



# Underestimation of Column NO<sub>2</sub> Amounts from the OMI Satellite Compared to Diurnally Varying Ground-Based Retrievals from Multiple Pandora Spectrometer Instruments

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#### 5 Abstract

Retrievals of Total Column NO<sub>2</sub> (TCNO<sub>2</sub>) are compared for 14 sites from the Ozone Measuring 6 Instrument (OMI using OMNO2-NASA v3.1) on the AURA satellite and from multiple ground-7 based PANDORA spectrometer instruments making direct-sun measurements. Six of these sites 8 9 with multi-year PANDORA data records are in the Northern Hemisphere (Busan, Seoul, 10 Washington DC, Waterflow New Mexico, Boulder Colorado, and Mauna Loa) and one site in the Southern Hemisphere (Buenos Aires Argentina). The first four of these sites and Buenos Aires 11 12 frequently have high TCNO<sub>2</sub> (TCNO<sub>2</sub> > 0.5 DU) and are likely have significant air quality problems that can affect human health. Eight additional sites have shorter term data records in the US and 13 South Korea. One of these is a one-year data record from a highly polluted site at City College in 14 New York City with pollution levels comparable to Seoul, South Korea. The result is that on a 15 16 weekly or monthly average basis, OMI almost always underestimates the amount TCNO<sub>2</sub> by 50 17 to 100%, while occasionally the daily OMI value exceeds that measured by PANDORA at very clean sites. OMI estimated air mass factor, surface reflectivity, and the OMI 24x13 km<sup>2</sup> FOV (field 18 of view) are three factors that can cause OMI to underestimate TCNO<sub>2</sub>. Because of the local 19 inhomogeneity of NO<sub>2</sub> emissions, the large OMI FOV is the most likely factor when comparing 20 OMI TCNO<sub>2</sub> to retrievals from the small PANDORA effective FOV calculated from the solar 21 diameter of 0.5°. As part of air quality assessments, OMI always misses the frequently much 22 23 higher values of TCNO<sub>2</sub> that occur after the OMI overpass time.

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#### Key Words: Nitrogen dioxide, OMI, PAN, PANDORA, ground-based, satellite

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# Underestimation of Column NO<sub>2</sub> Amounts from the OMI Satellite Compared to Ground-Based Retrievals from Multiple Pandora Spectrometer Instruments

#### 28 1.0 Introduction

29 Retrieval of Total Column NO<sub>2</sub> (TCNO<sub>2</sub>) from the Ozone Monitoring Instrument (OMI) has been a 30 scientific success story foe the past 14 years. Near total global coverage of the well-calibrated OMI has 31 enabled observation of all the regions where NO<sub>2</sub> is produced and has permitted monitoring of the 32 changes during the 2004 to 2019 period, especially in regions where there is heavy and growing industrial 33 activity (e.g., China and India). TCNO<sub>2</sub> amounts (data used: OMNO2-NASA v3.1) retrieved from OMI over 34 various specific land locations show a strong underestimate compared to co-located Pandora 35 Spectrometer Instruments (the abbreviation PAN is used for graph and table labels). The OMI TCNO2 36 overpass time underestimate compared to ground-based measurements has been previously reported at 37 a few specific locations (Bechle, 2013; Lamsal et al., 2015; Ialongo et al., 2017; Kollonige, et al., 2018; 38 Goldberg et al., 2018; Herman et al., 2018). For any location, the OMI overpass local standard time consists 39 of the central overpass near the 13:30 hour equator crossing solar time and occasionally a side viewing 40 overpass from adjacent orbits within ±90 minutes of the central overpass time. Independently from 41 instrument calibration and retrieval errors, there are two specific aspects to the underestimation of TCNO<sub>2</sub> 42 pollution levels. First, the mid-day OMI observations do not see the large diurnal variation of TCNO<sub>2</sub>, and 43 second, the very large field of view (FOV) footprint 13 x 24 km<sup>2</sup> at OMI nadir view tends to average regions 44 of high pollution with those from lower pollution areas.

45 There are other possible systematic retrieval errors with OMI TCNO<sub>2</sub>. The largest of these is 46 determining the air mass factor (AMF) needed to convert slant column measurements into vertical column 47 amounts. Accurately determining the AMF for TCNO<sub>2</sub> requires a-priori knowledge of the NO<sub>2</sub> profile shape 48 and using the correct surface reflectivity Rs. Currently Rs is found using a statistical process of sorting 49 through years of data to find relatively clear-sky scenes for each location (Kleipool, et al., 2008; O'Byrne 50 et al., 2010). Boersma et al., 2004 gave a detailed error analysis for the various components contributing 51 OMI TCNO<sub>2</sub> retrievals resulting an estimated "retrieval precision of 35-60%" in heavily polluted areas 52 dominated by determining the air mass factor. An improved V2.0 DOMINO retrieval (Boersma et al., 2011) 53 algorithm reduced the retrieval errors while increasing the estimated airmass factor, which reduces the 54 retrieved TCNO<sub>2</sub> up to 20% in winter and 10% in summer. The current version of OMNO2-NASA and v2.0 55 DOMINO are generally in good agreement (Marchenko et al., 2015; Zara et al., 2018). However, the 56 OMNO2-NASA retrievals are 10 to 15% lower than the v2.0 DOMINO retrievals. A subsequent detailed 57 analysis of surface reflectivity (Vasilkov et al., 2017) shows that retrieval of TCNO2 in highly polluted areas (e.g., some areas in China) can increase by 50% with the use of geometry-dependent reflectivities, but 58 59 only increase about 5% in less polluted areas. For PANDORA, calculation of the solar viewing AMF is a 60 simple geometric problem (AMF is approximately proportional to the cosecant of the solar zenith angle 61 SZA) and is independent of  $R_s$  (Herman et al., 2009). For a polluted region with TCNO<sub>2</sub> =  $5.34 \times 10^{16}$ 62 molecules/cm<sup>2</sup> or 2 DU, the PANDORA error is expected to be about ±2.5% with the largest uncertainty 63 coming from an assumed amount of stratospheric  $TCNO_2 = 0.1 DU$ .





Accurate satellite TCNO<sub>2</sub> retrievals (and for other trace gases) are important in the estimate of the effect of polluted air containing NO<sub>2</sub> on human health (Kim and Song, 2017 and references therein), especially from the viewpoint of NO<sub>2</sub> as a respiratory irritant and precursor to cancer (Choudhari et al., 2013). Since NO<sub>2</sub> is largely produced by combustion, satellite observations of NO<sub>2</sub> serve as a proxy for changing industrial activity. Another important application requiring accurate measurements of the amount of TCNO<sub>2</sub> and its diurnal variation is atmospheric NO<sub>2</sub> contribution to nitrification of coastal waters (Tzortziou et al., 2018).

