Evaluation of correlated Pandora column NO₂ and *in situ* surface NO₂ measurements during GMAP campaign

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1 Abstract.

To validate the Geostationary Environment Monitoring Spectrometer (GEMS), the GEMS 2 Map of Air Pollution (GMAP) campaign was conducted during 2020–2021 by integrating 3 Pandora Asia Network, aircraft, and in situ measurements. In the present study, GMAP-2020 4 measurements were applied to evaluate urban air quality and explore the synergy of Pandora 5 6 column (PC) NO₂ measurements and surface in situ (SI) NO₂ measurements for Seosan, South 7 Korea, where large point source (LPS) emissions are densely clustered. Due to the difficulty of interpreting the effects of LPS emissions on air quality downwind of Seosan using SI 8 9 monitoring networks alone, we explored the combined analysis of both PC-NO₂ and SI-NO₂ measurements. Agglomerative hierarchical clustering using vertical meteorological variables 10 combined with PC-NO₂ and SI-NO₂ yielded three distinct conditions: synoptic wind-dominant 11 (SD), mixed (MD), and local wind-dominant (LD). These results suggest meteorology-12 dependent correlations between PC-NO₂ and SI-NO₂. Overall, yearly daytime mean (11:00-13 17:00 KST) PC-NO₂ and SI-NO₂ statistical data showed good linear correlations (R = -0.73); 14 however, the differences in correlations were largely attributed to meteorological conditions. 15 16 SD conditions characterized by higher wind speeds and advected marine boundary layer heights suppressed fluctuations in both PC-NO₂ and SI-NO₂, driving a uniform vertical NO₂ 17 18 structure with higher correlations, whereas under LD conditions, LPS plumes were decoupled from the surface or were transported from nearby cities, weakening correlations through 19 20 anomalous vertical NO₂ gradients. The discrepancies suggest that using either PC-NO₂ or SI-NO₂ observations alone involves a higher possibility of uncertainty under LD conditions or 21 22 prevailing transport processes. However, under MD conditions, both pollution ventilation due to high surface wind speeds and daytime photochemical NO₂ loss contributed to stronger 23 24 correlations through a decline in both PC-NO₂ and SI-NO₂ toward noon. Thus, Pandora Asia Network observations collected over 13 Asian countries since 2021 can be utilized for detailed 25 investigation of the vertical complexity of air quality, and the conclusions can be also applied 26 when performing GEMS observation interpretation in combination with SI measurements. 27

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29 **1. Introduction**

Rapid developments in environmental remote sensing have led to a new era of air quality
observations, and recent hyperspectral data retrieval technologies have allowed for routine and

accurate monitoring of air pollutants at high spatial and temporal resolution. In particular, the Geostationary Environment Monitoring Spectrometer (GEMS), which was launched on February 18, 2020, measures the total and tropospheric air pollutant columns hourly at spatial resolutions of $7 \text{ km} \times 8 \text{ km}$ for gas and $3.5 \text{ km} \times 8 \text{ km}$ for aerosols (Kim et al., 2020), facilitating the tracking of pollution transport from local to synoptic scales.

37 Recent studies have revealed the potential of satellite observations to evaluate surface air 38 quality, particularly in regions with sparse air quality-monitoring networks. The main approach 39 is to convert column density to surface concentrations using a shape factor of the ratio of the 40 partial column (Ω_{z_0}) within the lowest layer (z_0) to the total column (Ω_{total}) (Zhao *et al.*, 2019), 41 as follows:

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$$S = \frac{\Omega_{z_0}}{\Omega_{total}} \times \frac{C}{\Delta z_o}$$

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where S, C, and Δz_o are the surface concentration, column density, and lowest layer thickness, 43 44 respectively. Acquiring accurate profile shape information is critical for determining the relationship between the column amount and surface concentration because the shape factor is 45 spatiotemporally variable. Considering this, numerous studies have obtained close 46 relationships using chemical transport model simulations, aircraft in situ measurements, and 47 satellite observations with high correlation coefficients (R) of 0.7 or more and used them to 48 49 scale up surface NO₂ to column NO₂ (Wang and Christopher, 2003; Boersma et al., 2009; Lamsal et al., 2010). This strong correlation can be explained by the generally uniform 50 planetary boundary layer height (PBLH), and by aerosol type and abundance, which is also the 51 52 case for trace gases.

