

The Dawn of Geostationary Air Quality Monitoring: Case Studies from Seoul and Los Angeles

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1 Abstract:

2 With the near-future launch of geostationary pollution monitoring satellite instruments over
3 North America, East Asia, and Europe, the air quality community is preparing for an integrated
4 global atmospheric composition observing system at unprecedented spatial and temporal
5 resolutions. One of the ways that NASA has supported this community preparation is through
6 demonstration of future space-borne capabilities using the Geostationary Trace gas and Aerosol
7 Sensor Optimization (GeoTASO) airborne instrument. This paper integrates repeated high-
8 resolution maps from GeoTASO, ground-based Pandora spectrometers, and low Earth orbit
9 measurements from the Ozone Mapping and Profiler Suite (OMPS), for case studies over two
10 metropolitan areas: Seoul, South Korea on June 9th, 2016 and Los Angeles, California on June
11 27th, 2017. This dataset provides a unique opportunity to illustrate how geostationary air quality
12 monitoring platforms and ground-based remote sensing networks will close the current
13 spatiotemporal observation gap. GeoTASO observes large differences in diurnal behavior
14 between these urban areas, with NO₂ accumulating within the Seoul Metropolitan Area through
15 the day but NO₂ peaking in the morning and decreasing throughout the afternoon in the Los
16 Angeles Basin. In both areas, the earliest morning maps exhibit spatial patterns similar to
17 emission source areas (e.g., urbanized valleys, roadways, major airports). These spatial patterns
18 change later in the day due to boundary layer dynamics, horizontal transport, and chemistry. The
19 nominal resolution of GeoTASO is finer than will be obtained from geostationary platforms, but
20 when NO₂ data over Los Angeles are up-scaled to the expected resolution of TEMPO, spatial
21 features discussed are conserved. Pandora instruments installed in both metropolitan areas
22 capture the diurnal patterns observed by GeoTASO, continuously and over longer time periods,
23 and will play a critical role in validation of the next generation of satellite measurement.. These
24 case studies demonstrate that different regions can have diverse diurnal patterns and that day-to-
25 day variability due to meteorology or anthropogenic patterns such as weekday/weekend

26 variations in emissions is large. Low Earth orbit measurements, despite their inability to capture
27 the diurnal patterns at fine spatial resolution, will be essential for intercalibrating the
28 geostationary radiances and cross-validating the geostationary retrievals in an integrated global
29 observing system.

30
31 **Key Words:** NO₂, atmospheric composition, Pandora, GeoTASO, OMPS, air quality, satellite,
32 geostationary

34 **1 Introduction**

35 The atmospheric chemistry community has long held a vision for an integrated observing system
36 that provides continuous long-term information at the spatial and temporal resolutions adequate
37 for monitoring air quality at local, regional, and global scales. This vision was first coherently
38 expressed in the Integrated Global Observing Strategy (IGOS) Atmospheric Chemistry Theme
39 Report over a decade ago (IGACO, 2004). While this vision is broadly similar to what has been
40 accomplished in the global meteorological community, its implementation for atmospheric
41 composition is still in its infancy. Satellite observations are an essential component, providing
42 continuous coverage over large areas globally. Observation requirements relevant to air quality
43 from satellites include temporal sampling at approximately one-hour frequency and horizontal
44 resolution on the order of 10 km (IGACO, 2004; Fishman et al., 2012). These temporal and
45 spatial requirements can be met globally by using a constellation approach that combines
46 multiple geostationary Earth orbit (GEO) platforms, which provide frequent observations over
47 portions of the globe, with low Earth orbit (LEO) platforms, which provide global once-daily
48 coverage (CEOS, 2011). Such a constellation strategy has been used for operational
49 meteorological observations for decades.

50 Measurements of ultraviolet-visible (UV-VIS) radiation needed to perform atmospheric
51 chemistry retrievals of ozone and its precursors have been made from platforms in LEO for the
52 past 22 years beginning with the launch of the Global Ozone Monitoring Experiment (GOME) in
53 1996 (Burrows et al., 1998), and continuing with the launch of the Ozone Monitoring Instrument
54 (OMI) in 2004 (Levelt et al, 2006), SCanning Imaging Absorption SpectroMeter for
55 Atmospheric CHartographY (SCIAMACHY) in 2002 (Bovensmann et al.,1999), GOME-2 in
56 2006 and 2013 (Callies et al., 2000), the Ozone Mapping and Profiler Suite Nadir Mapper
57 (OMPS NM) in 2011 and 2017 (Flynn et al, 2004, Yang et al., 2014), and the TROPOspheric
58 Monitoring Instrument (TROPOMI) in 2017. These data have been useful for understanding
59 global (e.g., Martin et al., 2003, Richter et al., 2005; Jaegle et al., 2005), regional (e.g., Duncan
60 et al., 2014; Duncan et al., 2016; Travis et al., 2016) and local air quality (e.g., Valin et al., 2013;
61 Zhu et al., 2017) over daily (e.g., Beirle et al., 2003; Valin et al., 2014; de Foy et al., 2016),
62 seasonal (e.g., Jaegle et al. 2005, Russell et al., 2010), interannual, and decadal time periods (van
63 der A et al., 2008; de Smedt et al. 2015). However, the relatively coarse spatial resolutions and
64 single daily observation times have substantially limited these applications, particularly within
65 the air quality management community which needs to be able to distinguish temporal profiles of
66 emissions from different source sectors and identify specific physical processes to justify
67 regulatory decisions.

68
69 Three GEO air quality missions are planned to be launched in the 2019-2023 period: Korea's
70 Geostationary Environmental Monitoring Spectrometer (GEMS) observing East Asia (Kim et al.,
71 2017), the United States' Tropospheric Emissions: Monitoring of Pollution (TEMPO) observing

72 North America (Zoogman et al., 2017), and Europe's Sentinel-4 observing Europe (Ingmann et
73 al., 2012), placing us on the cusp of a revolution in time-resolved air quality observations from
74 space. Similar to LEO instruments, these missions each consist of imaging spectrometers
75 measuring scattered light from the Earth's atmosphere in the UV-VIS wavelength range. Using
76 molecular absorption features within this range, the column-integrated atmospheric abundances
77 of certain trace gases and aerosols can be accurately retrieved. Target species relevant for air
78 quality include ozone (O₃), nitrogen dioxide (NO₂), formaldehyde (HCHO), and sulfur dioxide
79 (SO₂), as well as aerosol optical depth. Figure 1 shows the planned viewing regions for each
80 GEO mission overlaid on an image of the June 2016-2017 average OMPS NM NO₂ column
81 product (Yang et al., 2014). Unlike the single daily overpass and coarse footprint of legacy LEO
82 missions (e.g., OMPS NM, 50 km × 50 km, 13:30 LST), each GEO instrument will be capable of
83 scanning its field of regard every hour at spatial resolutions of better than 10 km. Recently
84 launched LEO instruments which are currently in check-out phase, TROPOMI and OMPS NM
85 aboard NOAA-20, have footprints of 3.5 km × 7 km and 17 km × 17 km at nadir, respectively
86 (van Geffen et al., 2017; L. Flynn, personal communication), providing global measurements
87 with sufficient spatial detail for cross-validating the three non-overlapping GEO components of
88 the constellation.

89
90 LEO measurements play a critical role in the global atmospheric composition constellation by
91 providing a means of intercalibrating and cross validating the GEO sensors and by providing
92 observations outside the fields of regard of the GEO sensors, as shown by Figure 1. The
93 importance of the LEO component of the Global Observing System for intercalibration of LEO
94 and GEO radiances has been recognized by the WMO-sponsored Global Space-based
95 Intercalibration System (GSICS, <http://gsics.wmo.int/>), which is responsible for operational
96 intercalibration of satellite instruments. To expand capability beyond existing activities for
97 sensors using visible and infrared wavelengths, GSICS has initiated a UV Subgroup that focuses
98 on cross-calibration of ultraviolet sensors, including existing LEO and future GEO instruments.
99 Harmonizing atmospheric composition retrievals among LEO and GEO sensors is also necessary
100 for effective utilization of the LEO and GEO measurements.

101
102 The spatial (< 10 km) and temporal (hourly) requirements for air quality satellites have largely
103 been determined by the desire to resolve the processes affecting the emissions, lifetime and
104 transport of tropospheric NO₂ (e.g., Beirle et al., 2011; Valin et al., 2011a; 2013; de Foy et al.,
105 2015) because of its fundamental role in the formation of tropospheric ozone and particulate
106 matter. There have been a variety of approaches for validating NO₂ products retrieved from LEO
107 platforms (e.g., Boersma et al., 2008; Bucsela et al., 2008, 2013; Irie et al., 2008; Lamsal et al.,
108 2010; Russell et al., 2011; Travis et al., 2016). These works have identified and addressed gaps
109 in the understanding of NO₂ retrievals, including methods for subtracting stratospheric NO₂
110 column contributions, a priori vertical profile deviations between urban and rural settings, and
111 surface reflectance variations (e.g., Zhou et al., 2010; Russell et al., 2011). The additional
112 retrieval assumptions relevant to GEO observations, for example changes in the a priori vertical
113 profile from morning to afternoon under different solar angles or downwind of a large point
114 source such as a power plant, are only beginning to be assessed.

115
116 To begin addressing the spatial and temporal challenges associated with GEO measurements
117 prior to launch, NASA funded the development of the suborbital Geostationary Trace gas and

118 Aerosol Sensor Optimization instrument (GeoTASO, Leitch et al., 2014, Nowlan et al., 2016)
119 and has deployed it during recent field experiments that also included networks of ground-based
120 UV-VIS solar spectrometers (Pandora, Herman et al., 2009; 2015). Analogous to how the LEO
121 observations are a transfer standard between the GEO domains, the airborne observations are a
122 transfer standard between the spatial scales of the surface-based validation instruments (i.e.
123 Pandora) and satellite observations. Here we use GeoTASO and Pandora datasets collected as
124 part of the KORUS-AQ study in Seoul, South Korea during spring 2016 and as part of the NASA
125 Student Airborne Research Program in Los Angeles, California, USA in summer 2017 to
126 demonstrate the spatial and temporal richness of the in anticipation of what will be routinely
127 provided in the near-future GEO-based measurements. We frame this discussion in the context of
128 LEO-based OMPS NM NO₂ column measurements to highlight both the spatial and temporal
129 limitations of past datasets but also to demonstrate how LEO-platforms will continue to provide
130 important global context to GEO-based sensors. Although field campaigns cover limited areas
131 and time periods, these measurements are providing a first taste of the air quality observations
132 that will be provided by scheduled GEO missions at an hourly timescale.

133 **2 Data**

134 **2.1 GeoTASO**

135 GeoTASO is an aircraft-based UV-VIS hyperspectral imaging spectrometer built by Ball
136 Aerospace (Leitch et al., 2014). It is being used to test air quality remote sensing retrievals for
137 the future GEO observations from TEMPO, GEMS, and Sentinel-4. The instrument was first
138 deployed during the NASA DISCOVER-AQ study in Houston, Texas in September 2013
139 (Nowlan et al., 2016). The data presented here were obtained by operating GeoTASO on the
140 NASA LaRC's UC-12B aircraft at a nominal altitude of 8.5 km. GeoTASO has two 2-
141 dimensional CCD detectors, gathering spectral data in the visible (VIS) wavelengths (410-690
142 nm) and in the UV wavelengths (300-380 nm). NO₂ retrievals only use data from the VIS
143 detector, which records spectra in one dimension (1056 pixels) and cross-track spatial data in the
144 second dimension (1033 pixels). The spectral integration time is fixed at 250 ms while traveling
145 at ground speeds of approximately 100 m/s. GeoTASO's nadir cross-track field of view is 45°
146 providing approximately 7 km of cross-track coverage at altitude. Prior to the NO₂ retrieval,
147 spectra are binned spatially to approximately 250 m x 250 m to increase the signal-to-noise ratio.
148 Gapless maps were created to simulate GEO observations by flying a series of parallel flight
149 lines spaced such that there was a small overlap between the adjacent swaths, taking into
150 consideration GeoTASO's 45° field of view and nominal flight altitude. Flight plans were
151 developed to cover areas of 4,000-8,000 km² so that as many as four repeat measurements could
152 be captured each day. A single traverse of this pattern across an area is referred to as a raster
153 pattern.

154
155 Spectra from 435-460nm, blue-visible light, are used to retrieve NO₂ differential slant columns
156 (DSCs) via Differential Optical Absorption Spectroscopy (DOAS). An open-source software
157 developed at the Royal Belgian Institute for Space Aeronomy called QDOAS (Danckaert et al.,
158 2016) is used to compute DSCs relative to an unpolluted reference spectrum taken in flight. The
159 resulting DSC retrievals represent the total amount of NO₂ molecular absorption along the slant
160 path of the light relative to what was present in the unpolluted reference measurement. For this
161 study, the native resolution (250 m x 250 m) DSCs are averaged to a spatial resolution of 750 m
162 x 750 m by co-adding three adjacent along-track and three adjacent across-track pixels, which is
163 still finer than any proposed GEO or LEO satellite. This averaging decreases the average DSC

164 error from 1.6×10^{15} molecules cm^{-2} to approximately 5×10^{14} molecules cm^{-2} and decreases the
165 noise observed over the area of the reference spectrum (the zero baseline for these
166 measurements) by over 50%.