71 We show that the use of OMI TCNO<sub>2</sub> for estimating local air quality and coastal nitrification on a 72 global basis is misleading for most polluted locations, and especially on days when the morning or 73 afternoon amounts are higher than those occurring at the OMI overpass time near 13:30 hours standard 74 time. OMI TCNO<sub>2</sub> data are extremely useful for estimating regional pollution amounts and for assessing 75 long-term changes in these amounts. Modelling studies (Lamsal et al., 2017 Fig. 1) based on the Global 76 Modelling Initiative model (Strahan et al., 2007) simulating TCNO<sub>2</sub> diurnal variation over Maryland USA 77 (37-40°N, 74-79°W) shows a late afternoon peak and shows that the stratospheric component does not 78 substantially contribute to this peak. Boersma et al. (2016) show that sampling strategy can cause 79 systematic errors between OMI TCNO<sub>2</sub> and model TCNO<sub>2</sub> with satellite results being up to 20% lower than 80 models. Duncan et al., (2014) reviews the applicability of satellite TCNO<sub>2</sub> data to represent air quality and 81 notes that TCNO<sub>2</sub> correlates well with surface levels of NO<sub>2</sub> in industrial regions and states that the portion 82 of TCNO<sub>2</sub> in the boundary layer could be over 75% of the total vertical column depending on NO<sub>2</sub> altitude 83 profile shape.

84 This paper presents 14 comparisons between retrieved OMI TCNO<sub>2</sub> overpass values that are co-85 located with PANDORA TCNO<sub>2</sub> amounts from various locations in the world. Six of the comparisons are 86 where PANDORAs have long-term data (1-year or longer) records. The comparisons are done using daily 87 data matched to the OMI overpass times ±6 minutes and with monthly running averages calculated using 88 Lowess(f) (Locally Weighted least squares fit to a fraction f of the data points, (Cleveland, 1981)) of time 89 matched TCNO<sub>2</sub>. This is combined with a discussion and presentation of data on the effect of diurnal 90 variation that are always missed at the local OMI mid-day overpass times. We show that OMI TCNO<sub>2</sub> 91 values are systematically lower than PANDORA values at sites with significant pollution (TCNO<sub>2</sub> > 0.3 DU). 92 We also present a unique view of a year of daily diurnal variation of TCNO<sub>2</sub> at two sites, Washington DC 93 and New York City, which are similar to other polluted locations.

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#### 95 2.0 Brief Instrument Descriptions

For the purposes of TCNO<sub>2</sub> retrievals, both OMI and PANDORA are spectrometer-based instruments using nearly the same spectral range and similar spectral resolution (about 0.5 nm). Both use spectral fitting retrieval algorithms that differ (Boersma et al. 2011; Herman et al., 2009) because of the differences between direct-sun viewing retrievals (PANDORA) and above the atmosphere downward viewing retrievals (OMI). The biggest difference is with the respective fields of view, 13 x 24 km<sup>2</sup> at OMI nadir view and larger off-nadir FOV compared to the much smaller PANDORA FOV (1.2<sup>o</sup>) measured in m<sup>2</sup> with the precise value depending on the NO<sub>2</sub> profile shape and the solar zenith angle. For example, if most





103 of the TCNO<sub>2</sub> is located below 2 km, then the PANDORA FOV is approximately given by 104  $(1.2\pi/180)(2/\cos(SZA))$ , which for SZA = 45° is about 59x59 m<sup>2</sup>.

#### 105 **2.1 OMI**

106 OMI is an east-west side (2600 km) and nadir viewing polar orbiting imaging spectrometer that 107 measures the earth's backscattered and reflected radiation in the range 270 to 500 nm with a spectral 108 resolution of 0.5 nm. The polar orbiting side scanning capabilities produce a pole to pole swath that is 109 about 2600 km wide displaced in longitude every 90 minutes by the earth's rotation to provide coverage 110 of nearly the entire sunlit Earth once per day at a 13:30 solar hour equator crossing time with spatial gaps 111 at low latitudes. OMI provides full global coverage every 2 to 3 days. Additional gaps are caused by a problem with the OMI CCD, "row anomaly" (Torres et al., 2018) that effectively reduces the number of 112 113 near-nadir overpass views. A detailed OMI instrument description is given in Levelt et al. (2006). TCNO2 is 114 determined in the visible spectral range from 405 to 465 nm where the NO<sub>2</sub> absorption spectrum has the 115 maximum spectral structure and where there is little interference from other trace gas species (there is a 116 weak water feature in this range). OMI overpass data are available for many sites (currently 719) from the following NASA website. https://avdc.gsfc.nasa.gov/index.php?site=666843934&id=13 117

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#### 119 2.2 PANDORA

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121 PANDORA is a sun-viewing instrument for SZA  $< 80^{\circ}$  that obtains about 4000 spectra for clear-sky 122 views of the sun in 20 seconds for each of two ranges UV (290 - 380 nm using a UV340 bandpass filter) 123 and visible plus UV (280 – 525 nm using no filter). The overall measurement time is about 80 seconds 124 including a 20 second dark-current measurements between each spectral measurement throughout the 125 day. About 4000 clear-sky spectra for the UV and visible portions are separately averaged together to achieve very high signal to noise ratios (SNR). The UV340 filter for UV portion of the spectra reduces stray 126 127 light effects from the visible wavelength range. A detailed description of PANDORA and its SNR is given 128 in Herman et al., (2009; 2015). The effect of moderate cloud cover (reduction of observed signal by a 129 factor of 8) in the PANDORA FOV on TCNO<sub>2</sub> retrievals is small (Herman et al., 2018). Cloud cover also 130 reduces the number of measurements possible in 20 seconds, which potentially increases the noise level. 131 PANDORA is driven by a highly accurate sun tracker that points an optical head at the sun and transmits 132 the received light to an Avantes 2048 x 32 pixel CCD spectrometer (AvaSpec-ULS2048 from 280 - 525 nm 133 with 0.6 nm resolution) through a 50 micron diameter fiber optic cable. The estimated TCNO<sub>2</sub> error is approximately 0.05 DU (1 DU = 2.69 x 10<sup>16</sup> molecules cm<sup>-2</sup>) out of a typical value of 0.3 DU in relatively 134 135 clean areas and over 3 DU in highly polluted areas. PANDORA data are available for 250 sites. Some sites 136 have multi-year data sets, but many of these sites are short-term 137 campaign sites. https://avdc.gsfc.nasa.gov/pub/DSCOVR/Pandora/DATA 01/.

