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Comparison of TROPOMI/Sentinel 5 Precursor NO_2 observations with ground-based measurements in Helsinki

Iolanda Ialongo[1], Henrik Virta[1], Henk Eskes[2], Jari Hovila[1], and John Douros[2]

[1]Space and Earth Observation Centre, Finnish Meteorological Institute, Helsinki, Finland.
[2]Royal Netherlands Meteorological Institute, De Bilt, Netherlands.

Correspondence: Iolanda Ialongo (iolanda.ialongo@fmi.fi)

Abstract. We present a comparison between satellite-based TROPOMI (TROPOspheric Monitoring Instrument) NO_2 products and ground-based observations in Helsinki (Finland). TROPOMI NO_2 total (summed) columns are compared with the measurements performed by the Pandora spectrometer during April–September 2018. We find a high correlation (r = 0.68) between satellite- and ground-based data, but also that TROPOMI total columns underestimate ground-based observations for relatively large Pandora NO_2 total columns, corresponding to episodes of relatively elevated pollution. This is expected because of the relatively large size of the TROPOMI ground pixel (3.5 × 7 km) and the a-priori used in the retrieval compared to the relatively small field-of-view of the Pandora instrument. Replacing the coarse a-priori NO_2 profiles with high-resolution profiles from the CAMS chemical transport model improves the agreement between TROPOMI and Pandora total columns for episodes of NO_2 enhancement.

We also analyse the consistency between satellite-based data and in situ NO_2 surface concentrations measured at the Helsinki-Kumpula air quality station (located a few metres from the Pandora spectrometer). We find similar day-to-day variability between TROPOMI, Pandora and in situ measurements, with NO_2 enhancements observed during the same days. Both satellite- and ground-based data show a similar weekly cycle, with lower NO_2 levels during the weekend compared to the weekdays as a result of reduced emissions from traffic and industrial activities (as expected in urban sites). The TROPOMI NO_2 maps reveal also spatial features, such as the main traffic ways and the airport area, as well as the effect of the prevailing south-west wind patterns.

This is one of the first works in which TROPOMI NO_2 retrievals are validated against ground-based observations and the results provide an early evaluation of their applicability for monitoring pollution levels in urban sites. Overall, TROPOMI retrievals are valuable to complement the ground-based air quality data (available with high temporal resolution) for describing the spatio-temporal variability of NO_2, even in a relatively small city like Helsinki.

1 Introduction

Nitrogen oxides (NO_x = NO + NO_2) play an important role in tropospheric chemistry, participating in ozone and aerosol production. NO_x are mainly generated by combustion processes from anthropogenic pollution sources (including transportation, energy production and other industrial activities), and they are toxic in high concentrations at the surface (US-EPA, 2019).

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The NO_2 amount in the atmosphere can be measured using satellite-based instruments. Launched in October 2017, TROPOMI (TROPOspheric Monitoring Instrument), the only payload on-board the European Space Agency's (ESA) Sentinel-5 Precursor (S5P) satellite, is expected to revolutionise the way we monitor air pollution from space because of its unprecedented spatial resolution (3.5×7 km at the beginning of the mission and 3.5×5.5 km since 6 August 2019) and high signal-to-noise ratio. TROPOMI (jointly developed by the Netherlands and ESA) is designed to retrieve the concentrations of several atmospheric constituents including ozone, NO_2, SO_2, CO, CH_4, CH_2O, aerosol properties as well as surface UV radiation. TROPOMI derives information on atmospheric NO_2 concentrations by measuring the solar light back-scattered by the atmosphere and the Earth's surface. Due to its high spatial resolution, TROPOMI observations are particularly suitable to monitoring polluting emission sources at city level. The S5P mission is part of the Space Component of the European Copernicus Earth Observation Programme.

TROPOMI builds on the experience from previous polar orbiting instruments such as the Dutch-Finnish Ozone Monitoring Instrument (OMI), which has been operating on-board NASA's EOS (Earth Observing System) Aura satellite (Levelt et al., 2006) since late 2004. OMI NO_2 observations have been used in several air quality applications and the main achievements have been recently summarised by Levelt et al. (2018). The results achieved using OMI NO_2 retrievals include estimating top-down polluting emissions, analysing changes in the pollution levels over the period of 13 years, and verifying the success of environmental policy measures (e.g., Beirle et al., 2011; Castellanos and Boersma, 2012; Streets et al., 2013; Lu et al., 2015; Lamsal et al., 2015; Duncan et al., 2016; Krotkov et al., 2016; Liu et al., 2017). Also, OMI observations have been used for monitoring the NO_2 weekly cycle over urban sites (Beirle et al., 2003; Boersma et al., 2009; de Foy et al., 2016). Recently, a reprocessing of the OMI NO_2 dataset has become available (Boersma et al., 2018) as deliverable of the European QA4ECV project. Many of the QA4ECV OMI retrieval developments have been incorporated in the TROPOMI NO_2 retrieval processor.

Since TROPOMI/S5P is a very recent mission, accurate validation against independent ground-based measurements is needed in order to evaluate the quality of the retrieval. Recently, the Pandonia Global Network (PGN), including a network of ground-based Pandora spectrometers, has been established to provide reference measurements of NO_2 total columns for validating satellite-based retrievals. Pandora measures direct sunlight in the UV-VIS (ultraviolet-visible) spectral range (280–525 nm) and provides NO_2 total columns using the direct-sun DOAS (Differential Optical Absorption Spectroscopy) technique (Herman et al., 2009). Recently, Zhao et al. (2019) presented a method to derive NO_2 total columns from Pandora zenith-sky measurements as well.

Very recently, Griffin et al. (2019) presented first results of the validation of TROPOMI NO_2 retrievals over the Canadian oil sands using air-mass factors calculated with the high-resolution GEM-MACH model. They show how the TROPOMI NO_2 vertical column densities are highly correlated with ground-based observations and agree within 15–30 %. In this work, we evaluate the quality of TROPOMI NO_2 vertical columns against ground-based observations in the urban site of Helsinki (60.2°N; 24.95°E). Helsinki is a city with about half a million inhabitants, surrounded by a larger urban area (including the city of Espoo in the west and Vantaa in the north-east). Satellite-based NO_2 observations from OMI instrument in Helsinki were previously validated by Ialongo et al. (2016), finding that OMI generally underestimates Pandora total columns by 5–30 %, depending on the retrieval algorithm and parameters. The improved resolution of TROPOMI retrievals is expected to reduce the effect of dilution, due to the relatively coarse pixel size as compared to the field-of-view of the ground-based observations.