167
168 DSCs are typically converted to vertical columns using a calculated air mass factor (AMF)
169 (Palmer et al., 2001; Lamsal et al., 2017). For a non-scattering atmosphere, the AMF simply
170 reflects a geometric correction of the slant path of light relative to a vertical path through the
171 atmosphere. However, because light traveling through Earth's atmosphere is heavily influenced
172 by scattering, AMF calculations require a radiative transfer model that incorporates a priori
173 assumptions about the vertical distribution of relevant trace gases (NO_2), surface albedo,
174 pressure, and aerosols, in addition to solar and viewing geometry. Ideally, the ancillary
175 information used to calculate AMFs should be at a spatial resolution similar to or better than the
176 DSC measurements to avoid introducing biases and artifacts (Russell et al., 2011). Many datasets
177 necessary for the AMF calculations do not yet exist at the sub-kilometer spatial scales at which
178 we are retrieving NO_2 and biases due to coarse a priori assumptions have not yet been evaluated
179 at this sub-kilometer spatial scale. For the early results shown in this study, fine spatial resolution
180 AMFs have not yet been calculated, therefore DSC values are shown to avoid potentially
181 introducing spatiotemporal artifacts associated with coarse AMF calculation. This simplification
182 does not fundamentally alter the conclusions of this study, as the variability of previously
183 calculated AMFs for GeoTASO (Nowlan et al., 2016) is much smaller than the spatiotemporal
184 patterns observed in this study.

185
186 The stratospheric contribution of NO_2 to the total column is small ($\sim 3 \times 10^{15}$ molecules cm^{-2}) and
187 spatially uniform relative to the tropospheric DSCs observed over Los Angeles and Seoul. The
188 temporal variation in this contribution is also small ($\sim 1 \times 10^{14}$ molecules $\text{cm}^{-2} \text{h}^{-1}$ (Sussmann et al.,
189 2005). When retrieving DSCs from GeoTASO, the contribution of stratospheric NO_2 is observed
190 similarly in the clean reference spectrum measurement as in all measurements, and thus is
191 implicitly subtracted in the fitting procedure. However, time difference between the reference
192 and retrieved observation introduces a bias in the DSCs due to the changing solar geometry
193 altering the path length of the solar beam through the stratospheric NO_2 layer. This time-
194 dependent bias in the stratospheric NO_2 is estimated and a correction is applied to results shown
195 here using the solar geometry and the stratospheric NO_2 vertical column observed from OMPS
196 NM aboard Suomi NPP (Yang et al., 2014) on the day of observation over the region of the
197 flight.

198
199 The urban areas in this study were mapped 3 to 4 times throughout one day to simulate how the
200 magnitude and spatial distribution varies diurnally at unprecedented spatial resolutions for each
201 location. In Korea, GeoTASO data were analyzed between longitudes of 126.4°E and 127.4°E
202 and latitudes of 37.2°N and 37.7°N to exclude areas outside of the Seoul Metropolitan Area
203 (SMA). Similarly, data over the Los Angeles (LA) Basin were analyzed between longitudes of -
204 118.5°W and -117.4°W and latitudes of 33.7°N and 34.165°N to restrict data from outside the
205 Basin.

206 207 **2.2 Pandora spectrometer**

208 In an effort to provide cost-effective methods for validating space-based UV-VIS trace gas
209 measurements, including those from GEO, NASA and ESA are collaborating on a global

210 network of ground-based Pandora Solar and sky-scanning spectrometers developed at NASA
211 GSFC (Herman et al., 2009). Pandora spectrometers are capable of retrieving accurate and
212 precise vertical columns of NO₂ using a direct-sun DOAS technique (Herman et al., 2009).
213 Pandora instruments are operated continuously to retrieve an NO₂ column approximately every
214 90 seconds during daylight hours, whenever the path between the surface and the sun is cloud-
215 free. These measurements are total NO₂ column with no differentiation of stratospheric or
216 tropospheric NO₂ contributions, but as discussed in section 2.1, stratospheric contributions are
217 relatively small and not variable over LA and Seoul. Data from these instruments have been used
218 to assess space- and aircraft- based retrievals of NO₂ columns (Flynn et al., 2014; Nowlan et al.,
219 2016; Goldberg et al., 2017), as well as to study the spatiotemporal variability of trace gases in
220 urban environments (Tzortziou et al., 2015) and column-to-surface relationships and their
221 relation to boundary layer depth (Flynn et al., 2014; Knepp et al., 2013). Further understanding
222 the effects of boundary layer depth on air quality has been identified as a ‘most important’
223 objective by the National Academy of Sciences’ most recent Decadal Survey (2017-2027)
224 (National Academies of Sciences, Engineering, and Medicine, 2018).

225
226 This study shows NO₂ data from three Pandoras in the SMA from May 5th-June 15th, 2016
227 (Yonsei, Olympic Park and Mount Taehwa) and six Pandoras within the LA Basin from June
228 15th-July 15th 2017 (UCLA, LA Main Street, Pico Rivera, CalTech, Fontana, and Ontario). For
229 each site, one-hour averages are calculated for analysis after the data are filtered according to
230 recommended data quality criteria (vertical column error of less than 2.69×10^{14} molecules cm⁻²
231 and normalized RMS less than 0.005). Each hourly average requires at least 5 valid observations
232 within the hour. Longer term diurnal averages (total, weekend, weekday) also calculated for
233 analysis require over 40 valid observations per hour.

234

235 **2.3 Ozone Mapping and Profiler Suite Nadir Mapper (OMPS NM)**

236 Data from the OMPS NM hyperspectral UV instrument aboard Suomi-NPP are used to
237 demonstrate legacy LEO measurement capability and to illustrate plans for incorporation of
238 recently launched (TROPOMI) and future LEO missions into the air quality observing
239 constellation. While OMI data have higher spatial resolution than OMPS NM, OMI was not
240 operational during part of the time period of this study. OMPS NM instruments are aboard
241 Suomi-NPP launched in 2011 and JPSS-1 (now NOAA-20) launched in 2017. NO₂ is retrieved
242 using an iterative spectral fitting algorithm at a nadir resolution of 50 km x 50 km (2500 km²)
243 (Yang et al., 2014), which will be further improved to 17 km x 17 km (289 km²) for OMPS NM
244 aboard NOAA-20 (L. Flynn, personal communication). OMPS NM NO₂ columns are also
245 separated into their tropospheric and stratospheric components. The measurement precision of
246 tropospheric NO₂ vertical column is estimated to be 3×10^{14} molecules cm⁻² (Yang et al., 2014).
247 For this analysis, both tropospheric and stratospheric columns are used from the instrument
248 aboard Suomi-NPP, with the latter helping correct the offset in GeoTASO’s DSCs due to the
249 stratospheric NO₂ layer as described in section 2.1. Data from OMPS NM are filtered for cloud
250 fractions greater than 25%.

251

252 **3 Results and discussion**

253 To demonstrate the capability and limitations of currently available data, Figure 2 shows single-
254 overpass and monthly-averaged OMPS NM NO₂ column measurements over South Korea and
255 California. On June 9, 2016, the OMPS NM nadir overpass was to the west of the Seoul

256 Metropolitan Area (SMA). Because of the viewing geometry and the curvature of the Earth, the
257 OMPS NM off-nadir detector elements that view the SMA cover twice as much surface area as
258 those at nadir (nominally 50 km × 50 km) on this day (Figure 2a). On the other hand, the OMPS
259 NM nadir overpass was directly over Southern California on June 27th, 2017 (Figure 2b), such
260 that OMPS NM was able to measure the tropospheric NO₂ column over LA near its finest spatial
261 resolution.

262
263 LEO observations can be refined spatially by ‘oversampling’ over a longer temporal range, as
264 the orbital track varies day-to-day leading to variable spatial sampling (e.g., the edge of swath
265 over Korea on June 9th, 2016 vs. the nadir observations over Los Angeles from June 27th, 2017).
266 This technique has been applied to trace-gas retrievals from imaging spectrometers, like OMI,
267 for NO₂, HCHO, and SO₂ data to identify and investigate pollution emitting sources, their
268 average plume extent, and emission rates (deFoy et al., 2009; Russell et al., 2010; McLinden et
269 al. 2012; Zhu et al., 2014). Figure 2c and d show the 0.25° x 0.25° (approximately 20 km x 30
270 km at 35°N) monthly average created by oversampling OMPS NM NO₂ data for June 2016 over
271 South Korea (Figure 2c) and June 2017 over California (Figure 2d). Here, the OMPS NM
272 average measurements show that NO₂ columns are locally maximum over Seoul and Los
273 Angeles. By providing the means to distinguish sources, long-term trends can be used to evaluate
274 the changes of emissions driven by regulatory programs (Kim et al., 2006), technological
275 controls (e.g., Russell et al., 2012) and economic activity (e.g., Russell et al., 2012; de Foy et al.,
276 2016; Duncan et al., 2016). Whether considering daily measurements or analysis of long term
277 monthly averages, instruments like OMPS NM provide a well-characterized, quantitatively
278 stable measurement reflecting a balance of NO₂ emissions and removal at spatial scales of ~25
279 km, with some limited information on pollutant transport (e.g., Beirle et al., 2011; Valin et al.,
280 2013; 2014; de Foy et al., 2016). As such, the measurements available from the past have not
281 been sufficient to address the more pressing air quality management needs: the ability to
282 distinguish sources within urban airsheds, characterization of local mesoscale flow patterns on
283 pollutant transport, quantification of NO₂ removal mechanisms (e.g., Valin et al., 2013), or better
284 characterization of photochemical ozone production to NO_x (NO + NO₂) or VOC control
285 strategies (e.g., Martin et al., 2004; Duncan et al., 2010; Jin et al., 2017; Schroeder et al. 2017) .

286
287 The LEO-based data in Figures 1 and 2 represent the standard measurement that has been
288 available to observe pollutants globally from space-based platforms for more than two decades.
289 While finer scale global LEO data will soon be available with the addition of TROPOMI and
290 NOAA-20 OMPS NM, the following two case studies demonstrate the information that will be
291 gained in adding temporally resolved GEO observations to this global observing system by
292 focusing on GeoTASO and Pandora measurements within the Seoul Metropolitan Area (SMA)
293 and the Los Angeles (LA) Basin in June 2016 and 2017, respectively. Figure 3 shows maps of
294 each metropolitan area discussed in these case studies. The white polygons encompass the area
295 observed by GeoTASO, white stars and labels are Pandora locations, red/blue lines are major
296 roadways (SEDAC, 2013), and icons and regions labeled in yellow are discussed in Section 3.1
297 and 3.2. Areas of elevated terrain appear are darker than the surrounding valleys and are
298 typically free of strong emission sources. Densely urbanized areas within the valleys appear
299 greyer in color. Using this map as a reference will help guide the discussion below.

300

3.1 Case study 1: Seoul Metropolitan Area, South Korea

The first execution of diurnal mapping over an urban area with GeoTASO was during the DISCOVER-AQ Front Range field study in summer 2014 (Crawford, et al., 2016). This same strategy was used more extensively during the KORUS-AQ field study in spring 2016 (<https://www-air.larc.nasa.gov/missions/korus-aq/>). Figure 4 shows maps of NO₂ DSC obtained by GeoTASO on June 9, 2016, at 4 different times of day between 08:00-18:00 LT over the Seoul Metropolitan Area (SMA). Each of the 4 rasters covers an area of approximately 40 km x 70 km in approximately two hours. This is also the approximate area of a single nadir OMPS NM pixel (Figure 2). Overlaid in panels a and c in Figure 4 are wind vectors averaged over the lowest 500 m agl from the full spectral resolution (~13-km) Global Data Assimilation System (GDAS) analyses for 00:00 UTC (09:00 LT) and 06:00 UTC (15:00 LT), respectively (Kleist and Ide, 2015a, 2015b). Output, archived at standard 6-hour intervals, is not available during the other two rasters.

Figure 5 shows percentile distributions of NO₂ DSCs for each SMA raster shown in Figure 4. Over the SMA area on this day, NO₂ pollution is at its minimum in the morning then increases and becomes more variable throughout the day, demonstrating the accumulation of NO₂ at a rate faster than its removal. The area median more than doubles from 20×10^{15} molecules cm⁻² to 49×10^{15} molecules cm⁻² over the course of the day with the interquartile range (representing the variability) expanding as well. There is not a significant change in the median from late morning to mid-afternoon, however the distribution is skewed upwards with the 75th percentile reaching 58×10^{15} molecules cm⁻² for Raster 3. Raster 4 exhibits the largest magnitude and variability of NO₂ columns on June 9th with the median DSCs approaching 50×10^{15} molecules cm⁻² and an interquartile range of 44×10^{15} molecules cm⁻². Maximum DSCs observed over the SMA during this day, up to 120×10^{15} molecules cm⁻² and well exceeding the 95th percentile, occurred during Rasters 3 and 4.

During the morning (Rasters 1 and 2), distinct patterns are apparent with maximum NO₂ DSCs over urbanized valleys and minimums located directly over elevated terrain. The western minima located south-southeast of Incheon is not due to elevated terrain, but instead due to the lack of large emission sources within this rural farmland region. The largest DSCs in the morning coincide with the areas with the largest temporal growth between Raster 1 and Raster 2, including Incheon, south central Seoul, and Suwon, where the DSCs grow to a magnitude outside of the interquartile range. These are areas with dense urbanization shown in Figure 3 and are likely the areas with the largest emissions in this domain.

The morning patterns reflect emission sources (i.e. roads and urban centers) that are confined spatially. In comparison, the spatial distribution of NO₂ DSCs in the afternoon changes dramatically. The boundary layer grows through the day due to surface heating, and from Raster 2 to Raster 3 grows deep enough to encompass the surrounding terrain. By the afternoon it appears the mixed layer is deep enough and advection is fast enough that the spatial pattern of NO₂ columns no longer reflects the pattern of emission sources. While muted in a deeper afternoon mixed layer, the terrain influence on the spatial pattern of NO₂ columns is still visible in Raster 4, where there is a local minimum in middle of the SMA plume with a magnitude of $\sim 70 \times 10^{15}$ molecules cm⁻² near Mt. Gwanaksan (green triangle in Figure 3 and 4). Over the nearby valley (5 km west), NO₂ DSC values are 20×10^{15} molecules cm⁻² larger. Assuming that

347 the mixed layer height is independent of terrain variations, the 20×10^{15} molecules cm^{-2} would
348 equate to an average mixing ratio of 20 ppbv within the 400 m between the valley floor and the
349 elevated terrain (assuming a temperature of 300 K and surface pressure of 1000hPa). This
350 mixing ratio estimate of 20 ppbv compares well with the mixing ratios measured nearby by
351 NCAR's 4-channel chemiluminescence instrument aboard the NASA DC-8 aircraft on this
352 afternoon during a KORUS-AQ flight (not shown). In such situations, local minima in column
353 amounts may not correspond to local minima in surface concentrations. GEO observations,
354 Pandora measurements, and routine air quality monitoring networks will begin to resolve some
355 of these differences in areas with complex terrain and provide translation to similar locations
356 without surface monitoring.