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#### 139 3.0 Overpass Comparisons and Diurnal Variation of TCNO<sub>2</sub>

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NO<sub>2</sub>'s contribution to air quality at the Earth's surface is usually a proportional function of TCNO<sub>2</sub>
 that varies with the time of day and with the altitude profile shape. Most of the NO<sub>2</sub> amount is usually





located between 0 and 3 km altitude with a small amount (about 0.1 DU) in the upper troposphere and
stratosphere. Because of the relatively short chemical lifetime, 3-4 hours (Liu et al., 2016), in the lower
atmosphere, most of the NO<sub>2</sub> is located near (0 to 20 km) its sources (industrial activity, power generation,
and automobile traffic). At higher altitudes or in the winter months, the life time of NO<sub>2</sub> is longer
permitting transport over larger distances from its sources.

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During the South Korean campaign (KORUS-AQ) in the spring of 2016 the diurnal variations of TCNO<sub>2</sub> vs days of the year DOY were determined for 6 sites (Herman et al., 2018), one of which is reproduced here (Fig. 1) for the city of Busan showing relatively low values of TCNO<sub>2</sub> in the morning (0.5 DU), moderately high values during the middle of the day (1.3 DU), and very high values in the afternoon (2 to 3 DU). Of these data, OMI only observes midday values near the 13:30 time marked on the Local Time axis of Fig.1 thereby missing very high values (2 to 3 DU) that frequently occur later in the afternoon coinciding with times when people are outdoors returning from work.

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Fig 1 Diurnal variation of TCNO<sub>2</sub> measured at Pusan University in Busan South Korea

Fig. 2. Monthly average values of TCNO<sub>2</sub> for OMI and PANDORA at OMI overpass times

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In addition to missing the TCNO<sub>2</sub> diurnal variation, the OMI values are about half those observed by PANDORA (Fig. 2) at the OMI overpass time, so that using OMI values to estimate NO<sub>2</sub> pollution seriously underestimates the air quality problem even at midday. The shaded area in Fig.2 corresponds to the period covered in the KORUS-AQ campaign 7 April to 11 June 2016 shown in Fig. 1. An extended time series for Busan location is shown in Fig. 3.







Fig. 3 Extended time series for Busan. Left Panel: individual matching PANDORA and OMI data points for the overpass time ± 6 minutes. Right Panel: monthly averages.

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Because of the different effective NO<sub>2</sub> FOV of PANDORA (measured in meters<sup>2</sup>) while tracking the moving sun position located in the heart of Busan (FOV distance d < 5 km for an SZA < 70° used for TCNO<sub>2</sub> retrievals), both the daily (Fig. 3, left panel) and PANDORA monthly average variation (Fig. 3, right panel), obtained at the OMI overpass time, differs from the variation in the OMI TCNO<sub>2</sub> because of the much larger OMI FOV (13 x 24 km<sup>2</sup> at OMI nadir view) retrieval. Because of this, the OMI time series has low correlation (r<sup>2</sup> = 0.1) with the PANDRA time series.

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The extended OMI vs PANDORA time series from 2012 – 2017 for Busan (Fig. 3) shows the same magnitude of differences seen during the KORUS-AQ period. A similar OMI vs PANDORA plot for total column ozone TCO<sub>3</sub> (Appendix Fig A1) shows good agreement between PANDORA and OMI indicating that the PANDORA instrument was operating and tracking the sun properly. Because the spatial variability of TCO<sub>3</sub>, which is mostly in the stratosphere, is much less than for TCNO<sub>2</sub>, the effect of different FOV's is minimized.

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The same type of differences, TCNO<sub>2</sub>(PAN) > TCNO<sub>2</sub>(OMI), are seen at a wide variety of sites (e.g., see Fig.4). Comparing extended Busan multi-year time series, some broad-scale correlation can be seen with peaks in February 2013, January 2014, and in 2016. The data from Busan are different than from many sites, since Busan is located very near the ocean causing a portion of the OMI FOV to be over the unpolluted ocean areas, whereas PANDORA is located inland in an area of dense automobile traffic and quite near mountains capable of trapping air.

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Fig. 4. PANDORA compared to OMI. Extended  $TCNO_2$  overpass time series for Mauna Loa Observatory, Hawaii, NASA Headquarters, Washington DC, and Waterflow, New Mexico.







Fig. 5. PANDORA compared to OMI. Extended TCNO<sub>2</sub> overpass time series for Seoul South Korea, Boulder, Colorado, and Buenos Aires, Argentina (Raponi et al. 2017).





TCNO<sub>2</sub>(PAN) comparisons with TCNO<sub>2</sub>(OMI) from Mauna Loa Observatory (Fig. 4) are not those 188 189 that might be expected, since the PANDORA observations are in an area where there are almost no 190 automobile emissions and certainly no power plants, yet PAN > OMI and TCNO<sub>2</sub>(PAN) values are large 191 enough so that the pollution values (0.18 DU) are well above the stratospheric values (approximately 0.1 192 DU). OMI, which mainly measures values over the clean ocean, has an average value of 0.1 DU. The 193 PANDORA values suggest upward airflow from the nearby circumferential ring road and resort areas. The 194 Mauna Loa TCNO<sub>2</sub> values do not show any correlation with the recent increased volcanic activity at Mt. 195 Kilauea after 2016. The calibration of the Mauna Loa PANDORA will be reviewed as part of a general data 196 quality assurance program that is starting with the most recently deployed PANDORA instruments.

197 An interesting inland site is near the very small town of Waterflow, New Mexico (Fig. 4), where 198 two power plants located near the PANDORA site ceased operation on December 30, 2013 (Lindenmaier 199 et al., 2014). According to a quote from AZCentral Newspaper (Tuesday 31 December 2013) "Three coal-200 fired generators that opened in the 1960s near Farmington, N.M., closed Monday as part of a \$182 million plan for Arizona Public Service Co. to meet environmental regulations, the utility reported". The TCNO2 201 202 data suggests that the actual shutdown occurred near October 15, 2013. After the shutdown, air quality 203 improved in the area with TCNO<sub>2</sub> decreasing from 0.4 DU to 0.28 DU. The remaining more efficient 204 generators continued to produce smaller NO<sub>2</sub> emissions. These were shut down at the end of 2016 with 205 little additional observed change in TCNO<sub>2</sub>, since these boilers used NO<sub>2</sub> scrubbers (Dubey at al., 2018 in 206 preparation). A nearby highway (Route 64) about 2 km from the PANDORA site has little automobile 207 traffic.