The satellite- and ground-based data used in the analysis are described in Sect. 2. The results of the comparison between TROPOMI NO_2 retrievals and ground-based Pandora total columns are shown in Sect. 3. The temporal correlation with in situ NO_2 surface concentration measurements and the NO_2 weekly cycle are also analysed. Finally, the conclusions are presented in Sect. 4.

2 Data and methodology

2.1 TROPOMI NO_2 observations

TROPOMI is a passive sensing hyperspectral nadir-viewing imager aboard the Sentinel-5 Precursor (S5P) satellite, launched on the 13th October 2017. S5P is a near-polar sun-synchronous orbit satellite flying at an altitude of 817 km, with an overpass time at ascending node (LTAN) of 13:30 (local time) and a repeat cycle of 17 days (KNMI, 2017). TROPOMI is operated in a non-scanning push broom configuration, with an instantaneous field-of-view of 108° and a measurement period of about 1 second. This results in a swath width of approx. 2600 km, an along-track resolution of 7 km and daily global coverage (KNMI, 2017). TROPOMI's four separate spectrometers measure the ultraviolet (UV), UV-visible (UVIS), near-infrared (NIR) and short-wavelength infrared (SWIR) spectral bands, of which the NIR and SWIR bands are new as compared to its predecessor OMI (Veefkind et al., 2012).

The NO_2 columns are derived using TROPOMI's UVIS spectrometer backscattered solar radiation measurements in the 405–465 nm wavelength range (van Geffen et al., 2015, 2019). The swath is divided into 450 individual measurement pixels, which results in a near-nadir resolution of 7×3.5 km. The total NO_2 slant column density is retrieved from the Level 1b UVIS radiance and solar irradiance spectra using Differential Optical Absorption Spectroscopy (DOAS) (Platt and Stutz, 2008). The species fitted by TROPOMI and their corresponding literature cross sections can be found in van Geffen et al. (2019). Tropospheric and stratospheric slant column densities are separated from the total slant column using a data assimilation system based on the TM5-MP chemical transport model, after which they are converted into vertical column densities using a look-up table of altitude dependent air-mass factors (AMF) and information on the vertical distribution of NO_2 from TM5-MP available with a horizontal resolution of $1° \times 1°$ and a time step of 30 minutes (van Geffen et al., 2019; Boersma et al., 2018; Williams et al., 2017).

The instrument, the NO_2 retrieval and assimilation scheme, and the data product have been described in detail by Veefkind et al. (2012), KNMI (2017), KNMI (2018), Eskes et al. (2019) and van Geffen et al. (2019).

We used reprocessed TROPOMI NO_2 data files, processor version 1.2.2, for the entire study period of 15.4.–30.9.2018. Reprocessed data files are occasionally generated using older sensing data as new processor algorithm versions become available. Version 1.2.x includes retrieval enhancements for high solar zenith angle and snow covered scenes (Eskes et al., 2019), both of which are important for high latitude locations such as Helsinki. Additionally, offline and near-real time (NRT) NO_2 products are also available. Offline data files are the main TROPOMI data product and are made available within about two

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weeks from sensing time, whereas NRT files are available within 3 hours of measurement time. NRT files are generated using forecast TM5-MP data rather than analysis data as with offline and reprocessed files (van Geffen et al., 2019) but the differences between the offline/reprocessed and near-real time products are generally small.

The TROPOMI NO_2 product used in the comparison was the summed total column, which is the sum of the tropospheric and stratospheric vertical column densities. It was chosen over the total column product, since the latter's sensitivity to the ratio between the stratospheric and tropospheric a-priori columns may lead to substantial systematic retrieval errors. The intermediate step of using data assimilation to first estimate the stratospheric column does remove part of this error. The precision values of the summed total columns used in the analysis stay within the range (0.5–4.5) $\times 10^{15}$ molec. cm^{-2} (or about 10–50 %). The data before 30 April 2018 were downloaded from the Sentinel-5P Expert Users Data Hub (https://s5pexp.copernicus.eu/dhus) as part of the S5P validation team activities, and starting from this date from the S5P Pre-Operations Data Hub (https://s5phub.copernicus.eu/dhus).

Figure 1 shows the TROPOMI NO_2 tropospheric columns over Helsinki averaged over the period 15.4.–30.9.2018. The largest enhancements are visible over the main traffic lanes as well as the Helsinki-Vantaa airport and surrounding area. Overall, the NO_2 levels during weekends (right panel) are smaller than those observed during weekdays (left panel) by about 30 %. This is typical for urban sites due to the weekly variability of traffic-related emissions, which are relatively higher during working days (from Monday to Friday). We also note that the NO_2 spatial distribution shown in Fig. 1 is partially affected by systematic wind patterns, which causes the NO_2 levels in the eastern part of the area to become relatively higher than the western part. Fig. S1 in the supplementary material shows the difference between the NO_2 tropospheric columns (normalised to the maximum value in the area) for all wind and low wind speed (less than 3 m s^{-1}) conditions. The pixels in red and blue in Fig. S1 indicate the area where the NO_2 levels are relatively higher or lower, respectively, due to the wind patterns. This is related to the prevailing wind directions from south-west over the Helsinki capital region.

Since the retrieval of TROPOMI vertical column densities (VCDs) is sensitive to the a-priori estimate of the NO_2 profile shape, the accuracy of the VCDs may be improved by using a-priori profiles from a chemical transport model (CTM) with a higher resolution than the $1° \times 1°$ of TM5-MP (Williams et al., 2017), as described by Eskes et al. (2019). In order to analyse their impact on the comparison, we used NO_2 profiles from the CAMS regional ENSEMBLE model (Météo-France, 2016; Marécal et al., 2015) as an alternative to TM5-MP profiles. It is a median of seven European CTMs, and the data are provided on a regular $0.1° \times 0.1°$ grid over Europe on 8 vertical levels up to 5 km altitude. In addition, the CAMS global model was used to generate the profiles above 3 km altitude with the assumption that this model gives a more reliable description of NO_x in the free troposphere. In particular, we used the ratios between TROPOMI tropospheric air-mass factors derived using the CAMS and the TM5-MP a-priori profiles. These ratios were available on the regular CAMS $0.1° \times 0.1°$ grid, between 30.4.–30.9.2018.