357
358 While NO_2 over the SMA generally accumulates throughout the day, this is not true for all
359 locations within the region. On June 9th, 2016, winds from the GDAS analyses near the SMA
360 shift from weak northerly flow in the morning (00:00 UTC – 09:00 LT) to stronger westerly flow
361 during the afternoon (06:00 UTC – 15:00 LT). The spatial pattern over the SMA does not change
362 from Raster 1 to Raster 2. However, during the afternoon there is a shift progressively to the east
363 between Rasters 3 and 4, indicative of horizontal transport. This is most apparent by observing
364 the edges of the SMA NO_2 DSC plume, such as at Incheon where there is significant growth
365 between Rasters 1 and 2, but then decay and/or extension toward the east during the afternoon
366 rasters. Additionally, it takes approximately two hours to cover the area of the domain in each
367 Rasters. In Raster 3, the spatial offsets of DSCs between successive overpasses (ranging from
368 15-30 minutes) between Incheon and south Seoul is likely caused by advection of the plume
369 between Raster line samples. On the eastern side of the domain, the Mount Taehwa area is
370 relatively unpolluted during Rasters 1 and 2, but between Raster 3 and Raster 4, NO_2 DSCs
371 increase and are consistent with what would be expected from the advection of the SMA plume
372 to southeast based on the 15:00 LT winds.

373
374 As part of efforts to demonstrate GEO validation plans, Pandora instruments provided direct sun
375 vertical column NO_2 measurements that are complementary to the GeoTASO backscatter DSCs
376 at three sites in the region (Figure 6). The selected sites cover a range of air quality conditions
377 across the SMA, spanning the domain of GeoTASO observations from the northwest (Yonsei is
378 just outside the raster domain due to airspace restrictions) to the east-southeast over Olympic
379 Park and Mount Taehwa another 40 km southeast (stars in Figures 2 and 4). Grey lines in Figure
380 6 show the hourly-averaged diurnal pattern for all days between May 5th and June 15th, 2016,
381 with the day of the GeoTASO observations, June 9th, highlighted in red. On June 9, 2016, the
382 observations at these sites are broadly consistent with the NO_2 column growth and transport
383 patterns observed by GeoTASO; NO_2 columns are large and growing over low-lying population
384 centers during the morning hours (e.g., Yonsei and Olympic Park) followed by transport to the
385 southeast, such that columns diminish over Yonsei in the early afternoon while growing over
386 Olympic Park briefly (also seen in Raster 3 from GeoTASO: Figure 4c) before finally
387 diminishing over Olympic Park and growing over Taehwa in the late afternoon. At Yonsei
388 University, Pandora measurements were made from the top of a campus building (180 m MSL,
389 ~130 m AGL). As a result, observations at Yonsei University are biased low at all times of the
390 day, especially in the morning hours, when the unsampled portion of the boundary layer (130 m)
391 is a larger component of the typically shallower NO_2 mixing depth. A similar bias has been
392 observed and quantified for previous Pandora measurements in Houston using coincident NO_2 in

393 situ measurements (Nowlan et al., 2016; Judd, 2016), however this potential bias does not
394 change the larger conclusions made here.

395
396 June 9th, 2016 was the only day that GeoTASO was used to acquire observations at 4 different
397 times throughout the day. However, the Pandora measurements show large day-to-day variations
398 of NO₂ column at these sites across the SMA (Figure 6, gray lines), particularly over the urban
399 sites of Yonsei and Olympic Park, but also over the rural Mount Taehwa site, downwind of
400 Seoul. The hourly and daily variations of NO₂ column can provide important constraints on
401 transport models, particularly when influences from local mountain and land-ocean circulations
402 are challenging to accurately simulate. The hourly values from Figure 6 are also averaged to
403 calculate the weekday diurnal average in solid black and weekend average in dashed black.
404 At all sites, the weekend column densities are lower than those during the weekdays,
405 highlighting the influence of anthropogenic activity on air quality. The longer-term averages of
406 the NO₂ column reveal important information on chemical transport, but primarily reveal
407 important lessons for understanding the chemical mass balance of NO₂ emissions and loss, or
408 insights on the importance of sources of pollutant emissions that are known to have a day-of-
409 week variation (e.g., Beirle et al., 2003; Harley et al., 2005; Valin et al., 2014).

410
411 The magnitude of NO₂ observed in the most polluted regions of the SMA by both Pandora and
412 GeoTASO is over an order of magnitude larger than observed by OMPS NM (Figure 3). The
413 LEO observations roughly coincide with the time of Raster 3 in the SMA. At Olympic Park at
414 this time, Pandora and GeoTASO both measured spikes in the local NO₂ column at the same
415 order of magnitude ($70\text{-}80 \times 10^{15}$ molecules cm⁻²) and the SMA as a whole had a median of
416 33×10^{15} molecules cm⁻². The order of magnitude discrepancy between the finer scale
417 measurements (GeoTASO and Pandora) and the coarse LEO observations (OMPS NM) reflect
418 spatial averaging of over an area that also includes less NO₂-polluted air. The SMA is observed
419 by four OMPS NM pixels, each averaging only a fraction of the enhanced NO₂ columns over the
420 SMA with a larger area of background NO₂ columns. Only moderate enhancements ($\sim 3 \times 10^{15}$
421 molecule cm⁻²) are observed over Seoul by the $\sim 20,000$ km² covered by the 4 OMPS NM pixels.
422 Looking at the oversampled monthly averaged data (Figure 2c), the spatial patterns correlate
423 better with those observed by GeoTASO observations with a peak centered over the SMA
424 region. Over this month there are fewer polluted days than the case shown on June 9th, as shown
425 by the Pandora measurements in Figure 6, resulting in a smaller magnitude of NO₂ over the
426 SMA on the month time-scale vs. the afternoon sample from Raster 3. Comparisons of OMPS
427 NM data (50 km × 50 km) with OMI data (24 km × 13 km) (Yang et al., 2014) and OMI
428 operational products with super-zoom OMI data (~ 7 km × 13 km; Valin et al., 2011b) confirm
429 that neither OMPS NM nor OMI operational footprints are sufficient to resolve the small-scale
430 NO₂ spatial variations over localized sources. Due to the nature of NO₂ emissions and its short
431 atmospheric lifetime, air quality applications require that the variability of NO₂ columns are
432 spatially resolved (e.g., Cohan et al., 2006; Valin et al., 2011a), a capability anticipated from
433 future LEO (TROPOMI: 3.5 km × 7 km) and GEO platforms.

434

435 **3.2 Case Study 2: Los Angeles, California**

436 Figure 7 shows NO₂ DSC maps obtained over the LA Basin at three different times on June 27th,
437 2017 capturing the morning, mid-day, and late afternoon periods. The left column shows the 750
438 m x 750 m resolution DSCs from GeoTASO and the right column shows a product that is co-

439 added to $3 \text{ km} \times 3 \text{ km}$ to emulate a sampling footprint that is more comparable to what is
440 anticipated from GEO. The area of this raster spans approximately $50 \text{ km} \times 50 \text{ km}$ in the
441 southern half and approximately 115 km east-to-west on the northern side of the Basin. Overlaid
442 are boundary layer averaged wind vectors from the North American Model (NAM)-CONUS 3-
443 km nest (Janjic and Gall, 2012) from 16:00 UTC (09:00 LT) on Raster 1, 20:00 UTC (13:00 LT)
444 on Raster 2, and 00:00 UTC (17:00 LT) for Raster 3. Figure 8 shows percentile distributions of
445 NO_2 DSCs for each LA Raster at the 750 m resolution (the left column of Figure 7). Unlike over
446 the SMA, maximum NO_2 columns are observed in the morning, with a median NO_2 DSC over
447 the LA Basin of $12.5 \times 10^{15} \text{ molecules cm}^{-2}$ during Raster 1. Notably, this value is lower than the
448 minimum median observed over the SMA during any of those four rasters. The median value
449 decreases approximately 50% between the morning and late afternoon (Raster 1 vs. Raster 3) in
450 the LA Basin. The opposite diurnal pattern observed by GeoTASO over Seoul and Los Angeles
451 may indicate more prevalent mid-day sources in Seoul relative to Los Angeles, differing
452 chemistry regimes, or perhaps just a difference in transport patterns during the case study
453 periods. With GEO platforms providing more data to test these hypotheses over many more
454 urban areas, we anticipate exciting opportunities for future air quality research.

455
456 Similar to the SMA example, the winds are relatively light in the morning and the spatial
457 distribution of NO_2 appears to mimic the distribution of emission sources. During Raster 1,
458 enhancements that likely reflect mobile emission sources are located over freeways (i.e. I10, I5,
459 and CA60: blue outlined roads in Figure 3) with the largest enhancements over downtown Los
460 Angeles (just west of LA Main Street) where many of these freeways intersect and traffic
461 congestion could lead to local emission enhancements. An additional maximum is observed over
462 LAX Airport on the coast (airplane icon in Figure 3), a large NO_x emission source. The lowest
463 columns measured coincide with areas of elevated terrain, such as the hills west of Long Beach,
464 and areas of the Santa Ana mountains.

465
466 On the western side of the Basin during Raster 2, GeoTASO observes a line of high NO_2 DSCs,
467 mimicking a frontal structure extending north-to-south from Glendale down to Long Beach,
468 peaking near downtown Los Angeles. At the same time, the hot spot over LAX airport during
469 Raster 1 is now more diffuse with a plume-like structure extending to the east, indicative of
470 horizontal transport inland. With Los Angeles's location on the Pacific Coast, the area is often
471 influenced by mesoscale land/water circulations (sea breezes) due to unequal heating over the
472 land and water, which could result in westerly transport of pollution within the LA Basin during
473 the daytime. Figure 9 illustrates the role of sea breeze transport on this day. Figure 9a shows
474 contoured 2-meter relative humidity (RH) and boundary layer averaged wind vectors from the
475 NAM-CONUS 3-km nest over the western half of the LA Basin at 20:00 UTC (13:00LT: the
476 midpoint of Raster 2). On this map, the largest gradient in relative humidity and shift in wind
477 vectors occurs around the 40% relative humidity contour indicating the boundary between the
478 land and marine air masses (i.e. the sea breeze front). The 40% contours from 19:00, 20:00, and
479 21:00 UTC are overlaid on the GeoTASO NO_2 DSCs from Raster 2 in Figure 9b to indicate the
480 movement of the sea breeze front during Raster 2 in relation to the NO_2 feature observed during
481 this time. These modeled results are similar in timing to the observed sea breeze arrival at the
482 South Coast Air Quality Monitoring District's LA Main Street monitoring location, which saw
483 an air mass transition at 13:30 LT with a slow increase in westerly wind speed and a 10%
484 increase in RH. The edge of the peninsula to the west of Long Beach has hilly terrain that acts as

485 a barrier to the penetrating sea breeze front, and due to the directional orientation of the coastline
486 in this area, there are two different sea breeze fronts pushing inland and converging around the
487 Long Beach area. The spatial structure of NO₂ during Raster 2 mimics the shape of the sea
488 breeze front that is pushing inland, suggesting that this front is advecting the pollution that was
489 along the coast to the east as the sea breeze progresses inland through the afternoon. In fact, it
490 appears that NO₂ is trapped within the convergence zone between the two sea breeze fronts in the
491 southern end of the Raster. The influence of air mass convergence on pollution build up has
492 been observed in other coastal regions, such as in Houston, Texas, where synoptically driven
493 offshore flow can converge with the sea breeze front allowing for the buildup of pollution within
494 its convergence zone and causing poor air quality (Banta et al., 2005). Although less defined, this
495 linear NO₂ feature within the continued presence of the convergence zone also appears in Raster
496 3 (Figure 7e) slightly further to the east, demonstrating the influence of this convergence zone
497 over the duration of the afternoon.

498
499 The appearance of enhanced NO₂ during Raster 3 between downtown Los Angeles and the
500 Inland Empire coincides with an area of enhancement also observed during the morning flight. It
501 is impossible to tell from the available data in this study whether this enhancement is due to
502 continued sea breeze transport or the result of increased local emissions during the late
503 afternoon, but as a whole, these datasets demonstrate the complexity of the spatial distribution of
504 NO₂ in a coastal urban metropolitan surrounded by complex terrain.

505
506 To provide an initial assessment of the data that will be routinely available from GEO
507 observations, the Los Angeles data are binned up to 9 km² (expected nadir areal resolution of
508 TEMPO: Zoogman et al., 2017) by averaging the data into 3 km x 3 km pixel bins (Figure 7
509 right). While the signatures are muted due to spatial averaging, the features discussed in the
510 preceding paragraphs remain spatially distinct, demonstrating how GEO observations from
511 TEMPO are expected to address salient air quality questions, even in a coastal region with
512 complex terrain, mesoscale circulations, and temporal emission patterns.