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Figures 4 and 5 show a variety of different sites, ranging from the Mauna Loa Observatory location at 3.4 km (11,161 feet) on a relatively clean Hawaiian Island surrounded by ocean to a polluted landlocked semi-arid site at Waterflow, New Mexico near a power plant. All the sites considered show a significant underestimate of OMI TCNO<sub>2</sub>. A summary of the monthly average underestimates is given in Tables 1 and 2. For some sites there is evident correlation between the two offset measurements. For example, the PANDORA at NASA Headquarters in Washington DC tracks the OMI measurement quite well on a monthly average basis. Other sites have only short periods of correlation.





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Table 1 Average values of  $TCNO_2$  for PANDORA and OMI from monthly averages in Figs. 4 and 5

Location	PAN (DU)	OMI (DU)
Mauna Loa Hawaii	0.16	0.11
NASA HQ Washington DC	0.34	0.25
Waterflow New Mexico	0.32	0.18
Seoul South Korea	1.2	0.58
Busan South Korea	0.68	0.32
Boulder Colorado	0.27	0.17
Buenos Aires Argentina	0.50	0.26
Average	0.49	0.27

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Table 2 Average values of TCNO<sub>2</sub> for PANDORA and OMI for additional sites

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Location	PAN (DU)	OMI (DU)
Essex Maryland	0.30	0.28
Baltimore Maryland	0.45	0.27
Fresno California	0.42	0.17
Denver La Casa Colorado	0.68	0.19
Gwangju Institute of Science and Technology (GIST) S. Korea	0.42	0.20
Hankuk University Foreign Studies (HUFS) South Korea	0.61	0.51
Average	0.48	0.29

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220 Table 2 contains a summary of sites that were part of short-term Discover-AQ campaigns in 221 Maryland, Texas, California, and Colorado, and two longer-term sites in South Korea. Essex, Maryland is located on the Chesapeake Bay 10 km east of the center of Baltimore. The site is relatively clean (PAN = 222 223 0.3 DU) compared to the center of Baltimore (PAN = 0.45 DU), while OMI measures about the same 224 amounts for both sites (0.28 and 0.27 DU) because the OMI FOV is larger than the distance between the 225 two sites. The Houston Texas site contains 7 months of data from January to July 2013 with widespread 226 NO<sub>2</sub> pollution permitting PANDORA and OMI to measure the same average values even though PANDORA 227 observes episodes on many days when TCNO<sub>2</sub> exceeds 1.5 DU for short periods at times not observed by 228 OMI. Observations in the small city of Fresno, California were during January when agricultural sources of 229 NO<sub>2</sub> were at a minimum (Almaraz, 2018), but automobile traffic in the center of Fresno was significant. In 230 this situation, PANDORA recorded the effect of automobile traffic while OMI averaged the city of Fresno 231 and surrounding fallow agricultural areas. The Denver La Casa location is in the center of the city in an 232 area with high amounts of local automobile traffic and near the Cherokee power generating plant. The 233 result is a high level of average pollution (0.42 DU) while OMI measures both the city center and the 234 surrounding relatively clean plains areas. The HUFS South Korean site is southeast of Seoul in a fairly 235 isolated valley. However, Seoul and its surrounding areas are a widespread transported source of pollution 236 so that both PANDORA and OMI measure elevated TCNO2 amounts. In contrast, the PANDORA GIST site is





237 on the outskirts of a small city in southwestern South Korea with significant traffic. The result is significant 238 amounts of localized TCNO<sub>2</sub> (PANDORA = 0.42) surrounded by areas that produce little NO<sub>2</sub> leading to 239 OMI observing a very clean 0.2 DU. The average of sites in the two tables are similar leading to ratios of 240 PAN/OMI of 1.8 and 1.7 respectively. The estimated 50% increase in OMI retrievals of TCNO<sub>2</sub> from using 241 the geometry-dependent reflectivity (Vasilkov, 2017) for the most polluted sites will narrow the 242 disagreement with PANDORA. For example, OMI Seoul TCNO<sub>2</sub> may become 0.87 DU (PANDORA = 1.2 DU) 243 and Buenos Aires 0.39 DU (PANDORA = 0.5 DU) still underestimating the amount of NO<sub>2</sub> pollution and 244 missing the significant diurnal variation.

245The average percent differences between OMI and PANDORA shown in Fig. 6 are relatively246constant over time for each site with small changes over each multi-year observation period. Of the six247sites, only two of the sites have a statistically significant change in the percent difference (Seoul South248Korea and NASA HQ Washington DC) at the 2-standard deviation level (2σ). The average differences range249from 24% to 46%.

250 For the six sites shown, the average OMI underestimate of  $TCNO_2$  is approximately a factor of 1.8 251 at the overpass time on a monthly average basis with occasional spikes that exceed this amount. The bias 252 values range from 1.1 to 3.6, with higher biases tending to be associated with higher TCNO<sub>2</sub> values. The 253 factor of 1.8 underestimate ignores the frequent large values of TCNO<sub>2</sub> at other times during the day (Fig. 254 7). In addition, averaging TCNO<sub>2</sub>(PAN) over each entire day yields average values for the whole period that 255 are 10 to 20% higher than just averaging over midday values that matched the OMI overpass time. Aside 256 from the absolute magnitude, the short-term variations (over several months) are similar for both OMI 257 and PANDORA although mostly not correlated. If correlation coefficients r<sup>2</sup> are generated from linear fits 258 to scatter plots of TCNO<sub>2</sub> from OMI vs PANDORA, the correlation is mostly poor (Examples,  $r^2 =:$  Seoul 259 0.06, Mauna Loa 0.3 NASA HQ 0.7, see Figs. 4 and 5). Additional sites with shorter PANDORA time series 260 of TCNO<sub>2</sub> show similar behavior.

261 An alternate view of the differences between OMI and PANDORA is provided by forming the 262 percent differences of the daily TCNO<sub>2</sub> values (Fig. 6) in the form 100(OMI – PAN)/PAN. Also shown are 263 the average percent differences and the linear fit slopes in percent change per year of the percent 264 differences over the multi-year period. Of the six sites in shown in Fig. 6, two have statistically significant 265 slopes, Seoul South Korea 2.1±0.5 %/Year and NASA Headquarters in Washington DC 3.4±0.9 %/Year at 266 the 2 $\sigma$  level suggesting a significant area average increase in pollution compared to PANDORA's local 267 values.

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Fig. 6 Percent differences between OMI and PANDORA. The slopes are the absolute change in the percent difference. For example, the Boulder percent difference goes from -31% to -23% over 4 years.