By contrast, the implications of weak correlations between column and surface measurements remain unclear. Engel-Cox *et al.* (2004) found a negative correlation of AOD and surface PM_{2.5} in northwestern USA, and explained it based on elevated haze decoupled

from the surface. Thompson et al. (2019) examined weak correlations between Pandora column 56 (PC) measurements and surface in situ (SI) observations of NO₂ over the Yellow Sea during 57 the Korea–US air quality (KORUS-AQ) field study, and suggested, as a possible reason, the 58 transported non-uniform plumes originated in China and Seoul hundreds of meters above the 59 ground from the surface layer. The estimated surface PM2.5 concentration was weakly 60 correlated (R = 0.4-0.49) with observed PM_{2.5} concentrations in Seoul, because only PBLH 61 62 was added to the multi-linear regression model to correlate AOD to surface PM2.5 (Kim et al., 2021). This effect may be related to the significant impact of long-range transport on $PM_{2.5}$, 63 64 with a contribution of up to 39% in Seoul (Lee et al., 2021). Thus, the wide variability in the degree of correlation between PC-PM and SI-PM is closely related to vertical profile variability 65 (Flynn et al., 2016). 66

67 It appears highly probable that several factors are responsible for the correlations between PC-NO₂ and SI-NO₂; therefore, it is necessary to improve our understanding of the degree of 68 correlation through detailed measurements, including column concentration. In this study, we 69 focused on the impact of meteorology and chemistry on correlation variability using PC, SI 70 and aircraft measurements, as well as meteorological observations. Understanding vertical 71 profile variability is also useful for evaluating the effects of various emissions on urban air 72 73 quality, particularly in areas neighboring active large point source (LPS) emissions sites. 74 Quantifying the impact of LPS emissions on downwind cities remains challenging due to the 75 lack of three-dimensional (3D) measurements. Accurate vertical profile data are also useful for improving remote sensing retrieval algorithms, because the profile shape contributes to the 76 conversion of slant column density into vertical column density as part of the air mass factor. 77 78 In mid-2019, the Pandonia Global Network (PGN; https://pandonia-global-network.org) was launched, with support from the National Aeronautics and Space Administration (NASA) and 79

80 European Space Agency (ESA), to facilitate the validation and verification of low-orbit or

geostationary environmental satellites. This network is attempting to expand air quality monitoring through integration with existing long-term air quality monitoring stations. Since 2020, the National Institute of Environmental Research, Economic and Social Commission for Asia and the Pacific, and Korea Environment Corporation have been extending the Pandora Asia Network to include 13 Asian countries, with support from the Korea International Cooperation Agency. The Pandora Asia Network is expected to be widely used to study urban air quality in Asia, which is increasingly deteriorating due to rapid economic growth.

As part of the GEMS Map of Air Pollution (GMAP) campaign, a suite of Pandora instruments was deployed in Seosan, a South Korean coastal city, from November 2020 to January 2021 (GMAP-2020), and we applied GMAP-2020 measurements to explore the synergy of PC observations when evaluating air quality over Seosan. Further results from this research project are also reported in this special issue, including GEMS validation and urban air quality evaluations based on Pandora, aircraft, surface flux, and *in situ* surface chemical measurements conducted during GMAP-2020.

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96 2. GMAP-2020 campaign

97 GEMS was launched on February 19, 2020; it is the first instrument to observe air quality from a geostationary Earth orbit. GEMS provides hourly air quality data on aerosols and gases at a 98 99 spatial resolution of 7 km \times 8 km. It is a scanning ultraviolet (UV)-visible spectrometer that 100 observes key atmospheric constituents including O₃, NO₂, CO, SO₂, CH₂O, CHOCHO, 101 aerosols, clouds, and UV indices. This mission heralded a new era of satellite air quality monitoring and will be joined by NASA's Tropospheric Emissions: Monitoring of Pollution 102 103 (TEMPO) and ESA's Sentinel-4 to form the GEO Air Quality Constellation in ~3 years, to cover the most polluted region in the Northern Hemisphere. 104

105 During GMAP-2020, Pandora instruments (PA₁–PA₄) were deployed near large point

106 sources (LPS₁–LPS₄) in Seosan; *in situ* surface air quality monitoring systems (AQM₁–AQM₆) and meteorological observations (Met₁–Met₃) were also used in this study (see their locations 107 108 in Fig. 1). Aircraft measurements were also used to validate GEMS and diagnose LPSs located in industrial areas surrounding Seosan. We explored the synergy of Pandora observations and 109 SI measurements, based on measurements collected during GMAP-2020, by evaluating air 110 quality in industrial Seosan (where LPSs are densely clustered). We particularly investigated 111 112 the impacts of vertical profile and sub-pixel variability for trace gases and aerosols, for further GEMS validation. All measurement sites for both GMAP-2020 campaigns are indicated in 113 114 Figure 1.