513
514 Figure 10 is the same format as Figure 6, but for the 6 Pandoras installed in the Los Angeles
515 Basin showing data between the dates of June 15th and July 15th, 2017. As is the case over
516 Seoul, Pandora NO₂ vertical column measurements in the LA Basin on June 27th, 2017 (Figure
517 10: red lines) are broadly consistent with the NO₂ column growth and transport patterns observed
518 by GeoTASO, most notably the early afternoon peak at LA Main Street coinciding with the sea
519 breeze front arrival on this day. In Los Angeles, unlike Seoul, NO₂ columns observed by
520 Pandora spectrometers are generally at a maximum in the mid-morning hours and decrease in the
521 early afternoon hours on weekdays. Over coastal and downtown Los Angeles sites (UCLA, LA
522 Main Street), NO₂ columns continue to decrease or remain steady in the late afternoon hours
523 whereas NO₂ columns grow at Pico Rivera, Ontario, and Fontana, reflecting the inland transport
524 of cleaner air at the coast and more polluted air at the downwind sites in the presence of westerly
525 prevailing winds.

526
527 While the overlying diurnal features are apparent on many days (e.g. the morning peak in NO₂ at
528 most sites), day-to-day variability in Pandora data (Figure 10: grey lines) shows significant
529 deviation from the average patterns. For example, the sea breeze front that shows a distinct
530 maximum over LA Main Street on June 27th only occurs on a handful of other days that month.

531 CalTech is also different from the other sites in that its NO₂ peak is around midday. Similar to
532 Taehwa, CalTech is not a primary NO₂ source area, but instead a potential receptor to Los
533 Angeles's early morning emissions under the right transport conditions (not seen on June 27th).
534 Additionally, CalTech is approximately 250 m asl (or about 150 m higher than the LA Main
535 Street site) and another midday contributor there may be mixing from lower-lying areas to the
536 elevation sampled by the Pandora as the mixed layer grows throughout the day. Like Seoul, NO₂
537 columns are smaller at all sites on the weekend (Saturday-Sunday) than during the week
538 (Monday-Friday), with a few sites exhibiting flat-shaped weekend temporal profiles, indicating
539 minimal change in column throughout the weekend day (CalTech, Pico Rivera). These
540 weekday-weekend differences are a fingerprint that can help identify the contribution of various
541 NO₂ sources based on our understanding of their day-of-week variation (e.g., heavy duty diesel
542 trucking, Harley et al., 2005) and important nonlinear chemical feedbacks (e.g, Valin et al.,
543 2014).
544

545 In contrast to the order of magnitude difference in NO₂ between GeoTASO and OMPS in Seoul,
546 South Korea (discussed in section 3.1), the near-nadir measurements over the LA Basin from
547 OMPS on June 27th, 2017 (Figure 2b: $2\text{-}5 \times 10^{15}$ molecules cm⁻²) were much closer to the midday
548 GeoTASO measurements during Raster 2 (median of 6.8×10^{15} molecules cm⁻²). The area covered
549 by GeoTASO is approximately 1.5 times the area of a nadir OMPS pixel, but the GeoTASO
550 raster does not encompass any single OMPS NM pixel in its entirety from this overpass in which
551 to do a one-to-one comparison. However, the oversampled image (Figure 2d) does suggest that
552 over a month-long timescale, OMPS NM observes NO₂ confined to the area measured by
553 GeoTASO within the LA Basin at the same order of magnitude as this GeoTASO case study day.
554

555 **4 Conclusions**

556 This work illustrates the spatiotemporal detail that will be resolved with the upcoming GEO air
557 quality measurements, using GeoTASO NO₂ retrievals as a proxy, and how ground-based and
558 LEO datasets will play important roles in validating and connecting these GEO observations
559 from the local- to global-scale. Data from GeoTASO, used as a testbed to address GEO
560 validation needs and to anticipate future opportunities for air quality management applications, is
561 used to resolve the spatiotemporal patterns of tropospheric column NO₂ over Seoul and Los
562 Angeles. In the morning, under the influence of weak winds, spatial patterns of NO₂ reflect the
563 spatial distribution of emission sources and topography over both the SMA and the LA Basin.
564 NO₂ column densities over the SMA grow throughout the day as emission rates outweigh NO₂
565 removal from the column, while NO₂ in Los Angeles typically peaks during the mid-morning
566 hours indicating that removal processes overtake emission rates before midday. GEO
567 observations will show whether these conclusions apply beyond the case studies shown here, as
568 well as expanding to other metropolitan areas around the globe. These spatially and temporally
569 refined measurements will begin to link the role of emissions and atmospheric dynamics with the
570 spatial distribution of pollutants in regions impacted by poor air quality, details that past LEO
571 observations were incapable of capturing.
572

573 In addition to the single days of GeoTASO data analyzed for each case study region, Pandora
574 observations are used to demonstrate day-to-day and hour-to-hour variability of NO₂ that will be
575 measured from GEO and provide a means of linking the satellite-based column measurements to
576 variations in in situ surface concentrations. Over both the SMA and the LA Basin, Pandora

577 measurements reveal that NO₂ columns vary between weekdays and weekends and between
578 source and receptor sites. They also fluctuate greatly on a day-to-day basis from the statistically
579 calculated diurnal averages, particularly near large sources. The frequent Pandora observations,
580 many times per hour, and their anticipated co-location with surface air quality and meteorology
581 monitoring instrumentation will also provide insight to transient local processes that better
582 inform the use of column-integrated measurements for monitoring surface-based pollution.
583

584 LEO observations are now attaining similar spatial resolutions as those expected from the GEO
585 instruments and are essential for intercalibrating radiances measured by each of the GEO
586 instruments as well as cross-validating their data products. Observations from decades of LEO
587 observations have provided compelling verification of multi-year changes in pollutant emissions
588 in different regions of the world. However, as illustrated by these case studies, pollutant
589 concentrations vary greatly through the day, particularly in urban areas. Variations are driven by
590 factors that also change through the day: emissions, photochemistry, and meteorology. Sparse
591 observations, including temporally sparse LEO observations (e.g. OMPS) and spatially sparse
592 surface measurements (e.g. Pandora), do not permit these factors to be disentangled, limiting
593 improvements in air quality assessment and prediction. The GeoTASO data shown in these case
594 studies illustrate one change in perspective the GEO observations will provide: moving beyond
595 coarse, static early-afternoon snapshots from LEO to dynamic visualization of chemical weather.
596 Together the pieces of this system will enable better understanding of the locations and
597 magnitudes of emissions and of meteorological influences, better monitoring of the air we
598 breathe, and ultimately more effective strategies for improving air quality.
599

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608

609 **6 Conflict of Interest**

610 The authors declare that the research was conducted in the absence of any commercial or
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612

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621
622

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626

627 **8 Author contributions**

628 LJ, JA, and BP formulated the central research idea. LJ and JA drafted the manuscript. LJ led the
629 processing and analysis of GeoTASO data. MT and MM processed the Pandora NO₂ retrievals
630 and LV led Pandora data analysis. BP provided the modeled meteorology dataset and relevant
631 processing scripts. KY provided OMPS NM data. JS was involved with Pandora measurements
632 and maintenance in South Korea. SJ, MK, and JA participated in the flight planning and data
633 gathering during GeoTASO flights in South Korea, with the addition of LJ in California. All
634 authors provided input, suggestions, and edits to the manuscript.

635

636 **9 Data availability**

- 637 • GeoTASO data can be provided upon request and will become publically available after
638 June 2018 on the KORUS-AQ data archive ([https://www-air.larc.nasa.gov/cgi-](https://www-air.larc.nasa.gov/cgi-bin/ArcView/korusaq?B200=1)
639 [bin/ArcView/korusaq?B200=1](https://www-air.larc.nasa.gov/cgi-bin/ArcView/lmos)) and the LMOS archive ([https://www-](https://www-air.larc.nasa.gov/cgi-bin/ArcView/lmos)
640 [air.larc.nasa.gov/cgi-bin/ArcView/lmos](https://www-air.larc.nasa.gov/cgi-bin/ArcView/lmos))
- 641 • Pandora data are available on data.pandonia.net
- 642 • OMPS data are available at <https://dx.doi.org/10.5067/N0XVLE2QAVR3>
- 643 • GFS and NAM meteorology are available at <http://nomads.ncep.noaa.gov/>
- 644 • SCAQMD hourly data is available <https://www.arb.ca.gov/aqmis2/metsselect.php>, and
645 higher temporal resolution is available upon request from SCAQMD.

646

647 **10 Disclaimer:**

648 The views, opinions, and findings contained in this report are those of the author(s) and should
649 not be construed as an official National Oceanic and Atmospheric Administration,
650 Environmental Protection Agency, or U.S. Government position, policy, or decision.

651

652 **References**

653 Banta, R. M., Senff, C. J., Nielsen-Gammon, J., Darby, L. S., Ryerson, T. B., Alvarez, R. J., et
654 al. (2005). A bad air day in Houston. *Bulletin of the American Meteorological Society* 86,
655 657–669.

656 Beirle, S., Platt, U., Wenig, M., and Wagner, T. (2003). Weekly cycle of NO₂ by GOME
657 measurements: A signature of anthropogenic sources. *Atmospheric Chemistry and*
658 *Physics Discussions* 3, 3451–3467. doi:10.5194/acpd-3-3451-2003.

659 Beirle, S., Boersma, K. F., Platt, U., Lawrence, M. G., and Wagner, T. (2011). Megacity
660 Emissions and Lifetimes of Nitrogen Oxides Probed from Space. *Science* 333, 1737–
661 1739. doi:10.1126/science.1207824.

662 Boersma, K. F., Eskes, H. J., and Brinksma, E. J. (2004). Error analysis for tropospheric NO₂
663 retrieval from space. *Journal of Geophysical Research: Atmospheres* 109,
664 doi:10.1029/2003JD003962.

- 665 Boersma, K. F., Jacob, D. J., Trainic, M., Rudich, Y., DeSmedt, I., Dirksen, R., et al. (2009).
666 Validation of urban NO₂ concentrations and their diurnal and seasonal variations
667 observed from the SCIAMACHY and OMI sensors using in situ surface measurements in
668 Israeli cities. *Atmospheric Chemistry and Physics* 9, 3867–3879. doi:10.5194/acp-9-
669 3867-2009.
- 670 Bovensmann, H., Burrows, J. P., Buchwitz, M., Frerick, J., Noël, S., Rozanov, V. V., et al.
671 (1999). SCIAMACHY: Mission objectives and measurement modes. *Journal of the*
672 *atmospheric sciences* 56, 127–150.
- 673 Broccardo, S., Heue, K.-P., Walter, D., Meyer, C., Kokhanovsky, A., v. d. A, R., et al. (2017).
674 Intra-pixel variability in satellite tropospheric NO₂ column densities derived from
675 simultaneous spaceborne and airborne observations. *Atmospheric Measurement*
676 *Techniques Discussions*, 1–32. doi:10.5194/amt-2016-366.
- 677 Bucsela, E. J., Perring, A. E., Cohen, R. C., Boersma, K. F., Celarier, E. A., Gleason, J. F., et al.
678 (2008). Comparison of tropospheric NO₂ from in situ aircraft measurements with near-
679 real-time and standard product data from OMI. *Journal of Geophysical Research* 113.
680 doi:10.1029/2007JD008838.
- 681 Bucsela, E. J., Krotkov, N. A., Celarier, E. A., Lamsal, L. N., Swartz, W. H., Bhartia, P. K., et al.
682 (2013). A new stratospheric and tropospheric NO₂ retrieval algorithm for nadir-viewing
683 satellite instruments: applications to OMI. *Atmos. Meas. Tech.* 6, 2607–2626.
684 doi:10.5194/amt-6-2607-2013.
- 685 Burrows, J. P., Weber, M., Buchwitz, M., Rozanov, V., Ladstätter-Weissenmayer, A., Richter,
686 A., et al. (1999). The global ozone monitoring experiment (GOME): Mission concept and
687 first scientific results. *Journal of the Atmospheric Sciences* 56, 151–175.
- 688 Callies, J., Corpaccioli, E., Eisinger, M., Hahne, A., and Lefebvre, A. (2000). GOME-2-Metop's
689 second-generation sensor for operational ozone monitoring. *ESA bulletin* 102, 28–36.
- 690 CEOS (2011). A Geostationary Satellite Constellation for Observing Global Air Quality: An
691 International Path Forward. Available at:
692 [http://ceos.org/document_management/Virtual_Constellations/ACC/Documents/AC-](http://ceos.org/document_management/Virtual_Constellations/ACC/Documents/AC-VC_Geostationary-Cx-for-Global-AQ-final_Apr2011.pdf)
693 [VC_Geostationary-Cx-for-Global-AQ-final_Apr2011.pdf](http://ceos.org/document_management/Virtual_Constellations/ACC/Documents/AC-VC_Geostationary-Cx-for-Global-AQ-final_Apr2011.pdf) [Accessed March 6, 2018].
- 694 Cohan, D. S., Hu, Y., and Russell, A. G. (2006). Dependence of ozone sensitivity analysis on
695 grid resolution. *Atmospheric Environment* 40, 126–135.
696 doi:10.1016/j.atmosenv.2005.09.031.
- 697 Crawford, J. H., Al-Saadi, J., Pierce, G., Long, R. W., Szykman, J. J., Leitch, J., et al. (2016).
698 Multi-perspective observations of NO₂ over the Denver area during DISCOVER-AQ:
699 Insights for Future Monitoring. *EM: Air and Waste Management Associations Magazine*
700 *for Environmental Managers*, 5.