271 3.1 Diurnal Variation at NASA HQ Washington DC





- 272 Figure 7 shows details of the daily diurnal variation of TCNO<sub>2</sub> on the roof of NASA Headquarters
- 273 Washington, DC adjacent to a major cross town highway (I695) for every day during each month of 2015
- for local time vs DOY. The midday observing local standard time for OMI is marked for each graph.



Fig. 7A. TCNO<sub>2</sub> diurnal variation (DU) from January to June, NASA Headquarters Washington, DC from January 2015 to June 2015. The approximate OMI overpass time near 13:30 hours is marked







Fig. 7B TCNO<sub>2</sub> diurnal variation (DU) from July to December, NASA Headquarters Washington, DC from July 2015 to December 2015. The approximate OMI overpass time near 13:30 hours is marked

277 The amount of  $TCNO_2$  is mostly from the adjacent highway and the surrounding urban area with 278 heavy traffic. The relatively moderate  $TCNO_2$  values (0.4 to 0.8 DU) are probably a testament to the 279 effectiveness of catalytic converters mandatory on all US automobiles in such a high traffic area.







Fig. 8A TCNO $_2$  diurnal variation (DU) at CCNY in New York City January to June 2018. The approximate OMI overpass time near 13:30 hours is marked







Fig. 8B TCNO<sub>2</sub> diurnal variation (DU) at CCNY in New York City July to December 2018. The peak near 5 DU occurs on 13 July 2018 between 11:20 and 12:30 EST. The approximate OMI overpass time near 13:30 hours is marked.

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Figure 8 contains the daily TCNO<sub>2</sub> diurnal variability vs DOY for each month measured by a PANDORA from the roof of a building on the CCNY (City College of New York) campus in the middle of Manhattan in New York City (NYC). From the values shown, the pollution levels are quite high, rivaling the





285 pollution levels in Seoul, South Korea. OMI at its mid-day overpass time would detect some of the high-286 level pollution events, but miss many others occurring mostly in the afternoon. There are a significant 287 number of days in all the months where the TCNO<sub>2</sub> levels appear to be low (e.g., blue color in July and 288 October), but the blue color still represents significant pollution levels (TCNO<sub>2</sub>(PAN) > 0.5 DU) that are 289 small only compared to the peak values during the month (TCNO<sub>2</sub>(PAN) > 1 DU). The highest amount of 290 TCNO<sub>2</sub> recorded during 2018 was about 5DU on 13 July 2018 from 11:20 and 12:30 EST (a time with very 291 light winds (1 km/hr) and moderate temperature (25°C). There were many smaller peaks between 2 and 292 3 DU throughout the year.

293 For both Washington DC (Fig. 7) and New York City (Fig. 8) there is strong day-to-day and month 294 to month variability that depends on the local weather and the amount of automobile traffic in the area. 295 High TCNO<sub>2</sub> events occur most often in the afternoon such that the OMI overpass near 13:30 would miss 296 most high TCNO<sub>2</sub> events. Poor air quality affecting respiratory health would be improperly characterized 297 by both the OMI average values being too low (Fig. 4) and by missing the extreme pollution events that 298 occur frequently in the late afternoon. The high value of TCNO<sub>2</sub> that occurred on 5 August (2.2 DU) at 299 07:45 EST for Washington DC is not a retrieval error (SZA less than  $70^{\circ}$ ), but is a one-time anomaly in 2015 300 compared to more usual high values of 1.5 DU with an occasional spike to 2 DU.



Fig. 9 TCNO<sub>2</sub> overpass time series for CCNY in Manhattan, New York City. Panel A: OMI overpass TCNO<sub>2</sub> (Black) compare with OMI (Red). Panel B: Monthly Lowess(0.08) fit to the daily overpass data. Panel C: Percent difference 100(OMI – PAN)/PAN calculated from the data in Panel A





Similar daily diurnal variation graphs of TCNO<sub>2</sub> (Figs. 7 and 8) could be shown for each site. However, the basic idea is the same for each site. OMI underestimates the amount of TCNO<sub>2</sub> because of its large FOV and misses most of the peak events at other times of the day. For some sites, such as Busan and Seoul, the peak values can reach 3 DU and above late in the afternoon, which are never seen by OMI (Herman et al., 2018).

Figure 9 for CCNY is similar to the graphs in Figs. 4 – 6 showing the relative behavior between PANDORA and OMI. However, there is a period in March 2018 when OMI TCNO<sub>2</sub> slightly exceeded that measured by PANDORA. OMI with its large FOV may be seeing part of the chemically driven seasonal variation, while PANDORA is seeing a nearly constant source driven amount mostly from automobile traffic. For most days during 2018, PAN(TCNO<sub>2</sub>) > OMI(TCNO<sub>2</sub>) with the average value for PAN = 0.6 DU and for OMI = 0.4 DU (Fig. 9 Panel B). The percent difference plot shows that there is a systematic increase between PANDORA and OMI TCNO<sub>2</sub> from a value 10% to a value of 52%.

#### 314 4.0 Summary

315 Examination of long-term TCNO<sub>2</sub> monthly average time series from OMI satellite and PANDORA 316 ground-based observations show that OMI systematically underestimates the amount of NO2 in the 317 atmosphere by an average factor of 1.5 to 2 at the local OMI overpass time near the equator crossing time 318 of 13:30±1:30. The OMI underestimate is much larger than error estimates for TCNO<sub>2</sub> retrievals for either 319 PANDORA or OMI. In addition, the PANDORA diurnal time series for every day during a year at each site 320 (only two typical sites are shown in this paper) shows peaks in  $TCNO_2$  that are completely missed by only 321 observing at mid-day. The result is that estimates of air quality related to health effects from OMI 322 observations are strongly underestimated almost everywhere as shown at all the sites with a long 323 PANDORA record. In comparisons to PANDORA, OMI data are mostly uncorrelated or weakly correlated 324 (e.g., Seoul correlation coefficient  $r^2 = 0.06$ , Mauna Loa  $r^2 = 0.3$ ), while NASA HQ in Washington, DC shows a correlation on a seasonal basis (NASA HQ  $r^2 = 0.7$ ) suggesting a wide area coordinated source of NO<sub>2</sub> 325 326 (most likely automobile traffic). The data from CCNY shows some correlation between the locations of the 327 peaks and troughs. Seven short term TCNO<sub>2</sub> time series were examined showing similar results (Table 1), 328 except when the pollution region is widespread as in the Seoul South Korea region. The conclusion is that 329 while OMI satellite TCNO<sub>2</sub> data are uniquely able to assess regional long-term trends in TCNO<sub>2</sub> and provide 330 a measure of the regional distribution of pollutants, the OMI data cannot properly assess local air quality 331 or the effect on human health over extended periods in urban or industrial areas. This will continue to be 332 the case, but to a lesser degree, when the OMI TCNO<sub>2</sub> data are improved by reprocessing with a new 333 geometry-dependent reflectivity (Vasilkov, 2017). The analysis shows that locating PANDORAs at polluted 334 sites could provide quantitative corrections for spatial and temporal biases that affect the determination 335 of local air quality from satellite data. To verify the proper operation of the various PANDORA instruments 336 a similar analysis for Total Column Ozone TCO was performed (see Appendix) and shows close agreement 337 between OMI and PANDORA, with the largest difference occurring for Mauna Loa Observatory at 3.4 km altitude, where PANDORA misses the ozone between the surface and 3.4 km. 338