2.2 Ground-based NO_2 observations

The NO_2 total columns measured by the ground-based Pandora instrument #105 located in the district of Kumpula, Helsinki, Finland (60.20°N, 24.96°E), are compared to the TROPOMI NO_2 retrievals. The Pandora system is composed of a spectrometer connected by a fibre optic cable to a sensor head with 1.6° FOV (field-of-view). A sun-tracking device allows the optical head

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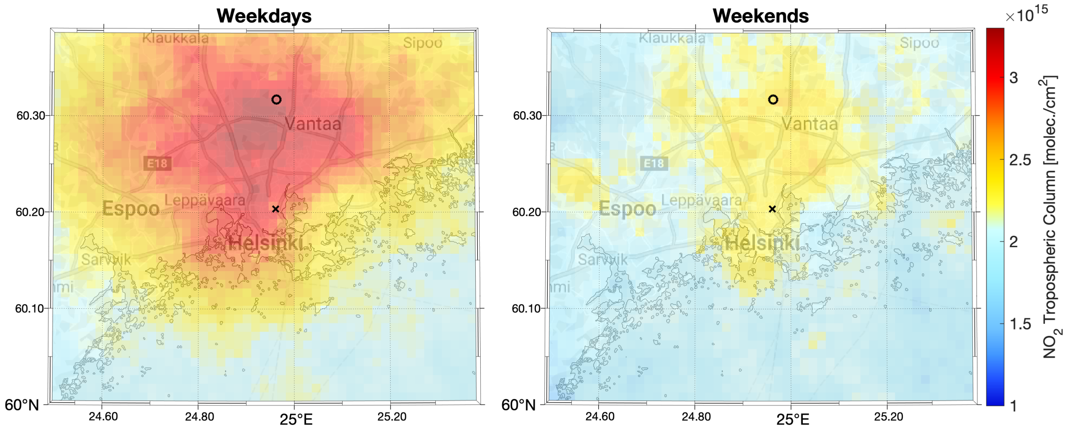

Figure 1. Average TROPOMI NO_2 tropospheric columns over Helsinki during the period 15.4.–30.9.2018. The left and right panels correspond to weekdays and weekends, respectively. The data have been binned and averaged to a 1 km resolution grid. The locations of the Kumpula ground-based station and the Helsinki-Vantaa airport are shown with a black cross and circle, respectively.

to point at the centre of the Sun with a precision of 0.013° (Herman et al., 2009). Pandora performs direct-sun measurements in the UV-VIS spectral range (280–525 nm) and provides NO_2 total vertical column densities, among other products.

The NO_2 total column retrieval is based on the DOAS spectral fitting technique (e.g., Cede et al., 2006), with NO_2 and O_3 being the trace gases fitted. The algorithm derives the relative NO_2 slant column densities (SCDs) from the 400–440 nm spectral band and converts them to absolute SCDs using a statistically estimated reference spectrum obtained using the Minimum-Amount Langley-Extrapolation method (MLE) (Herman et al., 2009).

The Pandora SCD retrieval employs a temperature correction to the cross-sections used in the spectral fitting procedure based on modelled monthly average NO_2 and temperature profiles and high-resolution temperature-dependent cross sections by Vandaele et al. (1998) for NO_2 and Serdyuchenko et al. (2014) for O_3 (as in the TROPOMI retrieval). We note that, while TROPOMI uses the ECMWF operational model as its source for atmospheric temperature profiles (van Geffen et al., 2019), Pandora uses a precalculated atmospheric temperature for a typical NO_2 profile (Cede, 2019). Due to the nature of direct-sun measurements no Ring effect correction is needed for Pandora (Herman et al., 2009).

The NO_2 columns are available about every 1.5 minutes. The full description of the Pandora instrument and the algorithm for the inversion methodology has been presented by Herman et al. (2009). The nominal clear-sky precision of the Pandora NO_2 total column retrievals is in the order of 3×10^{14} molec. cm^{-2} with an accuracy of about $\pm 1.3 \times 10^{15}$ molec. cm^{-2}. The accuracy depends on the uncertainties in the MLE-calculated reference spectrum, difference between the actual and assumed atmospheric temperature profiles, and uncertainties in the laboratory-determined absorption cross sections (Herman et al., 2009). At typical Helsinki concentrations (6×10^{15} molec. cm^{-2}) and AMF values (2.0) most of the systematic errors are due to uncertainties in the reference spectrum (Sect. 3.3 in Herman et al. (2009)). Pandora #105 is part of the Pandonia global network and the observations used in this paper were processed following the Pandonia procedure and distributed at http://pandonia.net/data/.

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The NO_2 surface concentrations available from the Kumpula, Helsinki, air quality (AQ) station were also used in order to analyse the temporal correspondence between surface NO_2 concentrations and TROPOMI vertical columns. The surface concentration data were obtained from FMI's databases as hourly averaged measurements. Kumpula station is classified as a semi-urban site, and its surface NO_2 concentrations are measured using an online trace level gas analyser based on the ultraviolet fluorescence method.

2.3 Methodology

We evaluate the agreement between TROPOMI and Pandora NO_2 vertical column densities by calculating the mean absolute difference (MD), the mean relative difference (MRD), the correlation coefficient (r), and the slope of an orthogonal linear fit for the measurements. The MD is defined as the average difference between the TROPOMI and Pandora VCDs in equation (1), whereas the MRD is the average of these differences when normalised with Pandora's VCD (equation (2)).

$$\mathrm{MD} = \frac{1}{n}\sum_{i=1}^{n}(\mathrm{VCD}_{\mathrm{TROPOMI},i} - \mathrm{VCD}_{\mathrm{Pandora},i}) \quad (1)$$

$$\mathrm{MRD} = 100\,\% \times \frac{1}{n}\sum_{i=1}^{n}\frac{\mathrm{VCD}_{\mathrm{TROPOMI},i} - \mathrm{VCD}_{\mathrm{Pandora},i}}{\mathrm{VCD}_{\mathrm{Pandora},i}} \quad (2)$$

A positive MD or MRD is thus an indication of TROPOMI overestimation, and negative an indication of TROPOMI underestimation. We also analyse weekdays and weekends separately, and the results are presented in Sect. 3.

Both TROPOMI and Pandora data were separately filtered according to a set of quality assurance criteria, after which the remaining temporally co-located measurements were compared with each other. For TROPOMI, only measurements with a data quality value (QA) >0.75 are used, which disqualifies scenes with a cloud radiance fraction >0.5, some scenes covered by snow or ice, and scenes that have been determined to include errors or problematic retrievals. Further details on the QA value are provided in the appendices of van Geffen et al. (2019). Only TROPOMI pixels including the Helsinki Pandora station were considered for the comparison. Also, only Pandora retrievals with data quality flag value of 0, 1, 10 or 11, corresponding to so-called assured and not-assured high or medium quality data (Cede, 2019), were taken into account. Pandora measurements within 10 minutes of TROPOMI overpass were averaged to get the Pandora-component of the validation data pairs. Wind speed data (average from the four lowest pressure levels: 925, 950, 975 and 1000 hPa) available from the European Centre for Medium-Range Weather Forecasts (ECMWF) as part of the ERA5 reanalysis product (https://cds.climate.copernicus.eu) were associated with each data pair in order to quantify the effect of advection on the NO_2 concentrations. The wind data were linearly interpolated to the Helsinki Pandora station's coordinates and the overpass time of each TROPOMI pixel used in the comparison.

