown in the last three columns.

Figure 6 shows histograms derived from the daily comparison of tropospheric NO$_2$ VCDs observed by TROPOMI and OMI over the Jing-Jin-Ji region during the measurement period. The absolute difference (OMI–TROPOMI) ranges from $-1$ to 2 ($\times 10^{16}$ molec cm$^{-2}$) with the mean value of 0.13 ($\times 10^{16}$ molec cm$^{-2}$), and nearly 2/3 of the values (65.6%) are between $-0.2$ and 0.2 ($\times 10^{16}$ molec cm$^{-2}$). Meanwhile, approximately 76.9% of OMI values are greater than TROPOMI. The positive difference mainly appears in winter and autumn, while the opposite often occurs in summer and spring. In addition, the mean R value is 0.80 and about 80.3% of days show R above 0.70, and only 6% are below 0.50. Lowest correlations are usually due to fewer pixels or the influence of
clouds. For instance, the pixels covered by cloud contamination or OMI records influenced by row anomaly could lead to low correlations. These results confirm that most of the time, the TROPOMI agrees very well with OMI on patterns.

![Image of histograms](image)

**Figure 6.** Histograms of the (a) absolute difference and (b) correlation coefficient between daily OMI and TROPOMI data over the Jing-Jin-Ji region.

Briefly, at the national and regional scale, the TROPOMI and OMI NO2 VCDs agree well in terms of spatiotemporal structure over China. However, the magnitudes in the OMI are generally larger than in the TROPOMI data by 20% to 30%, especially over highly polluted regions and during winter and autumn. Further, TROPOMI offers higher quality and finer details of NO2, but OMI does a poor job of capturing spatial diversity at the local scale along with substantial missing value problems.

3.2. Validation against Ground-Based MAX-DOAS Measurements

3.2.1. Diurnal Variations

In this section, the MAX-DOAS instrument is used to assess the skill of the TROPOMI and OMI NO2 datasets in matching the ground-based observations. We selected TROPOMI and OMI columns that have a pixel center within ±0.5° latitude/longitude of the Xianghe station and are measured under mostly clear situations (cloud radiance fractions less than 50%). Figure 7 provides the day-to-day variation of tropospheric NO2 levels inferred from satellite and ground-based measurements. From 15 to 26 June 2018, a complete picture during 08:00 to 16:00 local time was created by MAX-DOAS, as shown in Figure 7. Meanwhile, the open triangles and circles denote the collocated TROPOMI and OMI measurements, respectively. TROPOMI measurements are often in good agreement with OMI. The satellite values, for the most part, are fairly close to the MAX-DOAS observations. As for the characteristics of NO2 variations illustrated in Figure 7, the daily cycle is highly variable from day to day, depending on several factors, including the diurnal cycle of NOx emissions, the change of meteorological conditions as well as the photochemical reactions of NOx [17]. Among them, sun angle, boundary layer height and wind speeds are the main meteorological drivers. Meanwhile, the elevated NO2 column densities are particularly pronounced during the morning rush hour, which is due to the heavy transportation emissions during the peak hours to some extent, beyond that, low-boundary layer heights in the morning are also a reason for the increased NO2 during the morning hours [45]. It should be mentioned that the satellite overpass times generally occur just after the midday maximum in NO2 concentration, which may influence the range of NO2 values explored in the follow-up comparison to some extent.
The daily comparisons between the satellite- and ground-based measurements are quantitatively illuminated in the corresponding scatterplots (Figure 8). The MAX-DOAS data are averaged within 12:30 to 14:30 local time, which covered the satellite overpass time to ensure meaningful comparisons with the satellite measured values. According to the R values, on the one hand, quite good agreements were found among the three datasets. OMI data has a close relationship with the MAX-DOAS (R = 0.86) that is slightly higher than TROPOMI versus MAX-DOAS (R = 0.81). In addition, the R between TROPOMI and OMI daily observations is as high as 0.91 (samples = 73) during the 5-month period. On the other hand, the linear regression lines are made to compare the amplitude of the different datasets and gave the following results: regression slope = 0.57 for TROPOMI and regression slope = 0.71 for OMI. Therefore, it can be found that the satellite retrievals from TROPOMI and OMI tend to underestimate tropospheric NO$_2$ values over the Jing-Jin-Ji region by 30% to 50%, with OMI being less biased. There are several factors that may account for these findings. First, the MAX-DOAS concentrations are a focused path through a polluted area, whereas the satellite pixel is a spatially broad area that combines highly polluted and less polluted paths. Additionally, as we know, to convert a slant column to the corresponding vertical column, the influence of the light path and the corresponding parameters are calculated and expressed as AMF. The uncertainty in the estimate of the tropospheric AMF is the largest source of uncertainty in NO$_2$ satellite retrievals in situations with enhanced trace gas concentrations. Cloud fraction, aerosol characterization, surface albedo and profile shape are all important influential factors associated with the AMF computation [18]. Therefore, when the AMF is computed using a high-resolution chemical transport model, the tropospheric vertical column amount can be increased by large amounts.