#### 340

#### 341 Appendix

A1 **Ozone** This section shows the corresponding PANDORA total column ozone (TCO) values compared to OMI TCO for Busan South Korea (Fig. A1) that shows close agreement for the entire 2012 – 2017 period. The different fields of view for OMI and PANDORA have a much smaller effect because of the greater spatial uniformity of stratospheric ozone compared to tropospheric NO<sub>2</sub>. Additional sites are summarized in Table A1. The largest TCO difference (15 DU or 5.6%) occurs for Mauna Loa Observatory (Altitude = 3.4 km) compared to OMI (Average altitude = Sea Level). The close results show that the PANDORA was working properly and pointing accurately at the sun.



Fig. A1 Monthly average values of TCO for OMI and PANDORA at OMI overpass times for Busan South Korea

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	Table A1 Average values of TCO <sub>3</sub> for PANDORA and OMI			
	Location	PAN	OMI	Percent
		(DU)	(DU)	Difference
	Mauna Loa Observatory Hawaii (3.394 km)*	254	269	5.6
	NASA HQ Washington DC (0.02 km)	308	314	1.9
	Waterflow New Mexico (1.64 km)	293	292	0.3
	Yonsei University Seoul South Korea (0.07 km)	317	325	2.5
	Busan University Busan South Korea(0.03 km)	313	315	0.6
	Boulder, Colorado (NOAA Bldg) (1.617 km)	299	302	1.0
	Buenos Aires, Argentina (0.025 km)	279	284	1.8
	Essex, Maryland (0.012 km)	299	301	0.7
	Baltimore, Maryland (0.01 km)	296	296	0.0
	Fresno, California (0.939 km)	306	309	1.0
	Denver La Casa Colorado (1.6 km)	292	294	0.7
	Gwangju Institute of Science and Technology (GIST) S. Korea (0.021 km)	302	307	1.6
	Hankuk University Foreign Studies (HUFS ) South Korea (0.04 km)	318	326	2.5
	City College Manhattan New York City (0.04 km)	316	325	2.8
	Average	299	304	1.6
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353	* OMI observes the sea level value of $TCO_3$			
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365	References
366	Almaraz, Maya,Edith Bai, Chao Wang, Justin Trousdel, Stephen Conley, Ian Faloona and Benjamin Z.
367	Houlton, Agriculture is a major source of NOx pollution in California, SCIENCE ADVANCES, 31,
368	DOI:10.1126/sciadv.aao3477, 2018.
369	
370	Bechle, M. J.; Millet, D. B.; Marshall, J. D. Remote sensing of exposure to NO2: Satellite versus ground-
371	based measurement in a large urban area. Atmos. Environ., 69, 345-353, 2013
372	
373	Boersma, K. F., H. J. Eskes, E. J. Brinksma, Error analysis for tropospheric NO <sub>2</sub> retrieval from space, J.
374	Geophys. Res. Atmos., https://doi.org/10.1029/2003JD003962, 2004.
375	Boersma, K. F., Vinken, G. C. M., & Eskes, H. J., Representativeness errors in comparing chemistry
376	transport and chemistry climate models with satellite UV–Vis tropospheric column retrievals.
377	Geoscientific Model Development, 9(2), 875-898, 2016.
270	Chaudhari, Shaatal Karda, Minal Chaudhany, Sachin Pagda, Amol P. Gadhai and Vaishali Jashi, Nitris avida
370	and cancer: a review World Journal of Surgical Opcology 2013 11:118
380	http://www.wiso.com/content/11/1/18 2013
381	http://www.wjo.com/content/11/1/10,2010.
382	Cleveland, William S., LOWESS: A program for smoothing scatterplots by robust locally weighted
383	regression. The American Statistician. <b>35</b> (1): 54. <u>JSTOR</u> <u>2683591</u> . <u>doi:10.2307/2683591</u> , 1981.
384	
385	Goldberg, D. L., Saide, P. E., Lamsal, L. N., de Foy, B., Lu, Z., Woo, JH., Kim, Y., Kim, J., Gao, M., Carmichael,
386	G., and Streets, D. G.: A top-down assessment using OMI NO2 suggests an underestimate in the NOx
387	emissions inventory in Seoul, South Korea during KORUS-AQ, Atmos. Chem. Phys. Discuss.,
388	https://doi.org/10.5194/acp-2018-678, in review, 2018.
389	
390	Herman, J., A. Cede, E. Spinei, G. Mount, M. Tzortziou, and N. Abuhassan, NO2 column amounts from
391	ground-based Pandora and MFDOAS spectrometers using the direct-sun DOAS technique:
392	Intercomparisons and application to OMI validation, J. Geophys. Res., 114, D13307,
393	doi:10.1029/2009JD011848, 2009.
394	
395	Herman, J.R., R.D. Evans, A. Cede, N.K. Abuhassan, I. Petropavlovskikh, and G. McConville, Comparison
396	of Ozone Retrievals from the Pandora Spectrometer System and Dobson Spectrophotometer in Boulder
397	Colorado, Atmos. Meas. Tech., 8, 3407–3418, 2015 doi:10.5194/amt-8-3407-2015
398	Harman L Spinoi E Fried & Kim L Kim L Kim W Code & Abubassan N and Socal Dependations
399	Herman, J., Spinel, E., Fried, A., Kim, J., Kim, J., Kim, W., Cede, A., Abunassan, N., and Segai-Rozennamer,
400	w NO <sub>2</sub> and then to measurements in Korea from 2012 to 2010 from Pandora Spectrometer Instruments
401 402	Moss Tach 11 4582 4602 https://doi.org/10.5104/amt 11.4582 2019 2019
402	wicas. 1cuii, 11, 4909-4009, IIIIps.//uui.uig/10.9194/aiiii-11-4909-2010, 2010.
<ul> <li>391</li> <li>392</li> <li>393</li> <li>394</li> <li>395</li> <li>396</li> <li>397</li> <li>398</li> <li>399</li> <li>400</li> <li>401</li> <li>402</li> <li>403</li> </ul>	<ul> <li>ground-based Pandora and MFDOAS spectrometers using the direct-sun DOAS technique: Intercomparisons and application to OMI validation, J. Geophys. Res., 114, D13307, doi:10.1029/2009JD011848, 2009.</li> <li>Herman, J.R., R.D. Evans, A. Cede, N.K. Abuhassan, I. Petropavlovskikh, and G. McConville, Comparison of Ozone Retrievals from the Pandora Spectrometer System and Dobson Spectrophotometer in Boulder Colorado, Atmos. Meas. Tech., 8, 3407–3418, 2015 doi:10.5194/amt-8-3407-2015</li> <li>Herman, J., Spinei, E., Fried, A., Kim, J., Kim, J., Kim, W., Cede, A., Abuhassan, N., and Segal-Rozenhaimer, M.: NO<sub>2</sub> and HCHO measurements in Korea from 2012 to 2016 from Pandora spectrometer instruments compared with OMI retrievals and with aircraft measurements during the KORUS-AQ campaign, Atmos. Meas. Tech., 11, 4583-4603, https://doi.org/10.5194/amt-11-4583-2018, 2018.</li> </ul>