- 701 Danckaert, T., Fayt, C., Van Roozendael, M., De Smedt, I., Letocart, V., Merlaud, A., et al.
702 (2017). QDOAS Software User Manual. Available at: [http://uv-](http://uv-vis.aeronomie.be/software/QDOAS/QDOAS_manual.pdf)
703 [vis.aeronomie.be/software/QDOAS/QDOAS_manual.pdf](http://uv-vis.aeronomie.be/software/QDOAS/QDOAS_manual.pdf) [Accessed March 27th, 2018].
- 704 de Foy, B., Krotkov, N. A., Bei, N., Herndon, S. C., Huey, L. G., Martínez, A.-P., et al. (2009).
705 Hit from both sides: tracking industrial and volcanic plumes in Mexico City with surface
706 measurements and OMI SO₂ retrievals during the MILAGRO field campaign.
707 *Atmospheric Chemistry and Physics* 9, 9599–9617. doi:10.5194/acp-9-9599-2009.
- 708 de Foy, B., Lu, Z., Streets, D. G., Lamsal, L. N., and Duncan, B. N. (2015). Estimates of power
709 plant NO_x emissions and lifetimes from OMI NO₂ satellite retrievals. *Atmospheric*
710 *Environment* 116, 1–11. doi:10.1016/j.atmosenv.2015.05.056.
- 711 de Foy, B., Lu, Z., and Streets, D. G. (2016). Impacts of control strategies, the Great Recession
712 and weekday variations on NO₂ columns above North American cities. *Atmospheric*
713 *Environment* 138, 74–86. doi:10.1016/j.atmosenv.2016.04.038.
- 714 De Smedt, I., Stavrakou, T., Hendrick, F., Danckaert, T., Vlemmix, T., Pinardi, G., et al. (2015).
715 Diurnal, seasonal and long-term variations of global formaldehyde columns inferred from
716 combined OMI and GOME-2 observations. *Atmospheric Chemistry and Physics*
717 *Discussions* 15, 12241–12300. doi:10.5194/acpd-15-12241-2015.
- 718 Duncan, B. N., Yoshida, Y., Olson, J. R., Sillman, S., Martin, R. V., Lamsal, L., et al. (2010).
719 Application of OMI observations to a space-based indicator of NO_x and VOC controls on
720 surface ozone formation. *Atmospheric Environment* 44, 2213–2223.
721 doi:10.1016/j.atmosenv.2010.03.010.
- 722 Duncan, B. N., Prados, A. I., Lamsal, L. N., Liu, Y., Streets, D. G., Gupta, P., et al. (2014).
723 Satellite data of atmospheric pollution for U.S. air quality applications: Examples of
724 applications, summary of data end-user resources, answers to FAQs, and common
725 mistakes to avoid. *Atmospheric Environment* 94, 647–662.
726 doi:10.1016/j.atmosenv.2014.05.061.
- 727 Duncan, B. N., Lamsal, L. N., Thompson, A. M., Yoshida, Y., Lu, Z., Streets, D. G., et al.
728 (2016). A space-based, high-resolution view of notable changes in urban NO_x pollution
729 around the world (2005-2014). *Journal of Geophysical Research: Atmospheres* 121, 976–
730 996. doi:10.1002/2015JD024121.
- 731 Fishman, J., Iraci, L. T., Al-Saadi, J., Chance, K., Chavez, F., Chin, M., et al. (2012). The United
732 States' Next Generation of Atmospheric Composition and Coastal Ecosystem
733 Measurements: NASA's Geostationary Coastal and Air Pollution Events (GEO-CAPE)
734 Mission. *Bulletin of the American Meteorological Society* 93, 1547–1566.
735 doi:10.1175/BAMS-D-11-00201.1.
- 736 Flynn, C. M., Pickering, K. E., Crawford, J. H., Lamsal, L., Krotkov, N., Herman, J., et al.
737 (2014). Relationship between column-density and surface mixing ratio: Statistical
738 analysis of O₃ and NO₂ data from the July 2011 Maryland DISCOVER-AQ mission.
739 *Atmospheric Environment* 92, 429–441. doi:10.1016/j.atmosenv.2014.04.041.

- 740 Flynn, L. E., Homstein, J., and Hilsenrath, E. (2004). The ozone mapping and profiler suite
741 (OMPS). The next generation of US ozone monitoring instruments. in *IGARSS 2004.*
742 *2004 IEEE International Geoscience and Remote Sensing Symposium*, 155.
743 doi:10.1109/IGARSS.2004.1368968.
- 744 Goldberg, D. L., Lamsal, L. N., Loughner, C. P., Swartz, W. H., Lu, Z., and Streets, D. G.
745 (2017). A high-resolution and observationally constrained OMI NO₂ satellite retrieval.
746 *Atmospheric Chemistry and Physics* 17, 11403–11421. doi:10.5194/acp-17-11403-2017.
- 747 Harley, R. A., Marr, L. C., Lehner, J. K., and Giddings, S. N. (2005). Changes in Motor Vehicle
748 Emissions on Diurnal to Decadal Time Scales and Effects on Atmospheric Composition.
749 *7. Environmental Science & Technology*.
- 750 Herman, J., Cede, A., Spinei, E., Mount, G., Tzortziou, M., and Abuhassan, N. (2009). NO₂
751 column amounts from ground-based Pandora and MFDOAS spectrometers using the
752 direct-sun DOAS technique: Intercomparisons and application to OMI validation.
753 *Journal of Geophysical Research* 114. doi:10.1029/2009JD011848.
- 754 Herman, J., Evans, R., Cede, A., Abuhassan, N., Petropavlovskikh, I., and McConville, G.
755 (2015). Comparison of ozone retrievals from the Pandora spectrometer system and
756 Dobson spectrophotometer in Boulder, Colorado. *Atmospheric Measurement Techniques*
757 8, 3407–3418. doi:10.5194/amt-8-3407-2015.
- 758 IGACO (2004). International Global Atmospheric Chemistry Observations Strategy Theme
759 Report. (No. ESA SP-1282, GW No. 159, WMO TD No. 1235). Available at:
760 <http://www.fao.org/gtos/igos/docs/IGACO-Theme-Report-2004-4.pdf> [Accessed March
761 6, 2018].
- 762 Ingmann, P., Veihelmann, B., Langen, J., Lamarre, D., Stark, H., and Courrèges-Lacoste, G. B.
763 (2012). Requirements for the GMES Atmosphere Service and ESA's implementation
764 concept: Sentinels-4/-5 and -5p. *Remote Sensing of Environment* 120, 58–69.
765 doi:10.1016/j.rse.2012.01.023.
- 766 Irie, H., Kanaya, Y., Akimoto, H., Tanimoto, H., Wang, Z., Gleason, J. F., et al. (2008).
767 Validation of OMI tropospheric NO₂ column data using MAX-DOAS measurements
768 deep inside the North China Plain in June 2006: Mount Tai Experiment 2006.
769 *Atmospheric Chemistry and Physics* 8, 6577–6586. doi:10.5194/acp-8-6577-2008.
- 770 Jaeglé, L., Steinberger, L., Martin, R. V., and Chance, K. (2005). Global partitioning of NO_x
771 sources using satellite observations: Relative roles of fossil fuel combustion, biomass
772 burning and soil emissions. *Faraday Discussions* 130, 407. doi:10.1039/b502128f.
- 773 Janjic, Z., and Gall, R. (2012). Scientific documentation of the NCEP nonhydrostatic multiscale
774 model on the B grid (NMMB). Part 1 Dynamics. UCAR/NCAR
775 doi:10.5065/D6WH2MZX.
- 776 Jin, X., Fiore, A. M., Murray, L. T., Valin, L. C., Lamsal, L. N., Duncan, B., et al. (2017).
777 Evaluating a Space-Based Indicator of Surface Ozone-NO_x-VOC Sensitivity Over

- 778 Midlatitude Source Regions and Application to Decadal Trends: Space-Based Indicator
779 of O₃ Sensitivity. *Journal of Geophysical Research: Atmospheres* 122, 10,439–10,461.
780 doi:10.1002/2017JD026720.
- 781 Judd, L. M. (2016). Investigating the spatiotemporal variability of NO₂ and photochemistry in
782 urban areas. Dissertation. Houston, TX: University of Houston.
- 783 Kim, J., Kim, M., and Choi, M. (2017). “Monitoring Aerosol Properties in East Asia from
784 Geostationary Orbit: GOCI, MI and GEMS,” in *Air Pollution in Eastern Asia: An
785 Integrated Perspective* ISSI Scientific Report Series. (Springer, Cham), 323–333.
786 doi:10.1007/978-3-319-59489-7_15.
- 787 Kim, S.-W., Heckel, A., McKeen, S. A., Frost, G. J., Hsie, E.-Y., Trainer, M. K., et al. (2006).
788 Satellite-observed U.S. power plant NO_x emission reductions and their impact on air
789 quality. *Geophysical Research Letters* 33. doi:10.1029/2006GL027749.
- 790 Kleist, D. T., and Ide, K. (2015a). An OSSE-Based Evaluation of Hybrid Variational–Ensemble
791 Data Assimilation for the NCEP GFS. Part I: System Description and 3D-Hybrid Results.
792 *Monthly Weather Review* 143, 433–451. doi:10.1175/MWR-D-13-00351.1.
- 793 Kleist, D. T., and Ide, K. (2015b). An OSSE-Based Evaluation of Hybrid Variational–Ensemble
794 Data Assimilation for the NCEP GFS. Part II: 4D-EnVar and Hybrid Variants. *Monthly
795 Weather Review* 143, 452–470. doi:10.1175/MWR-D-13-00350.1.
- 796 Knepp, T., Pippin, M., Crawford, J., Chen, G., Szykman, J., Long, R., et al. (2015). Estimating
797 surface NO₂ and SO₂ mixing ratios from fast-response total column observations and
798 potential application to geostationary missions. *Journal of Atmospheric Chemistry* 72,
799 261–286. doi:10.1007/s10874-013-9257-6.
- 800 Krotkov, N. A., Lamsal, L. N., Celarier, E. A., Swartz, W. H., Marchenko, S. V., Bucsela, E. J.,
801 et al. (2017). The version 3 OMI NO₂ standard product. *Atmospheric Measurement
802 Techniques* 10, 3133–3149. doi:10.5194/amt-10-3133-2017.
- 803 Lamsal, L. N., Martin, R. V., van Donkelaar, A., Celarier, E. A., Bucsela, E. J., Boersma, K. F.,
804 et al. (2010). Indirect validation of tropospheric nitrogen dioxide retrieved from the OMI
805 satellite instrument: Insight into the seasonal variation of nitrogen oxides at northern
806 midlatitudes. *Journal of Geophysical Research* 115. doi:10.1029/2009JD013351.
- 807 Lamsal, L. N., Janz, S. J., Krotkov, N. A., Pickering, K. E., Spurr, R. J. D., Kowalewski, M. G.,
808 et al. (2017). High-resolution NO₂ observations from the Airborne Compact
809 Atmospheric Mapper: Retrieval and validation: High-Resolution NO₂ Observations.
810 *Journal of Geophysical Research: Atmospheres* 122, 1953–1970.
811 doi:10.1002/2016JD025483.
- 812 Leitch, J. W., Delker, T., Good, W., Ruppert, L., Murcray, F., Chance, K., et al. (2014). The
813 GeoTASO airborne spectrometer project. doi:10.1117/12.2063763.

- 814 Levelt, P. F., Oord, G. H. J. van den, Dobber, M. R., Malkki, A., Visser, H., Vries, J. de, et al.
815 (2006). The ozone monitoring instrument. *IEEE Transactions on Geoscience and Remote*
816 *Sensing* 44, 1093–1101. doi:10.1109/TGRS.2006.872333.
- 817 Liang, J., Horowitz, L. W., Jacob, D. J., Wang, Y., Fiore, A. M., Logan, J. A., et al. (1998).
818 Seasonal budgets of reactive nitrogen species and ozone over the United States, and
819 export fluxes to the global atmosphere. *J. Geophys. Res.* 103, 13435–13450.
820 doi:10.1029/97JD03126.
- 821 Martin, R. V. (2003). Global inventory of nitrogen oxide emissions constrained by space-based
822 observations of NO₂ columns. *Journal of Geophysical Research* 108.
823 doi:10.1029/2003JD003453.
- 824 Martin, R. V., Fiore, A. M., and Van Donkelaar, A. (2004). Space-based diagnosis of surface
825 ozone sensitivity to anthropogenic emissions. *Geophysical Research Letters* 31.
826 doi:10.1029/2004GL019416.
- 827 McLinden, C. A., Fioletov, V., Boersma, K. F., Krotkov, N., Sioris, C. E., Veeffkind, J. P., et al.
828 (2012). Air quality over the Canadian oil sands: A first assessment using satellite
829 observations. *Geophysical Research Letters* 39, n/a-n/a. doi:10.1029/2011GL050273.
- 830 National Academies of Sciences, Engineering, and Medicine (2018). Thriving on Our Changing
831 Planet: A Decadal Strategy for Earth Observation from Space. Washington, DC: The
832 National Academies Press. <https://doi.org/10.17226/24938>.
833
- 834 Nowlan, C. R., Liu, X., Leitch, J. W., Chance, K., González Abad, G., Liu, C., et al. (2016).
835 Nitrogen dioxide observations from the Geostationary Trace gas and Aerosol Sensor
836 Optimization (GeoTASO) airborne instrument: Retrieval algorithm and measurements
837 during DISCOVER-AQ Texas 2013. *Atmospheric Measurement Techniques* 9, 2647–
838 2668. doi:10.5194/amt-9-2647-2016.
- 839 Palmer, P. I., Jacob, D. J., Chance, K., Martin, R. V., Spurr, R. J. D., Kurosu, T. P., et al. (2001).
840 Air mass factor formulation for spectroscopic measurements from satellites: Application
841 to formaldehyde retrievals from the Global Ozone Monitoring Experiment. *J. Geophys.*
842 *Res.* 106, 14539–14550. doi:10.1029/2000JD900772.
- 843 Richter, A., Burrows, J., Nüss, H., Granier, C., and Niemeier, U. (2005). Increase in tropospheric
844 nitrogen dioxide over China observed from space. *Nature* 437, 129–32.
845 doi:10.1038/nature04092.
- 846 Russell, A. R., Valin, L. C., Bucsele, E. J., Wenig, M. O., and Cohen, R. C. (2010). Space-based
847 Constraints on Spatial and Temporal Patterns of NO_x Emissions in California,
848 2005–2008. *Environmental Science & Technology* 44, 3608–3615.
849 doi:10.1021/es903451j.
- 850 Russell, A. R., Perring, A. E., Valin, L. C., Bucsele, E. J., Browne, E. C., Wooldridge, P. J., et al.
851 (2011). A high spatial resolution retrieval of NO₂ column densities from OMI: method

852 and evaluation. *Atmospheric Chemistry and Physics* 11, 8543–8554. doi:10.5194/acp-11-
853 8543-2011.