**Figure 7.** Average diurnal variation of tropospheric NO$_2$ VCDs measured by MAX-DOAS (plus symbols) during 08:00 to 16:00 local time at Xianghe station. The open triangles and circles denote the TROPOMI75 and OMI measurements for the site, respectively (units: 10$^{16}$ molec cm$^{-2}$).
Figure 8. (a) Correlation between daily TROPOMI tropospheric NO$_2$ columns and MAX-DOAS NO$_2$ observations made from 1 February through 30 June 2018. (b) Same as (a), but for correlation between OMI and MAX-DOAS. (c) Same as (a), but for correlation between TROPOMI75 and OMI (units: $10^{16}$ molec cm$^{-2}$).

The time series of NO$_2$ columns derived from the three datasets and their direct differences are displayed in Figure 9. The direct bias between the MAX-DOAS NO$_2$ concentrations and the satellite NO$_2$ columns at Xianghe station is noticeable. The ground MAX-DOAS NO$_2$ columns are larger than the corresponding TROPOMI and OMI values covering the research period, since most of the difference values (ground-based minus satellite-based) are positive. This confirms that both daily mean TROPOMI and OMI NO$_2$ are negatively biased relative to MAX-DOAS over this region. Moreover, the difference values are closer to 0 from May to July than other months. As mentioned before, the NO$_2$ product of TROPOMI and OMI both show some agreement at low values, while it has obvious discrepancies for episodes of NO$_2$ enhancement. This is due to the AMF computed with a low-resolution priori NO$_2$ profiles, among other potential factors. Goldberg et al. [46] also shows that high-resolution models in NO$_2$ retrieval could simulate larger concentrations near the surface in urban areas.
The AMF could improve the agreement between TROPOMI and ground-based Pandora total columns over regions with NO2 enhancements (from $-28.5 \pm 3.3\%$ for TMS-MP to $-23.7 \pm 3.5\%$ for CAMS). A similar investigation has also been reported by Dimitropoulou et al. [43]. They generated a modified version of the TROPOMI tropospheric NO2 VCDs using the daily-averaged MAX-DOAS concentration profiles. The change in NO2 profile shape has a strong impact on validation results, leading to a better agreement between satellite and ground-based MAX-DOAS data. On average, the TROPOMI results have been systematically lower than the MAX-DOAS results, and the biases seem to be proportional to the NO2 VCD magnitude [21,47]. Specifically, OMI data are 32.0% lower than the MAX-DOAS data on average, and TROPOMI data are generally 45.5% smaller than MAX-DOAS. Such negative bias is possibly induced by the unrealistic a priori assumptions used in the calculation of AMF, as already noted, a key parameter in the retrieval algorithm [16,34]. For the TROPOMI NO2 retrieval algorithm, the NO2 vertical profiles are specified by the TM5-MP model, for 34 vertical layers at the horizontal resolution of $1^\circ \times 1^\circ$ in latitude–longitude, which are very coarse and cannot express spatial gradients at the scale of a city. As shown by Ialongo et al. [33], replacing the coarse a priori NO2 profiles with high-resolution profiles from the Copernicus Atmosphere Monitoring Service (CAMS) chemical transport model of the AMF could improve the agreement between TROPOMI and ground-based Pandora total columns over regions with NO2 enhancement (from $-28.5 \pm 3.3\%$ for TMS-MP to $-23.7 \pm 3.5\%$ for CAMS). A similar investigation has also been reported by Dimitropoulou et al. [43]. They generated a modified version of the TROPOMI tropospheric NO2 VCDs using the daily-averaged MAX-DOAS concentration profiles. The change in NO2 profile shape has a strong impact on validation results, leading to a better agreement between satellite and ground-based MAX-DOAS data. On average, the TROPOMI results have been