404 405 406	lalongo, I., Herman, J., Krotkov, N., Lamsal, L., Boersma, K. F., Hovila, J., and Tamminen, J.: Comparison of OMI NO₂observations and their seasonal and weekly cycles with ground-based measurements in Helsinki, Atmos. Meas. Tech., 9, 5203-5212, https://doi.org/10.5194/amt-9-5203-2016, 2016.
407	
408 409	Kleipool, Q.L., M.R. Dobber, J.F. De Haan and P.F. Levelt, Earth Surface Reflectance Climatology from Three Years of OMI Data, Journal of Geophysical Research, 113, doi:10.1029/2008JD010290, 2008.
410	
111	Vollaging D.F. Theremann A.M. Josingvis M. Teartriev M. Devikos J.D. Durger, D. Joshos J. (2010)
411	Kolionige, D. E., Hiompson, A. W., Josipović, W., Tzortziou, W., Beukes, J. P., Burger, R., Laakso, L. (2016).
412	OMI satellite and ground-based Pandora observations and their application to surface NO2 estimations
413	at terrestrial and marine sites. Journal of Geophysical Research: Atmospheres, 123, 1441–1459.
414	https://doi.org/10.1002/2017JD026518, 2018.
415	
116	Lamcal I. N. Krotkov, N. A. Colarior, F. A. Swartz, W. H. Diakoring, K. F. Duasala, F. I. Martin, P.
410	V Dhilin S. Irio H. Codo A. Harmon I. Wainhaiman A. Strikman I. L. and Knann T. N. Evaluation
417	V., Philip, S., Irie, H., Cede, A., Herman, J., weinneimer, A., Szykman, J. J., and Knepp, T. N.: Evaluation
418	of OMI operational standard $NO_2$ column retrievals using in situ and surface-based $NO_2$ observations,
419	Atmos. Chem. Phys. Discuss., 14, 14519-14573, doi:10.5194/acpd-14-14519-2014, 2014.
420	
421	Lamsal, L. N., S. J. Janz, N. A. Krotkov, K. E. Pickering, R. J. D. Spurr, M. G. Kowalewski, C. P. Loughner, J. H.
422	Crawford, W. H. Swartz, and J. R. Herman, High-resolution NO <sub>2</sub> observations from the Airborne Compact
423	Atmospheric Mapper Retrieval and validation. J. Geophys. Res. Atmos., 122, 1953–1970.
424	doi:10.1002/2016/D025483_2017
425	001.10.1002/2010/0025405, 2017
425	Levelt D. E. Man den Oand C. H. L. Dekken M. D. Malldi, A. Missen H. de Miser, J. Gennesser, D. Lundell
426	Levelt, P. F., Van den Oord, G. H. J., Dobber, M. R., Malkki, A., Visser, H., de Vries, J., Stammes, P., Lundell,
427	J. O. V., and Saari, H.: The Ozone Monitoring Instrument, IEEE T. Geosci. Remote, 44, 1093–1101,
428	doi:10.1109/tgrs.2006.872333, 2006.
429	
430	Lindenmaier, Rodica, Manvendra K. Dubey, Bradley G. Henderson, Zachary T. Butterfield, Jay R. Herman,
431	Thom Rahn · SH. Lee, Multiscale observations of CO2, (CO2)-C-13, and pollutants at Four Corners for
432	emission verification and attribution, Proceedings of the National Academy of Sciences 06/2014;
433	111(23):8386-8391. DOI:10.1073/pnas.132188311, 2014
434	
435	Liu, F., Beirle, S., Zhang, Q., Dörner, S., He, K., and Wagner, T.: NO <sub>x</sub> lifetimes and emissions of cities and
436	power plants in polluted background estimated by satellite observations, Atmos. Chem. Phys., 16, 5283-
437	5298, https://doi.org/10.5194/acp-16-5283-2016, 2016.
438	
439	O'Byrne, G., R. V. Martin, A. van Donkelaar, J. Joiner, and E. A. Celarier, Surface reflectivity from the Ozone
440	Monitoring Instrument using the Moderate Resolution Imaging Spectroradiometer to eliminate clouds:
441	Effects of snow on ultraviolet and visible trace gas retrievals, J. Geophys. Res., VOL. 115, D17305,
442	doi:10.1029/2009JD013079, 2010.
443	
444	Marchenko, S., N. A. Krotkov, L. N. Lamsal, E. A. Celarier. W. H. Swartz. and E. I. Bucsela. Revising the
445	slant column densityretrieval of nitrogen dioxide observed by the Ozone Monitoring Instrument, J.
446	Geophys. Res. Atmos., 120, 5670–5692, doi:10.1002/2014JD022913, 2015
447	





- Raponi, Marcelo, Cede, A, Santana Diaz, Daniel, Sanchez, R, A. Otero, L, O. Salvador, J, R. Ristori, P, Quel,
  Eduardo. Total Column Ozone Measured In Buenos Aires Between March And November 2017, Using A
- 450 Pandora Spectrometer System. Anales AFA. 29(2), 46-50. 10.31527/analesafa.2018.29.2.46, 2018.
  451
- 452 Torres, O., Bhartia, P. K., Jethva, H., and Ahn, C.: Impact of the ozone monitoring instrument row
- anomaly on the long-term record of aerosol products, Atmos. Meas. Tech., 11, 2701-2715,
- 454 https://doi.org/10.5194/amt-11-2701-2018, 2018.
- 455
- 456 Tzortziou, M.; Parker, O.; Lamb, B.; Herman, J.R.; Lamsal, L.; Stauffer, R.; Abuhassan, N. Atmospheric
- 457 Trace Gas (NO<sub>2</sub> and O<sub>3</sub>) Variability in South Korean Coastal Waters, and Implications for Remote
- 458 Sensing of Coastal Ocean Color Dynamics. *Remote Sens.*, 10, 1587, ; doi:10.3390/rs10101587, 2018.
- 459