854 Russell, A. R., Valin, L. C., and Cohen, R. C. (2012). Trends in OMI NO₂ observations over the
855 United States: effects of emission control technology and the economic recession.
856 *Atmospheric Chemistry and Physics* 12, 12197–12209. doi:10.5194/acp-12-12197-2012.

857 Schroeder, J. R., Crawford, J. H., Fried, A., Walega, J., Weinheimer, A., Wisthaler, A., et al.
858 (2017). New insights into the column CH₂O/NO₂ ratio as an indicator of near-surface
859 ozone sensitivity: CH₂O/NO₂ as Indicator of O₃ Sensitivity. *Journal of Geophysical*
860 *Research: Atmospheres* 122, 8885–8907. doi:10.1002/2017JD026781.

861 SEDAC (NASA Socioeconomic Data and Applications Center) Center for International Earth
862 Science Information Network - CIESIN - Columbia University, and Information
863 Technology Outreach Services - ITOS - University of Georgia. 2013. Global Roads
864 Open Access Data Set, Version 1 (gROADSv1). Palisades, NY: NASA Socioeconomic
865 Data and Applications Center (SEDAC). <http://dx.doi.org/10.7927/H4VD6WCT>.
866 [Accessed March 18, 2018].
867

868 Sussmann, R., Stremme, W., Burrows, J. P., Richter, A., Seiler, W., and Rettinger, M. (2005).
869 Stratospheric and tropospheric NO₂ variability on the diurnal and annual scale: a
870 combined retrieval from ENVISAT/SCIAMACHY and solar FTIR at the Permanent
871 Ground-Truthing Facility Zugspitze/Garmisch. *Atmospheric Chemistry and Physics* 5,
872 2657–2677.

873 Travis, K. R., Jacob, D. J., Fisher, J. A., Kim, P. S., Marais, E. A., Zhu, L., et al. (2016). Why do
874 models overestimate surface ozone in the Southeast United States? *Atmospheric*
875 *Chemistry and Physics* 16, 13561–13577. doi:10.5194/acp-16-13561-2016.

876 Tzortziou, M., Herman, J. R., Cede, A., Loughner, C. P., Abuhassan, N., and Naik, S. (2015).
877 Spatial and temporal variability of ozone and nitrogen dioxide over a major urban
878 estuarine ecosystem. *Journal of Atmospheric Chemistry* 72, 287–309.
879 doi:10.1007/s10874-013-9255-8.

880 Valin, L. C., Russell, A. R., Hudman, R. C., and Cohen, R. C. (2011a). Effects of model
881 resolution on the interpretation of satellite NO₂ observations. *Atmospheric Chemistry and*
882 *Physics* 11, 11647–11655. doi:10.5194/acp-11-11647-2011.

883 Valin, L. C., Russell, A. R., Bucsela, E. J., Veefkind, J. P., and Cohen, R. C. (2011b).
884 Observation of slant column NO₂ using the super-zoom mode of AURA-OMI.
885 *Atmospheric Measurement Techniques Discussions* 4, 1989–2005. doi:10.5194/amtd-4-
886 1989-2011.

887 Valin, L. C., Russell, A. R., and Cohen, R. C. (2013). Variations of OH radical in an urban
888 plume inferred from NO₂ column measurements. *Geophysical Research Letters* 40,
889 1856–1860. doi:10.1002/grl.50267.

- 890 Valin, L. C., Russell, A. R., and Cohen, R. C. (2014). Chemical feedback effects on the spatial
891 patterns of the NO_x weekend effect: a sensitivity analysis. *Atmospheric Chemistry and*
892 *Physics Discussions* 13, 19173–19192. doi:10.5194/acpd-13-19173-2013.
- 893 van der A, R. J., Eskes, H. J., Boersma, K. F., van Noije, T. P. C., Van Roozendaal, M., De
894 Smedt, I., et al. (2008). Trends, seasonal variability and dominant NO_x source derived
895 from a ten year record of NO₂ measured from space. *Journal of Geophysical Research*
896 113. doi:10.1029/2007JD009021.
- 897 van Geffen, J. H. G., Boersma, K. F., Eskes, H. J., Maasakkers, J. D., and Veefkind, J. P. (2017).
898 TROPOMI ATBD of the total and tropospheric NO₂ data products. KNMI Available at:
899 [http://www.tropomi.eu/sites/default/files/files/S5P-KNMI-L2-0005-RP-](http://www.tropomi.eu/sites/default/files/files/S5P-KNMI-L2-0005-RP-TROPOMI_ATBD_NO2_data_products-v1p1p0-20170816_signed.pdf)
900 [TROPOMI_ATBD_NO2_data_products-v1p1p0-20170816_signed.pdf](http://www.tropomi.eu/sites/default/files/files/S5P-KNMI-L2-0005-RP-TROPOMI_ATBD_NO2_data_products-v1p1p0-20170816_signed.pdf) [Accessed
901 January 16, 2018].
- 902 Veefkind, J. P., Aben, I., McMullan, K., Förster, H., de Vries, J., Otter, G., et al. (2012).
903 TROPOMI on the ESA Sentinel-5 Precursor: A GMES mission for global observations of
904 the atmospheric composition for climate, air quality and ozone layer applications. *Remote*
905 *Sensing of Environment* 120, 70–83. doi:10.1016/j.rse.2011.09.027.
- 906 Yang, K., Carn, S. A., Ge, C., Wang, J., and Dickerson, R. R. (2014). Advancing measurements
907 of tropospheric NO₂ from space: New algorithm and first global results from OMPS.
908 *Geophysical Research Letters* 41, 4777–4786. doi:10.1002/2014GL060136.
- 909 Zhou, Y., Brunner, D., Spurr, R. J. D., Boersma, K. F., Sneep, M., Popp, C., et al. (2010).
910 Accounting for surface reflectance anisotropy in satellite retrievals of tropospheric NO₂.
911 *Atmospheric Measurement Techniques Discussions* 3, 1971–2012. doi:10.5194/amtd-3-
912 1971-2010.
- 913 Zhu, L., Jacob, D. J., Mickley, L. J., Marais, E. A., Cohan, D. S., Yoshida, Y., et al. (2014).
914 Anthropogenic emissions of highly reactive volatile organic compounds in eastern Texas
915 inferred from oversampling of satellite (OMI) measurements of HCHO columns.
916 *Environmental Research Letters* 9, 114004. doi:10.1088/1748-9326/9/11/114004.
- 917 Zhu, L., Jacob, D. J., Keutsch, F. N., Mickley, L. J., Scheffe, R., Strum, M., et al. (2017).
918 Formaldehyde (HCHO) As a Hazardous Air Pollutant: Mapping Surface Air
919 Concentrations from Satellite and Inferring Cancer Risks in the United States.
920 *Environmental Science & Technology* 51, 5650–5657. doi:10.1021/acs.est.7b01356.
- 921 Zoogman, P., Liu, X., Suleiman, R. M., Pennington, W. F., Flittner, D. E., Al-Saadi, J. A., et al.
922 (2017). Tropospheric emissions: Monitoring of pollution (TEMPO). *Journal of*
923 *Quantitative Spectroscopy and Radiative Transfer* 186, 17–39.
924 doi:10.1016/j.jqsrt.2016.05.008.
- 925 Figure 1: Global map of OMPS tropospheric NO₂ vertical columns for June 2016 and June 2017
926 averaged to 0.25° x 0.25° with the overlaid approximate spatial coverages of the planned
927 geostationary platforms: TEMPO over North America, Sentinel-4 over Europe, and GEMS over
928 East Asia.

929

930 Figure 2: Tropospheric NO₂ data from OMPS aboard Suomi-NPP for single overpasses on (a)
931 June 9th 2016 over South Korea and (b) June 27th, 2017 over California. The monthly averaged
932 0.25° x 0.25° tropospheric NO₂ from OMPS is shown for (c) June 2016 over South Korea and
933 (d) June 2017 over California. White polygons in each map outline the area of the GeoTASO
934 flights.

935

936 Figure 3: Maps of (a) the Seoul Metropolitan Area (SMA) and (b) the Los Angeles Basin. Major
937 roads (SEDAC, 2013) are drawn in red (I-5, I-10, and CA-60 are outlined in blue in Panel b).
938 Pandora sites are labeled with white star icons, and regions discussed in the paper labeled in
939 yellow. The green triangle in the SMA is Mount Gwanaksan, and the airplane icon in Los
940 Angeles depicts the location of LAX Airport. GeoTASO rasters cover the approximate area
941 depicted by the white polygons.

942

943 Figure 4: Maps of GeoTASO NO₂ DSCs over SMA on June 9th, 2016 for (a) Raster 1 from
944 08:00-10:00 LT, (b) Raster 2 from 10:00-12:00 LT, (c) Raster 3 from 14:00-16:00 LT, and (d)
945 Raster 4 from 16:00-18:00 LT. Pandora sites are labeled with white star icons. Rasters 1 and 3
946 includes wind vectors averaged through the lowest 500 m agl from the full resolution Global
947 Data Assimilation System (GDAS) at (a) 00:00 UTC (09:00 LT) and (c) 06:00 UTC (15:00 LT).

948

949 Figure 5: Box plots showing the percentile distributions of NO₂ DSCs for each Raster in SMA
950 from Figure 4. The shaded box shows 25th-75th percentile range with the whiskers extending to
951 the 5th and 95th percentiles. The solid line dividing each shaded box is the median.

952

953 Figure 6: Hourly averaged NO₂ vertical columns observed by ground-based Pandora
954 spectrometers at Yonsei University, Olympic Park, and Mount Taehwa. Grey lines are the
955 individual day diurnal averages from May 5th through June 15th, 2016. June 9th, 2016 is overlaid
956 in red, the weekday (Monday-Friday) averages are the black solid lines, and weekend (Saturday-
957 Sunday) average observations are the black dashed lines.

958

959 Figure 7: Maps of GeoTASO NO₂ DSCs over the LA Basin on June 27th, 2017. Raster 1 from
960 08:30-10:00 LT is shown in a and b, Raster 2 from 12:15-13:45 LT is shown in c and d, and
961 Raster 3 from 16:45-18:15 LT is shown in e and f. Panels a, c, and e are at 750 m x 750 m
962 resolution, whereas b, d, and f are the DSCs binned to 3 km x 3 km spatial resolution. Overlaid
963 are the boundary layer averaged wind vectors from the NAM-CONUS 3-km nest analysis for
964 16:00 UTC (09:00 LT) in a and b, 20:00 UTC (13:00 LT) in c and d, and 00:00 UTC (17:00 LT)
965 in e and f.

966

967 Figure 8: Box plots showing the percentile distributions of NO₂ DSCs for each raster in the LA
968 Basin from Figure 7. The shaded box shows 25th-75th percentile range with the whiskers
969 extending to the 5th and 95th percentiles. The solid line dividing each shaded box is the median.