Figure 9. (a) Time series of tropospheric NO2 VCDs from February to June in 2018. Solid lines represent the fit curves of the three datasets. (b) Daily mean absolute differences between MAX-DOAS and satellite tropospheric NO2 VCDs. Blue triangles indicate MAX-DOAS minus OMI data, and red dots represent MAX-DOAS minus TROPOMI75 (unit: $10^{16}$ molec cm$^{-2}$).

3.2.2. Monthly Variations

To minimize the uncertainties associated with natural and instrument noise, monthly averages are further employed in these comparisons. Figure 10(a–c) displays the relation plots between the monthly mean satellite-measured and ground-based-measured NO2 VCDs. Excellent linear agreement was seen between the TROPOMI and MAX-DOAS, with the R reaching about 0.99. It is also shown that OMI and MAX-DOAS NO2 were well-correlated ($R = 0.96$). However, the slopes between satellite- and ground-based data deviate from the unity line, again confirming that the satellite underestimates the signal in reference to MAX-DOAS.

In Figure 10(d), the monthly mean values are plotted. Both OMI and TROPOMI tropospheric NO2 columns are systematically lower than the MAX-DOAS results, and the biases seem to be proportional to the NO2 VCD magnitude [21,47]. Specifically, OMI data are 32.0% lower than the MAX-DOAS data on average, and TROPOMI data are generally 45.5% smaller than MAX-DOAS. Such negative bias is possibly induced by the unrealistic a priori assumptions used in the calculation of AMF, as already noted, a key parameter in the retrieval algorithm [16,34]. For the TROPOMI NO2 retrieval algorithm, the NO2 vertical profiles are specified by the TM5-MP model, for 34 vertical layers at the horizontal resolution of $1^\circ \times 1^\circ$ in latitude–longitude, which are very coarse and cannot express spatial gradients at the scale of a city. As shown by Ialongo et al. [33], replacing the coarse a priori NO2 profiles with high-resolution profiles from the Copernicus Atmosphere Monitoring Service (CAMS) chemical transport model of the AMF could improve the agreement between TROPOMI and ground-based Pandora total columns over regions with NO2 enhancement (from $-28.5 \pm 3.3\%$ for TMS-MP to $-23.7 \pm 3.5\%$ for CAMS). A similar investigation has also been reported by Dimitropoulou et al. [43]. They generated a modified version of the TROPOMI tropospheric NO2 VCDs using the daily-averaged MAX-DOAS concentration profiles. The change in NO2 profile shape has a strong impact on validation results, leading to a better agreement between satellite and ground-based MAX-DOAS data. On average, the TROPOMI results have been
increased by about 50% compared to previous validation after their recalculation. This suggests that using high-resolution input will improve the tropospheric AMF, and as such, the tropospheric NO$_2$ VCDs for satellite retrieval. Nevertheless, it is also worth noting that the R between satellite- and ground-based datasets are quite good in all cases. Finally, it should be noted that successive efforts are required due to a limited number of samples on monthly scales.