460 Vasilkov, A., Qin, W., Krotkov, N., Lamsal, L., Spurr, R., Haffner, D., Joiner, J., Yang, E.-S., and Marchenko,

- 461 S.: Accounting for the effects of surface BRDF on satellite cloud and trace-gas retrievals: a new approach 462 based on geometry-dependent Lambertian equivalent reflectivity applied to OMI algorithms, Atmos.
- 463 Meas. Tech., 10, 333-349, https://doi.org/10.5194/amt-10-333-2017, 2017.
- 464

Zara, M., Boersma, K. F., De Smedt, I., Richter, A., Peters, E., van Geffen, J. H. G. M., Beirle, S.,
Wagner, T., Van Roozendael, M., Marchenko, S., Lamsal, L. N., and Eskes, H. J.: Improved slant column
density retrieval of nitrogen dioxide and formaldehyde for OMI and GOME-2A from QA4ECV:
intercomparison, uncertainty characterization, and trends, Atmos. Meas. Tech., 11, 4033-4058,
https://doi.org/10.5194/amt-11-4033-2018, 2018.





- 471 Figure Captions
- 472 Fig 1 Diurnal variation of TCNO<sub>2</sub> measured at Pusan University in Busan South Korea
- 473 Fig. 2. Monthly average values of TCNO<sub>2</sub> for OMI and PANDORA at OMI overpass times
- 474 Fig. 3 Extended time series for Busan. Left Panel: individual matching PANDORA and OMI data
- 475 points for the overpass time ± 6 minutes. Right Panel: monthly averages.
- 476 Fig. 4. PANDORA compared to OMI. Extended TCNO<sub>2</sub> overpass time series for Mauna Loa
- 477 Observatory, Hawaii, NASA Headquarters, Washington DC, and Waterflow, New Mexico.
- Fig. 5. PANDORA compared to OMI. Extended TCNO<sub>2</sub> overpass time series for Seoul South Korea,
  Boulder, Colorado, and Buenos Aires, Argentina (Raponi et al. 2018).
- 480 Fig. 6 Percent differences between OMI and PANDORA. The slopes are the absolute change in the
- 481 percent difference. For example, the Boulder percent difference goes from -31% to -23% over 4 years.
- 482 Fig. 7A TCNO<sub>2</sub> diurnal variation (DU) from January to June, NASA Headquarters Washington, DC
- 483 from January 2015 to June 2015. The approximate OMI overpass time near 13:30 hours is marked.
- Fig. 7B TCNO<sub>2</sub> diurnal variation (DU) from July to December, NASA Headquarters Washington, DC from
  July 2015 to December 2015. The approximate OMI overpass time near 13:30 hours is marked
- Fig. 8A TCNO<sub>2</sub> diurnal variation (DU) at CCNY in New York City January to June 2018. The approximate
  OMI overpass time near 13:30 hours is marked.
- Fig. 8B TCNO<sub>2</sub> diurnal variation at CCNY in New York City July to December 2018. The peak near 5 DU
  occurs on 13 July 2018 between 11:20 and 12:30 EST. The approximate OMI overpass time near 13:30
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- 491 Fig. 9 TCNO<sub>2</sub> overpass time series for CCNY in Manhattan, New York City. Panel A: OMI overpass
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- 493 data. Panel C: Percent difference 100(OMI PAN)/PAN calculated from the data in Panel A
- 494 Fig. A1 Monthly average values of TCO for OMI and PANDORA at OMI overpass times for Busan South495 Korea







Fig 1 Diurnal variation of TCNO<sub>2</sub> measured at Pusan University in Busan South Korea



FIGURE 2

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499 **FIGURE 1** 

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Fig. 3 Extended time series for Busan. Left Panel: individual matching PANDORA and OMI data points for the overpass time ± 6 minutes. Right Panel: monthly averages.

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### 504 **FIGURE 3**







Fig. 4. PANDORA compared to OMI. Extended TCNO<sub>2</sub> overpass time series for Mauna Loa Observatory, Hawaii, NASA Headquarters, Washington DC, and Waterflow, New Mexico.







Fig. 5. PANDORA compared to OMI. Extended TCNO<sub>2</sub> overpass time series for Seoul South Korea, Boulder, Colorado, and Buenos Aires, Argentina (Raponi et al. 2017).

507 FIGURE 5







Fig. 6 Percent differences between OMI and PANDORA. The slopes are the absolute change in the percent difference. For example, the Boulder percent difference goes from -31% to -23% over 4 years.

## **FIGURE 6**





512



Fig. 7A TCNO<sub>2</sub> diurnal variation (DU) from January to June, NASA Headquarters Washington, DC from January 2015 to June 2015. The approximate OMI overpass time near 13:30 hours is marked

513

**FIGURE 7A** 







Fig. 7B TCNO<sub>2</sub> diurnal variation (DU) from July to December, NASA Headquarters Washington, DC from July 2015 to December 2015. The approximate OMI overpass time near 13:30 hours is marked

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### 515 **FIGURE 7B**







Fig. 8A TCNO<sub>2</sub> diurnal variation (DU) at CCNY in New York City January to June 2018. The approximate OMI overpass time near 13:30 hours is marked

517

## 518 Figure 8A







Fig. 8B TCNO<sub>2</sub> diurnal variation (DU) at CCNY in New York City July to December 2018. The peak near 5 DU occurs on 13 July 2018 between 11:20 and 12:30 EST. The approximate OMI overpass time near 13:30 hours is marked.

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520 Figure 8B







Fig. 9 TCNO<sub>2</sub> overpass time series for CCNY in Manhattan, New York City. Panel A: OMI overpass TCNO<sub>2</sub> (Black) compare with OMI (Red). Panel B: Monthly Lowess(0.08) fit to the daily overpass data. Panel C: Percent difference 100(OMI – PAN)/PAN calculated from the data in Panel A

521

522 Figure 9







Fig. A1  $\,$  Monthly average values of  $\mathsf{TCO}_3$  for OMI and PANDORA at OMI overpass times for Busan South Korea

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524

525 FIGURE A1