Furthermore, we analyse the effect of the co-location choices on the MD and MRD value by varying both the maximum distance from the ground-based station and the averaging time interval for Pandora measurements around the S5P overpass time. The results are presented in Sect. 3. When calculating these values for increasing maximum distances, we also required that in all cases the TROPOMI pixel above the station had to have a valid measurement fulfilling our quality criteria.

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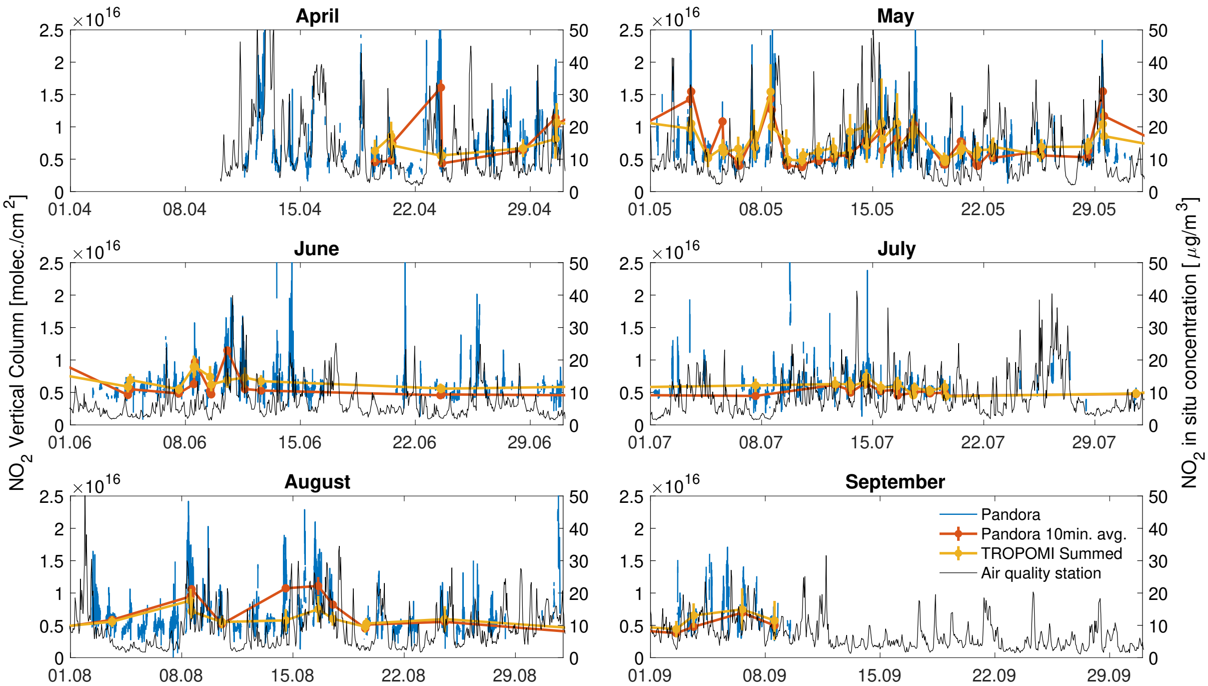

Figure 2. Time series of co-located NO_2 total/summed columns during the period 15.4.–30.9.2018. The blue line indicates all available Pandora observations; the red line indicates Pandora observations averaged 10 minutes before and after S5P's overpass; the yellow line indicates TROPOMI summed columns of the pixels located above the ground-based Pandora station.

The effect of using CAMS regional ENSEMBLE instead of TM5-MP a-priori NO_2 profiles (as used in the standard product) in the calculation of TROPOMI VCDs was analysed by calculating an alternative summed column using the ratio (R) between the tropospheric column retrievals derived from the CAMS and TM5-MP NO_2 profile shapes, computed on the CAMS-regional grid with 0.1° resolution. For each available orbit we used the value of R in the CAMS grid pixel that included the Pandora station. The CAMS a-priori summed column was then calculated from the tropospheric and stratospheric NO_2 VCDs of the standard L2 product as

$$\mathrm{VCD}_{\text{summed, CAMS}} = \mathrm{R} \times \mathrm{VCD}_{\text{tropos, TM5-MP}} + \mathrm{VCD}_{\text{stratos, TM5-MP}}\,. \tag{3}$$

The new TROPOMI-CAMS summed columns calculated using equation (3) were then also compared to the Pandora total columns, and the results are presented in table 2 and figure 7. Apart from these two instances, all tables and figures in this paper use standard TROPOMI data products (i.e. based on TM5-MP a-priori profiles).