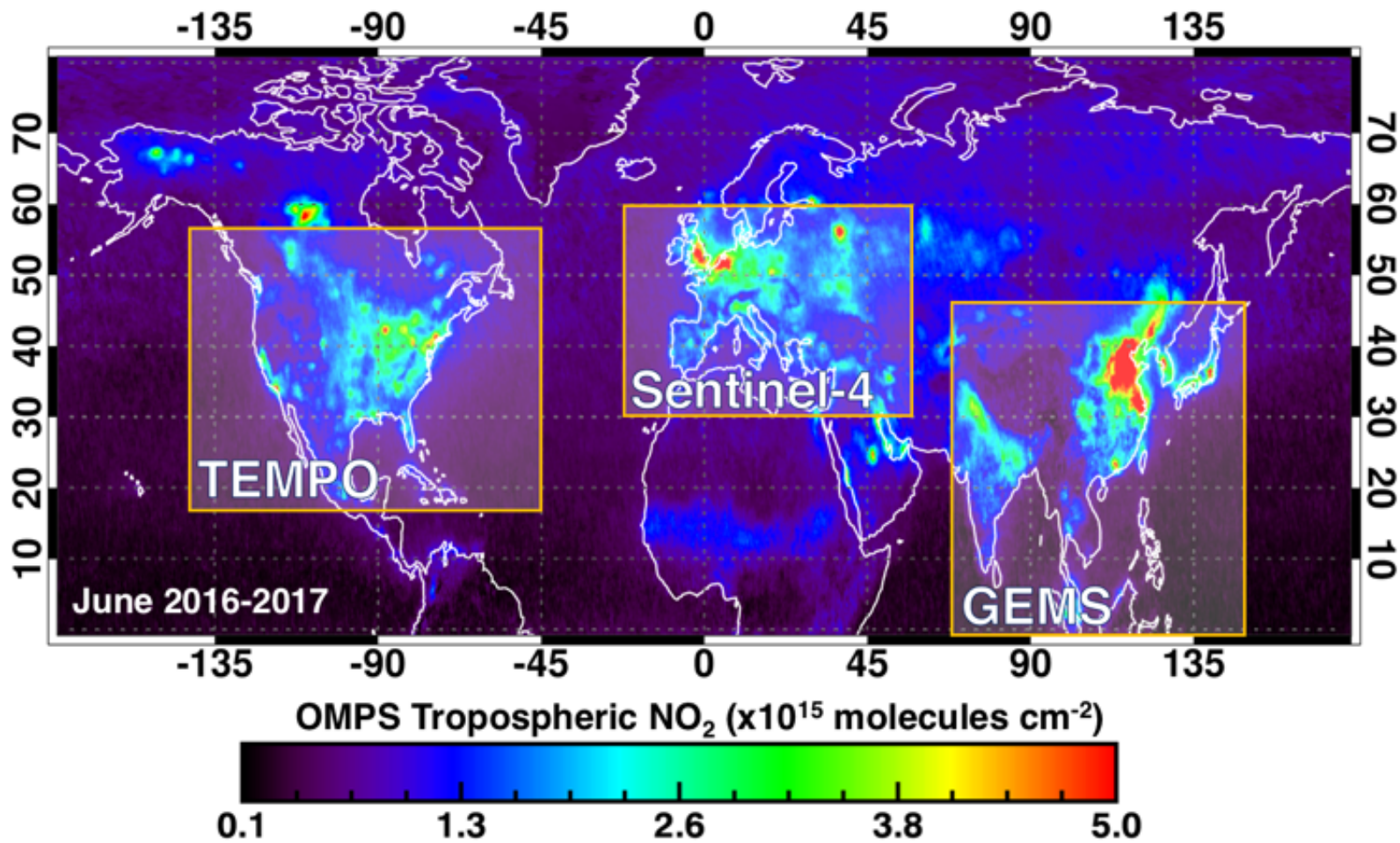
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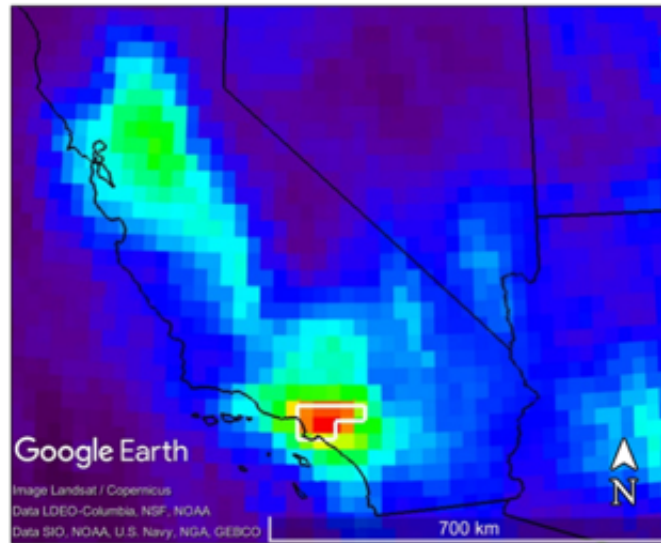
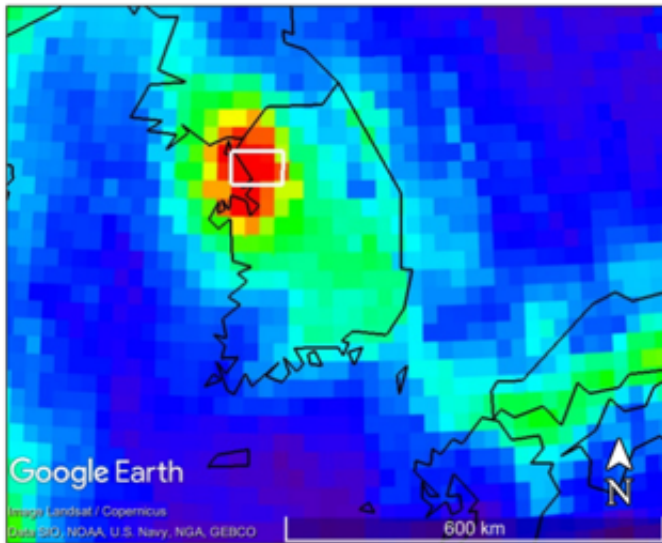
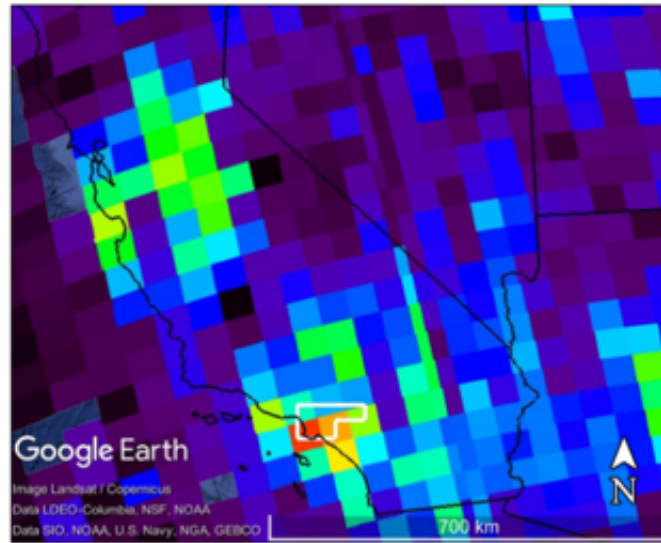
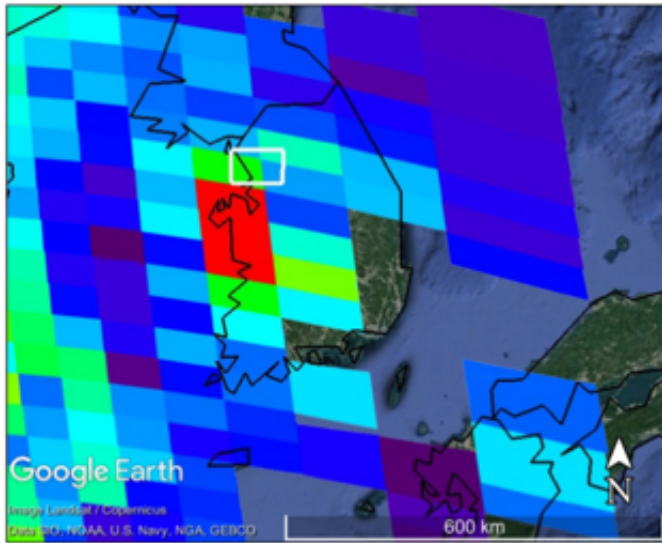
971 Figure 9: (a) Relative humidity from the NAM-CONUS 3-km nest at 20:00 UTC (13:00 LT)
972 with overlaid modeled boundary layer averaged wind vectors and a white contour at 40% relative
973 humidity boundary indicating the sea breeze front position and (b) NO₂ DSCs over the western
974 side of the LA Basin during Raster 2 with the indicated sea breeze front position identified from

975 the NAM-CONUS 3-km nest 40% relative humidity contour at 19:00UTC (long dashes: 12:00
976 LT), 20:00UTC (solid: 13:00 LT) and 21:00UTC (dotted: 14:00 LT).

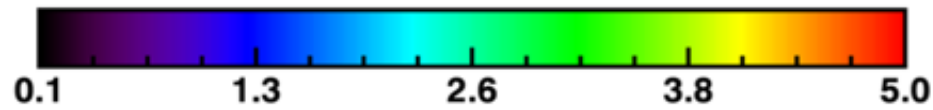
977

978 Figure 10. Hourly averaged NO₂ vertical columns observed by ground-based Pandora
979 spectrometers at UCLA, Los Angeles Main Street, CalTech, Pico Rivera, Ontario and Fontana.
980 Grey lines are the individual day diurnal averages from June 15th through July 15th, 2017. June
981 27th, 2016 is overlaid in red, the weekday (Monday-Friday) averages are the black solid lines,
982 and weekend (Saturday-Sunday) average observations are the black dashed lines.

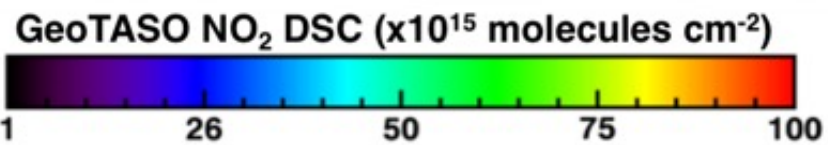
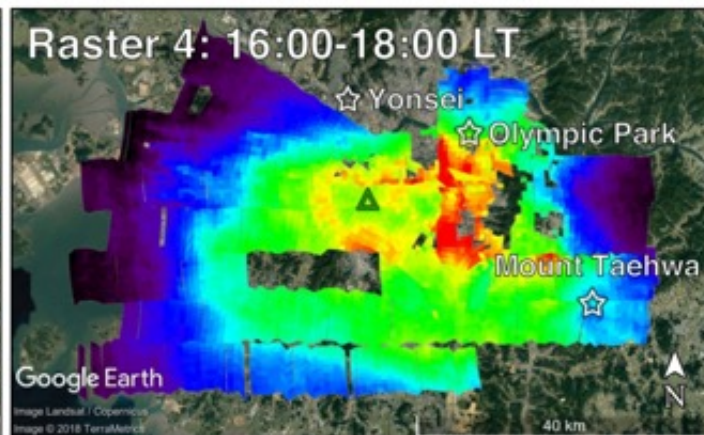
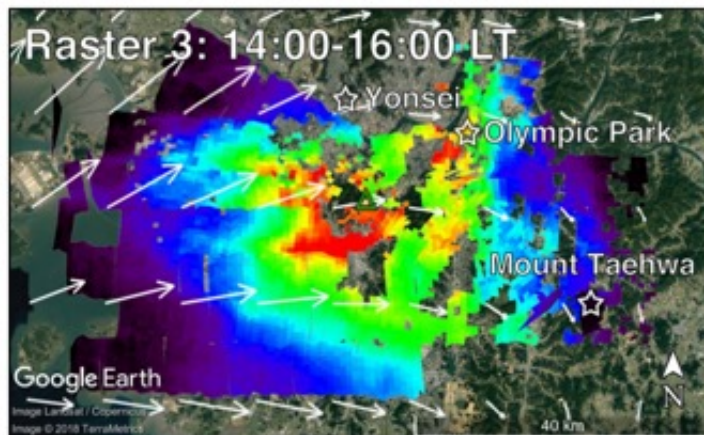
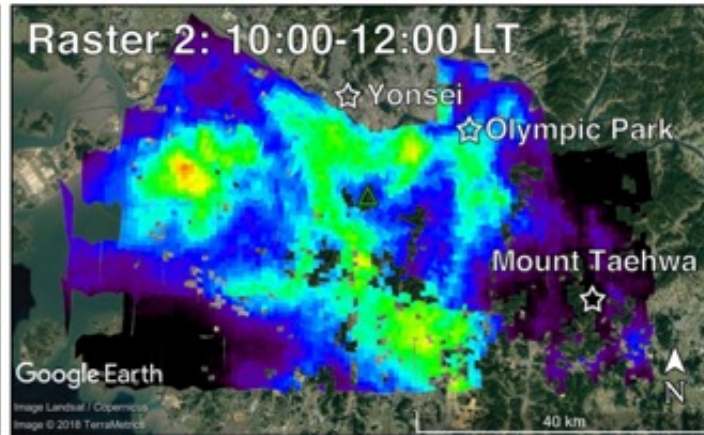
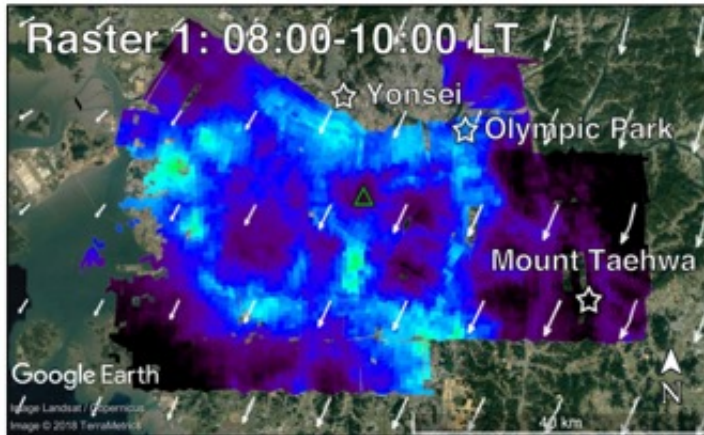