![Figure 10](image-url) (a) Correlation plot showing the collocated monthly mean TROPOMI75 tropospheric NO$_2$ columns and MAX-DOAS NO$_2$ measurements. (b) Same as (a), but for correlation between OMI and MAX-DOAS. (c) Same as (a), but for correlation between TROPOMI75 and OMI. (d) Monthly mean NO$_2$ tropospheric columns based on TROPOMI75, OMI and MAX-DOAS, from February to June 2018. The error bars correspond to the standard deviation of the monthly mean tropospheric NO$_2$ VCDs (units: 10$^{16}$ molec cm$^{-2}$).

4. Conclusions

In this work, the first year of the tropospheric NO$_2$ product from TROPOMI was compared with OMI and validated against the ground MAX-DOAS instrument, in order to evaluate their applicability for air quality monitoring, especially in the highly polluted Jing-Jin-Ji region. The major conclusions are summarized below.

(1) At the nationwide scale, the tropospheric NO$_2$ column derived from TROPOMI produces consistent spatial NO$_2$ patterns to OMI, with correlation coefficient above 0.93. Moreover, both datasets could capture monthly and seasonal cycles of NO$_2$ equally well. However, the magnitude of NO$_2$ is generally lower in TROPOMI than in OMI by 20% to 30%, and the absolute difference is prominent over the high-pollution areas, such as the Jing-Jin-Ji region and the Yangtze River Delta and during winter and autumn, exceeding 0.6 \times 10^{16} \text{molec cm}^{-2}. However, the NO$_2$ concentrations retrieved from TROPOMI in Southern China may be somewhat higher than OMI.

(2) When it comes to the local-scale Jing-Jin-Ji hotspot, TROPOMI manifests high quality and exhibits clear and structural features of NO$_2$ distribution, indicating a significantly better performance
of representing spatial variability. That is, the TROPOMI observations are more likely to reveal NO\textsubscript{2} information at a local and regional scale. In contrast, OMI provides more uniform spatial distribution and contains lots of noise grids, which to some extent is unable to unravel spatial diversity. If the regional mean is considered, the two datasets are highly consistent for the annual and monthly average of NO\textsubscript{2} columns (\(R > 0.85\)), while TROPOMI produces lower NO\textsubscript{2} content and this discrepancy is more outstanding for high NO\textsubscript{2} loadings. Meanwhile, both the datasets indicate a similar seasonal fluctuation with the highest value during winter and the lowest during summer. At the same time, the difference implies the same seasonal behavior, which is most striking in the winter (\(0.31 \times 10^{16}\) molec cm\(^{-2}\)) and is close to each other in the summer (\(0.05 \times 10^{16}\) molec cm\(^{-2}\)). In over-polluted areas, the differences are possibly due to the uncertainties in the cloud and aerosol characterization and surface albedo used for the retrievals. They are all important parameters which have significant influences on satellite tropospheric NO\textsubscript{2} AMFs. For NO\textsubscript{2} retrieval, aerosols and clouds can both mask some of the tropospheric NO\textsubscript{2}, hence introducing inaccuracies in the retrieved columns.

(3) Finally, we have conducted the comparison between the satellite data and ground-based measurements at Xianghe site. It turns out that both OMI and TROPOMI underestimate the observations by ranging from 30\% to 50\%, with OMI being less biased. In spite of the negative drift, the temporal structures of changes derived from OMI and TROPOMI closely match the ground-based records, since the R values are above 0.8 and 0.95 for daily and monthly scales, respectively.

In summary, TROPOMI has a much finer footprint than any previous satellite instruments, which could provide NO\textsubscript{2} observations with improved spatial resolution and low instrumental noise. Most of the time, the TROPOMI agrees very well with OMI on patterns; however, the NO\textsubscript{2} values derived from TROPOMI are generally underestimated compared with OMI and MAX-DOAS measurements when NO\textsubscript{2} loadings are high. Thus, further investigations are needed to examine the causes of these differences and propose improvement of the NO\textsubscript{2} inversion over China.

**Author Contributions:** Conceptualization, C.W. and T.W.; Formal analysis, C.W.; Supervision, T.W., P.W. and V.R.; Validation, C.W.; Writing—original draft, C.W.; Writing—review & editing, T.W., P.W. and V.R. All authors have read and agreed to the published version of the manuscript.

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