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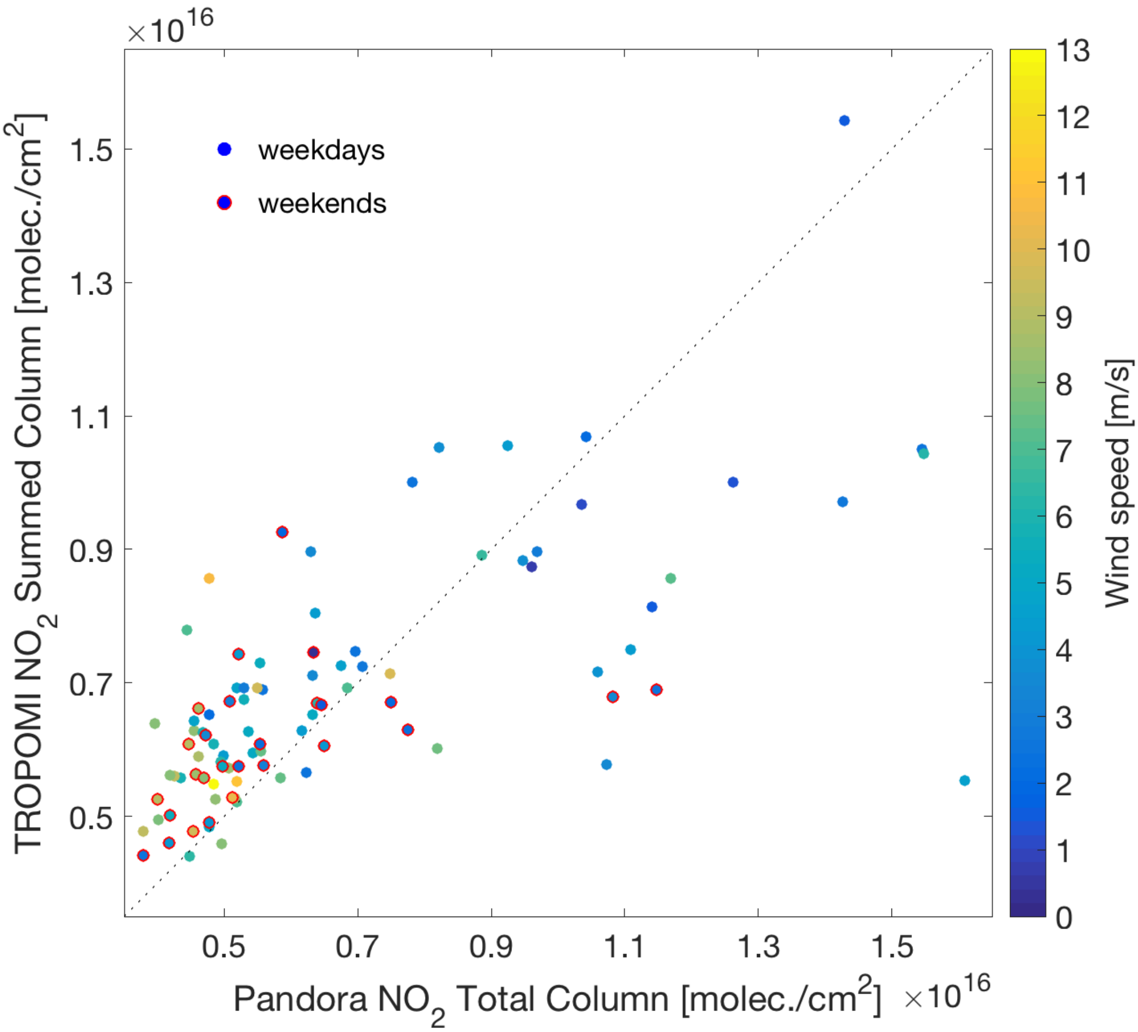

Figure 3. Scatter plot between Pandora and TROPOMI vertical columns. Filled colours indicate the corresponding wind speed, while red circles correspond to weekend overpasses. The 1:1 line is plotted as dotted line.

3 Results

Figure 2 shows the time series of the NO_2 measurements used in the analysis, covering the period April–September 2018. The Pandora NO_2 total columns are shown in their original time resolution (blue line) as well as averaged 10 minutes around the S5P overpass (red line). The latter are used in the quantitative comparison to the TROPOMI NO_2 summed columns (yellow line). We note that S5P often has two valid overpasses per day (ranging from 12 to 15 local time) at the latitude of Helsinki

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Table 1. Statistics of the comparison between TROPOMI and Pandora NO_2 total columns. The values outside (inside) the parentheses are obtained for Pandora retrievals with at least medium (high) quality.

	MRD[a] (%)	MD[b] ($\times 10^{14}$ molec. cm^{-2})	r[c]	slope[d]	n[e]
all data	9.9 ± 2.6 (10.1 ± 3.6)	1.2 ± 2.2 (0.8 ± 3.2)	0.68 (0.66)	0.51 (0.50)	94 (56)
weekdays	9.0 ± 3.3	0.2 ± 2.9	0.68	0.51	67
weekends	12.1 ± 4.4	3.8 ± 3.2	0.46	0.34	27
Pandora high[f]	-28.2 ± 4.8	-36.0 ± 7.0	0.31	1.80	15
Pandora low[g]	17.1 ± 2.2	8.3 ± 1.2	0.72	0.95	79

[a] Mean Relative Difference; [b] Mean Difference; [c] Correlation coefficient; [d] Orthogonal linear fit slope; [e] Number of collocations; [f] Pandora NO_2 total columns $\geq 10^{16}$ molec. cm^{-2}; [g] Pandora NO_2 total columns $< 10^{16}$ molec. cm^{-2}.

(60°N). The hourly NO_2 surface concentrations measured at Kumpula AQ station are also shown on the right hand y-axis (black line). The Pandora total columns and the surface concentrations show similar peaks and day-to-day variability (blue and black line, respectively), which shows how the Pandora observations are sensitive to the changes in the NO_2 levels occurring at the surface. We note that the collocated TROPOMI and Pandora vertical columns (yellow and red line, respectively, in Fig. 2) also mostly follow the same day-to-day variability. The largest differences between TROPOMI and Pandora vertical columns, with TROPOMI smaller than Pandora, correspond to relatively high NO_2 enhancements measured at the surface (black line in Fig. 2). This is expected, as the comparatively large size of the TROPOMI pixels leads to greater spatial averaging compared to the Pandora field-of-view.

In order to further compare satellite- and ground-based collocated observations, Figure 3 shows the scatter plot between Pandora and TROPOMI total columns from the overpasses presented in Fig. 2. The colour of the filled dots indicates the wind speed, and the red circles correspond to weekend observations. The weekend overpasses fall mostly into the bottom-left area of the scatter plot, corresponding to relatively small NO_2 total columns from both Pandora and TROPOMI retrievals. This is expected due to the NO_2 weekly cycle over urban sites, i.e. reduced polluting emissions from traffic during the weekend compared to the weekdays. Furthermore, the overpasses corresponding to high wind speed values (green-yellow dots in Fig.3) also fall into the bottom-left area of the scatter plot. In these cases, the dilution by the wind acts to reduce the NO_2 levels. Overall, the data points are quite close to the one-to-one line, except for some cases with elevated NO_2 total columns measured by Pandora. These cases correspond to NO_2 enhancements with small wind speed (below 3 m s^{-1}), when the spatial dilution effect of TROPOMI's ground footprint as compared to Pandora's narrow field-of-view is especially pronounced.

Table 1 summarises the results of the comparison between TROPOMI and Pandora in terms of mean relative difference (MRD), mean difference (MD), correlation coefficient (r), orthogonal linear least squares fit slope and number of overpasses (n). The overall MRD and MD values are (9.9 ± 2.6) % and $(1.2 \pm 2.2) \times 10^{14}$ molec. cm^{-2}, respectively, meaning that on average TROPOMI slightly overestimates the NO_2 total columns. The correlation coefficient is high ($r = 0.68$). When considering only weekdays, the MD and MRD values become slightly smaller (MRD=(9.0 ± 3.3) %). This is expected, as weekday observations contain a number of collocations where the difference between TROPOMI and Pandora vertical columns

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is exceedingly negative (Fig. 3), corresponding to NO_2 enhancements measured by Pandora. Correspondingly, the MRD and MD values for the weekend (typically associated with lower NO_2 levels) are larger. When taking into account only overpasses with Pandora NO_2 columns larger than 10^{16} molec. cm^{-2}, the bias becomes exceedingly negative (about −28 % or $(-36.0 \pm 7.0) \times 10^{14}$ molec. cm^{-2}), meaning that TROPOMI underestimates the NO_2 total columns when NO_2 enhancements occur. When considering overpasses below that threshold, the bias is positive (about 17 %). These two effects partially cancel each other when the data set is considered as a whole. We also note that taking into account only Pandora retrievals with the highest quality flagging (0 or 10) does not have a substantial effect on the results of the comparison (shown in parentheses in Tab. 1), but it reduces the amount of data available for the comparison by about 30 % (as compared to the case where also medium quality data are included).