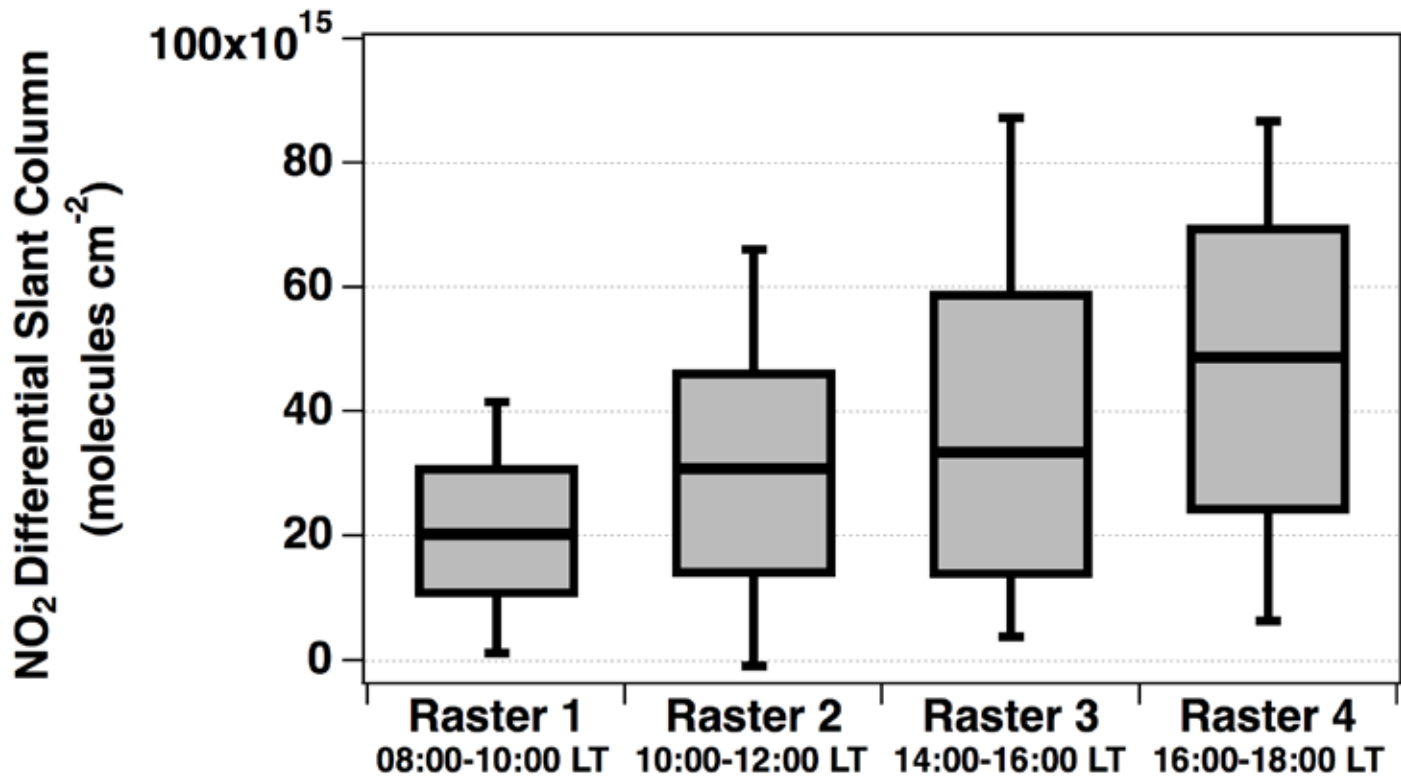


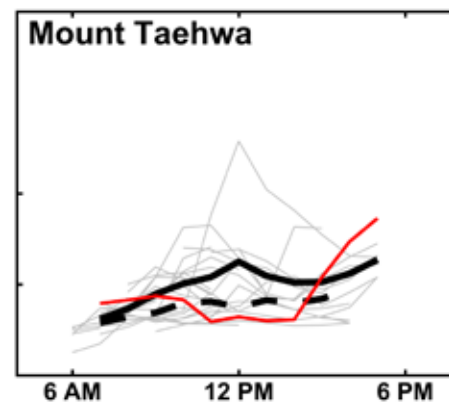
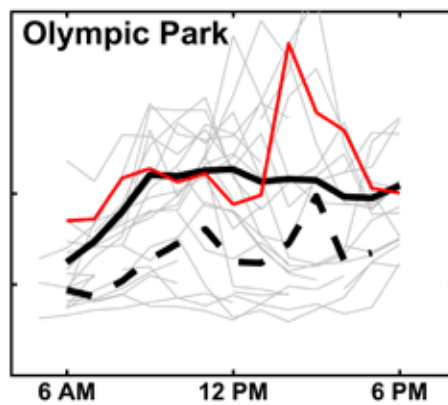
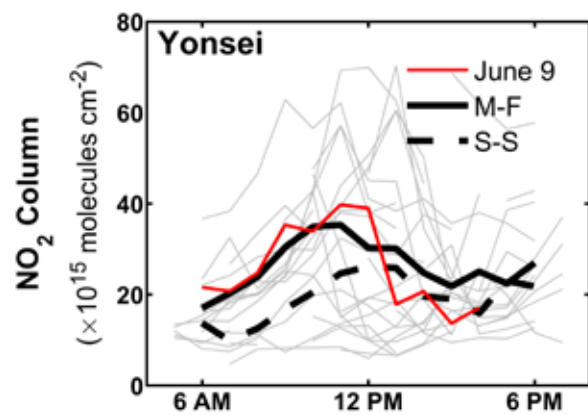
OMPS Tropospheric NO₂ (x10¹⁵ molecules cm⁻²)

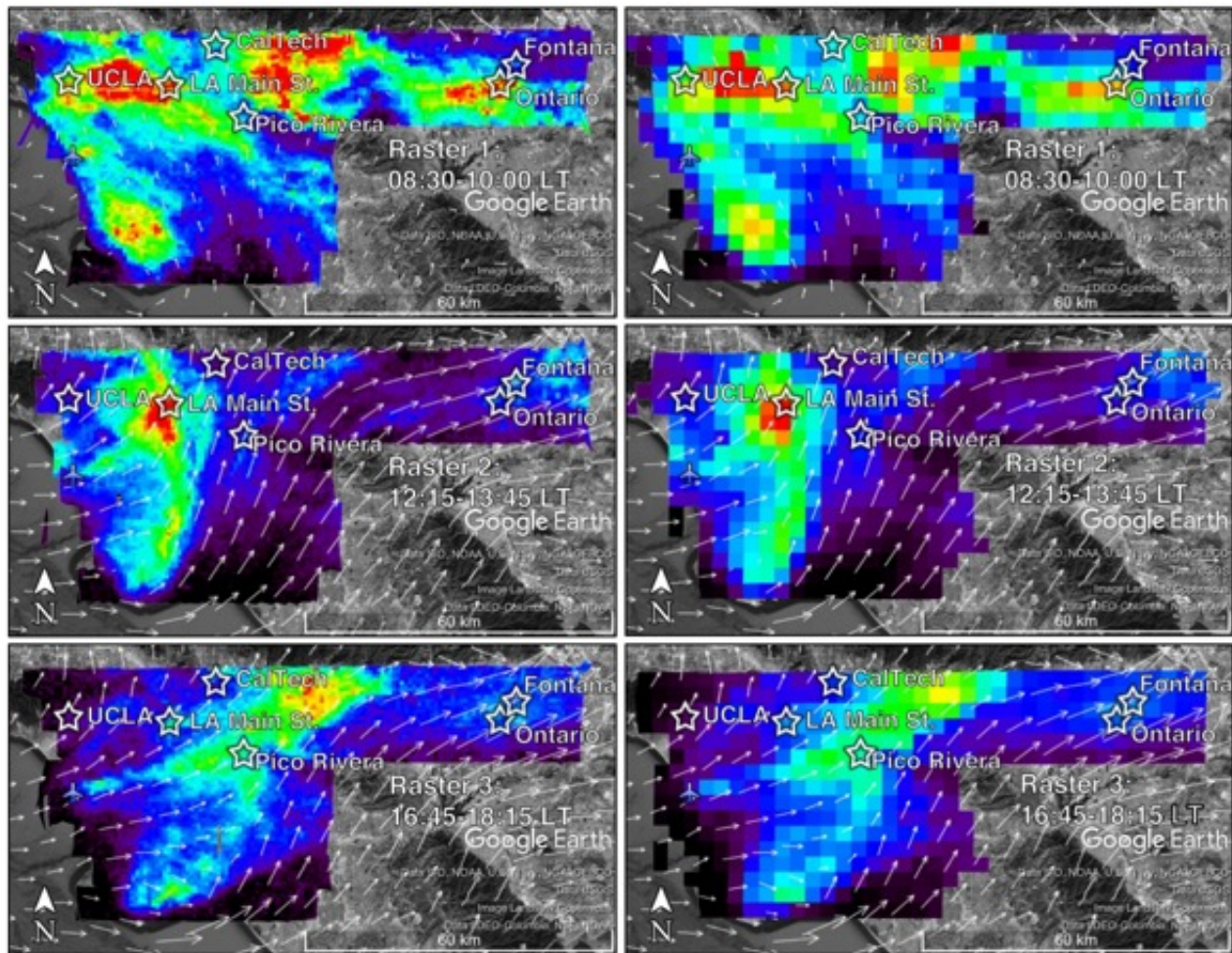




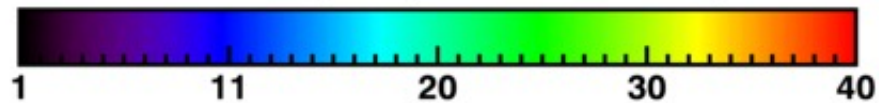


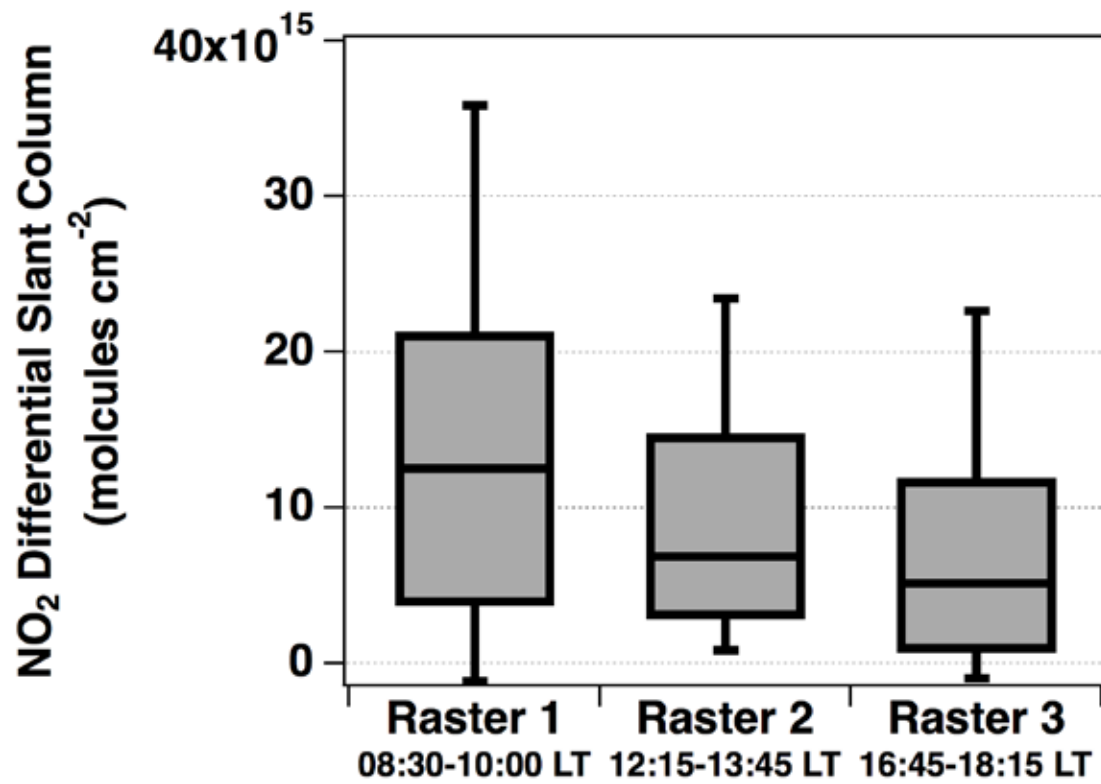


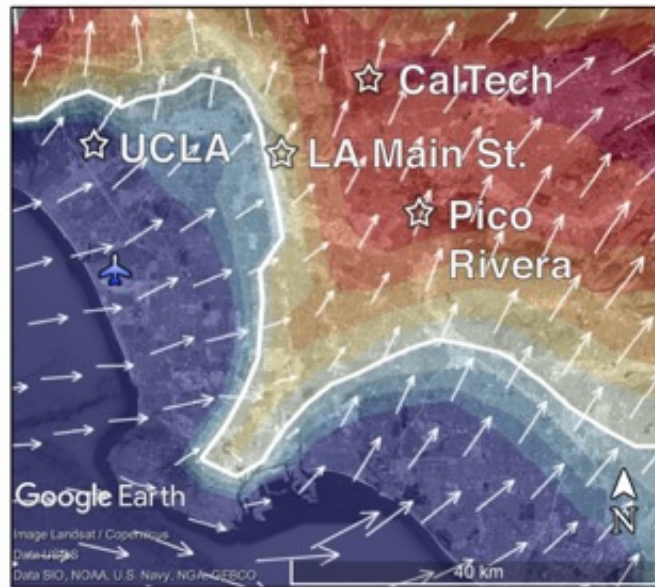




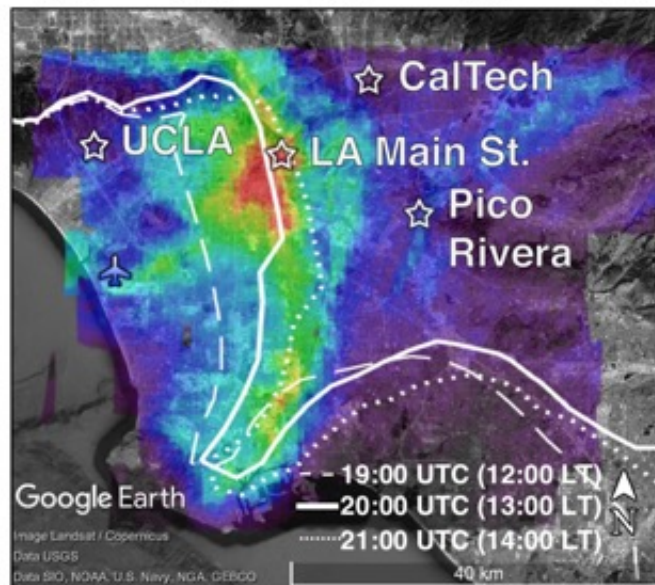
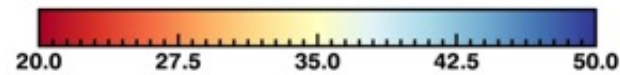
GeoTASO NO₂ DSC (x10¹⁵ molecules cm⁻²)







Relative Humidity (%)
NAM-CONUS 3-km



GeoTASO NO₂ DSC
(x10¹⁵ molecules cm⁻²)