Figure 4 shows how the choice of the overpass criteria affects the calculated MD value (a similar plot for the MRD is shown in Figure S2 of the Supplement). In the analysis presented so far we have included measurements from only those TROPOMI pixels which include the Pandora ground-based station. It is also possible to average the contribution from all those pixels which fall within a certain distance from the station. Figure 4 (upper panel) shows how the MD gradually shifts towards negative values when the radius increases from 5 to 30 km. This suggests that averaging over a larger area causes the resulting TROPOMI vertical columns (used in the comparison) to become smaller than those obtained from the single overlaying pixel because of the inhomogeneous spatial distribution of NO_2, so that the mean concentrations decrease with increasing distance. The MD (and MRD) value for the overlaying pixel criterion is very similar to the value obtained for the distance of 5 km, even if the number of collocations is not exactly the same. The overall effect of the spatial collocation choice stays within about 6 percentage points (or 6×10^{14} molec. cm^{-2}).

Similarly, Fig. 4 (lower panel) shows how the MD value changes when the Pandora observations are averaged over an increasing time range, from 5 to 55 min around the overpass time of the satellite. The MD value increases with increasing temporal averaging interval. Averaging over an increasing time range generally slightly reduces the Pandora total column values used in the comparison with TROPOMI, making the MD more positive. The variability remains around 2 percentage points (or 3×10^{14} molec. cm^{-2}).

In order to better compare the temporal variability of the NO_2 vertical columns and surface concentrations, we employ a simple empirical conversion based on the linear regression between Pandora vertical columns and surface concentrations measured at the Kumpula AQ site, at the satellite overpass time (Fig. 5, left panel). From the results of the linear fit (showing high correlation, $r = 0.71$), we convert the surface concentrations into total columns and compare the results to the TROPOMI and Pandora time series (Fig. 5, right panel). We note how the three datasets show a very similar temporal variability, with NO_2 peaks occurring during the same days. We particularly note NO_2 enhancements in May and during the first half of August.

We also analyse the NO_2 weekly cycle as seen from the different datasets. Figure 6 shows the Pandora NO_2 total columns, TROPOMI summed and tropospheric columns and surface concentrations at the Kumpula air quality station as a function of the day of the week. The data are normalised by the corresponding weekly mean value. We note that all datasets show smaller values on Saturdays and Sundays, as expected from the weekly cycle of NO_x emissions typical of urban sites. The NO_2 surface concentrations show about 50 % smaller values in the weekend compared to the weekly average, while TROPOMI

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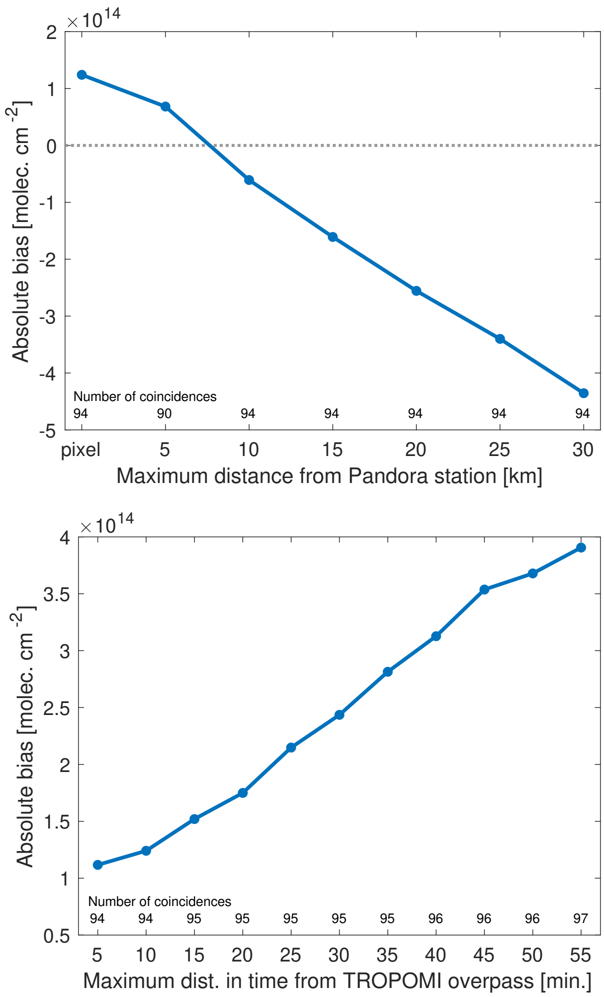

Figure 4. Mean absolute difference between TROPOMI and Pandora total columns as a function of the maximum distance between the centre of the pixel and the ground-based station (upper panel), and as a function of the maximum time difference from TROPOMI overpass (lower panel). The number of coincidences for different collocation criteria are shown above the x-axis. Note that in the upper panel we also require that the TROPOMI pixel above Pandora station contains a valid measurement (QA value >0.75). Thus the number of coincidences does not increase with distance.

tropospheric columns are about 30 % lower. Pandora and TROPOMI summed NO_2 vertical columns are also lower in the weekends (compared to the corresponding weekly means), but only by about 15–20 %. This is because no weekend effect is expected in the stratospheric fraction of the NO_2 column. Surface NO_2 concentration measurements can be expected to show a larger difference between weekend and weekdays due to their greater sensitivity to changes in polluting emissions at the surface (especially from traffic in the urban environment). The results are consistent with those found using nine years of OMI NO_2 observations in Helsinki (Ialongo et al., 2016).

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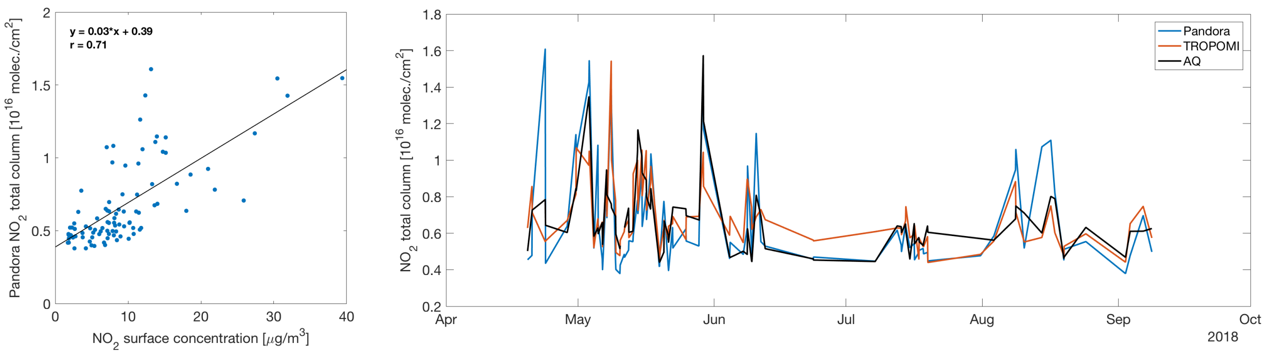

Figure 5. NO_2 time series from Pandora, TROPOMI and Kumpula AQ station at the satellite overpass time (right panel). The surface concentrations are empirically converted to total columns using the results of the linear regression between Pandora total columns and surface concentration data (left panel).

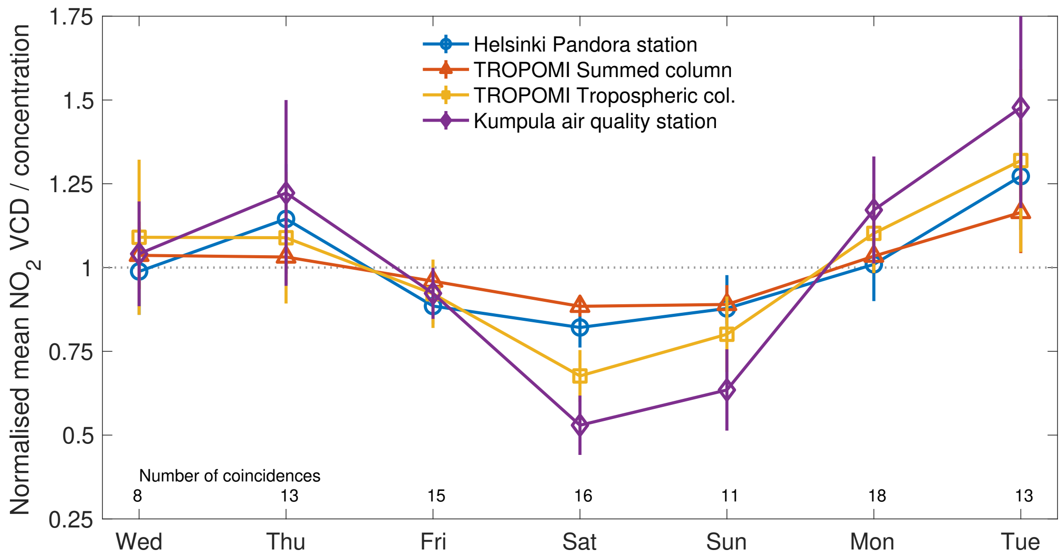

Figure 6. NO_2 weekly cycle in Helsinki. The time-averaged Pandora total columns (blue line), TROPOMI summed (red line) and tropospheric (yellow line) columns, and surface concentrations at the Kumpula AQ site (purple line) for each day of the week are shown. The values have been normalised by the corresponding weekly mean.

Finally, we evaluate the effect of using the NO_2 a-priori profiles derived from the high-resolution CAMS regional ENSEMBLE model, instead of profiles from the TM5-MP CTM as used in TROPOMI's standard product, in the calculation of NO_2 vertical column densities. In Fig.7 we compare both the standard product summed columns and the summed columns derived using the CAMS a-priori profiles, calculated as described in Section 2.3, to the Pandora total columns (analogously to Fig. 3). Only those overpasses (n=75) for which both a-priori summed columns were available were included in the comparison. The

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Table 2. Statistics of the comparisons between TROPOMI summed columns calculated using two different a-priori NO_2 profiles (TM5-MP and CAMS regional ENSEMBLE) and Pandora total columns during 30.4.–30.9.2018.

	MRD^a (%)	MD^b ($\times 10^{14}$ molec. cm^{-2})	r^c	slope^d	n^e
TM5-MP	11.5 ± 2.7	3.1 ± 2.0	0.74	0.52	75
CAMS	14.0 ± 2.6	4.9 ± 1.8	0.80	0.58	75

^{a}Mean Relative Difference; ^{b}Mean Difference; ^{c}Correlation coefficient; ^{d}Orthogonal linear fit slope; ^{e}Number of collocations.

statistics are presented in Table 2 and the corresponding time series in Fig. S3 of the supplement. The comparison shows that the largest differences between the two summed columns are mostly found in cases of relatively high concentrations. In these cases, the use of CAMS profiles generally increases the TROPOMI summed columns and reduces the difference between TROPOMI and Pandora. On the other hand, in cases of low concentrations, where TROPOMI tends to overestimate the VCDs compared to Pandora, the use of CAMS a-priori profiles slightly worsens the agreement with Pandora by increasing the positive bias. Because the largest improvement is achieved for relatively high concentrations and negative biases becoming less negative, the overall MRD value increases from 11.5 % to 14 % (Table 2). For this smaller subset of 75 co-locations with Pandora the correlation between TM5-MP summed columns and Pandora is 0.74 and the slope of an orthogonal linear fit is 0.52. Using the CAMS profiles improves the agreement with Pandora in terms of correlation and slope, with their values increasing to 0.80 and 0.58, respectively. The time series in Fig. S3 in the supplement further shows how using the high-resolution CAMS profiles increases the TROPOMI tropospheric columns so that the summed columns (yellow line) become closer to Pandora's peak values (blue line), corresponding to episodes of NO_2 enhancement.

4 Conclusions

We showed the results of the comparison between satellite-based TROPOMI/S5P NO_2 products and ground-based observations at a medium-sized urban site, Helsinki (Finland). We find that the differences between the total columns derived from the TROPOMI and Pandora instruments are on average less than 10 % (or 1.2×10^{14} molec. cm^{-2}), which is well below the requirements for TROPOMI observations (25–50 % for the NO_2 tropospheric column; ESA, 2017). We also note that the day-to-day and weekly NO_2 variability (typical of urban sites) is reproduced well by the TROPOMI retrievals, similarly to Pandora and in situ surface observations from the local air quality station. This confirms that the satellite-based TROPOMI/S5P NO_2 retrievals are sensitive to changes in air pollution levels occurring at the surface.

In general, we find that TROPOMI NO_2 summed columns are smaller than Pandora total columns for relatively high concentrations, while low values are overestimated. We find this partially related to the TM5-MP model profile shapes used in the TROPOMI retrieval to compute the tropospheric air-mass factors and thus the tropospheric vertical columns. Because of the relatively coarse resolution of the TM5-MP a-priori profiles in the standard product, TROPOMI tropospheric columns are expected to have a negative bias over polluted areas where the peak in the NO_2 profile is close to the surface, and where the

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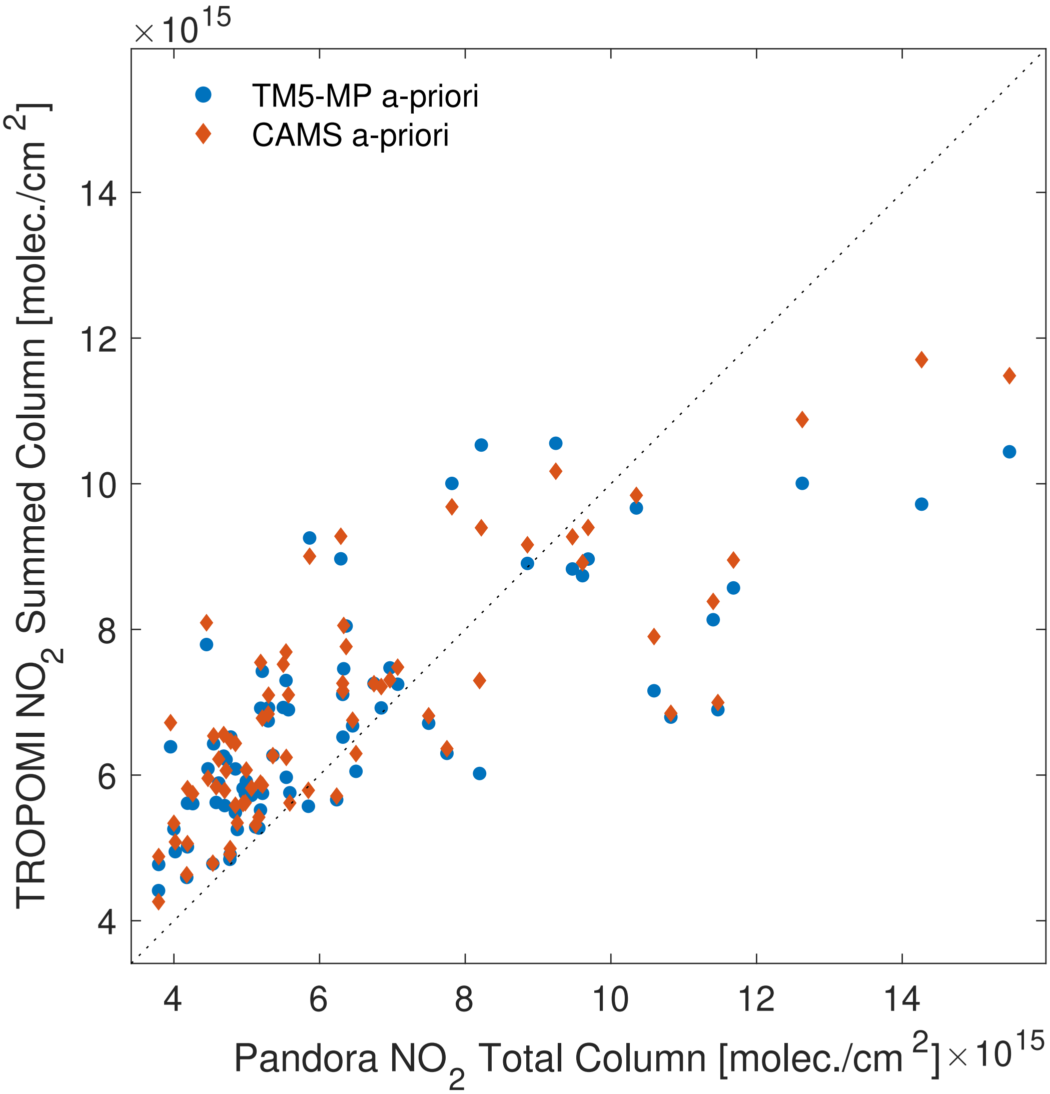

Figure 7. Scatter plot between Pandora and TROPOMI summed columns derived using CAMS regional ENSEMBLE and TM5-MP a-priori NO_2 profiles (blue dots and red diamonds, respectively). The comparison includes only those overpasses for which both summed columns were available at the same time during the time interval 30.4.–30.9.2018. The 1:1 line is plotted as dotted line.

boundary layer column is underestimated in the a-priori. Also, the time variability of the column amounts at the measurement site may be underestimated due to the a-priori. In the same way, the concentrations away from major sources may be somewhat overestimated. In Helsinki we find that replacing the original profiles with those derived from the high-resolution regional CAMS ensemble model increases the TROPOMI NO_2 tropospheric columns and reduces the discrepancy between TROPOMI and Pandora VCDs for situations with relatively high NO_2 concentrations.

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As compared to previous satellite-based instruments such as OMI, the mean bias against ground-based observations in Helsinki is of the same order of magnitude while the correlation coefficient is generally higher (r = 0.68 for TROPOMI and r = 0.5 for OMI, see Ialongo et al., 2016). The correlation between Pandora and TROPOMI NO_2 retrievals is also in line with the results obtained by Griffin et al. (2019) over the Canadian oil sands.

Overall, the analysis of TROPOMI NO_2 observations in the Helsinki area shows high correlation with ground-based observations, as well as demonstrates TROPOMI's capability to properly reproduce the temporal (day-to-day and weekly) variability of the surface NO_2 concentrations. This is a confirmation that satellite-based observations can bring additional information on the temporal and spatial variability of NO_2 in the neighbourhood of major cities, in addition to traditional air quality measurements.

Data availability. The re-processed TROPOMI data before the 30 April 2018 were downloaded from the Sentinel-5P Expert Users Data Hub (https://s5pexp.copernicus.eu/dhus) as part of the S5P validation team activities, and after that date from the S5P Pre-Operations Data Hub (https://s5phub.copernicus.eu/dhus). Pandora #105 total column data belong to the Pandonia network and are available at http://pandonia.net/data/. Surface concentration data at the Kumpula air quality station were obtained on request from the FMI database. The wind data were part of the ECMWF ERA5 reanalysis product and were downloaded from https://cds.climate.copernicus.eu.

Competing interests. No competing interest are present.

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