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116 **3. Methods**

117 **3.1 Study area**

Seosan, the target area of the GMAP-2020 campaign, is a small city with a population of 118 174,780 in 2017; it is accessed via three expressways to the east and four national highways 119 120 cross the city. It is located in midwestern South Korea, and is affected by >300 emissions point sources including LPSs. Coal-fired power plants including Taean, Dangjin, the Hyundai 121 Dangjin steelworks, and the Daesan petrochemistry industrial complex (LPS₁-LPS₄, 122 123 respectively, in Fig. 1) have the highest emissions rates in South Korea. The Hyundai Dangjin steelworks (LPS₃) and Taean and Dangjin power plants (LPS₁ and LPS₂) emit 10.5, 11, and 124 8.8 Gg of NOx per year, respectively. Although Seosan accounts for only 1.8% of the 125 population of Seoul, its NOx emissions (10.2 Gg year⁻¹) account for 13.2% of its total NOx 126 emissions. The transportation sector of Seosan is a far greater NOx source than the industrial 127 sector of Seoul (ratio of 99:1); however, within Seosan, the industrial sector is on par with the 128 129 transport sector (52:48; http://airemiss.nier.go.kr).

130 During the past decade, the annual mean NO₂ level in Seosan has been 17 ppb, which is

approximately half of that in Seoul (31.2 ppb). NO₂ exhibits strong seasonal variation, reaching
a minimum in summer and maximum in winter, due to meteorological factors and greater
energy use during winter (Kim and Kim, 2020). Therefore, the timing of the GMAP-2020
campaign was well suited to tracking pollution.

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136 **3.2 Pandora measurements**

137 Pandora measures the UV and visible wavelengths (280-525 nm) of direct sunlight with a spectral resolution of 0.6 nm, to determine the vertical column density of NO₂, O₃, and HCHO 138 139 (Herman *et al.*, 2009). For measurements in Dobson units (DU; 1 DU = 26.9 Pmol cm⁻²), column NO₂ has a very high signal-to-noise ratio (700:1) and very high precision (0.01 DU) 140 for clear skies (Herman et al., 2009). The vertical column density of NO₂ can be determined 141 142 using DOAS software (Van Roozendael and Fayt, 2001). Pandora direct-sun measurements are advantageous in that the air mass factor is simplified, and therefore is dependent only on the 143 geography for a known solar zenith angle. 144

From retrieved Pandora measurements, tropospheric and total (=tropospheric + stratospheric) vertical column densities are both available. However, it should be noted that appreciable uncertainties cannot be neglected in the tropospheric NO₂ profiles obtained from Pandora instruments, particularly for the high aerosol-loading areas such as East Asia. In this background, we used total vertical column densities in the present study, and also confirmed that they have a high correlation with the tropospheric column densities observed in our study period with little change in stratospheric column density in space and time at the local scale.

Four Pandora instruments were installed at sites to the south of LPSs (Fig. 1) during the GMAP-2020 campaign, i.e., at Seosan Daehoji, Seosan Dongmun, Seosan City Council, and Seosan Super Site (PA_1 – PA_4 in Fig. 1). The presence of clouds reduces vertical column density precision by decreasing the number of photons arriving at Pandora instruments within a fixed 156 integration time. Therefore, the retrieved Pandora measurements were cloud-screened using an 157 observed cloud cover of 0.6. Cloud cover was provided by the Korea Meteorological 158 Administration (KMA), and the precision improvement afforded by cloud screening was 159 verified by comparing each Pandora-derived vertical column density with the median vertical 160 column density, with and without cloud screening within the inter-comparison period.

At PA₄, the operating period was extended to cover almost the entire year (November 12, 2020–October 30, 2021) including the GMAP-2020 campaign period, and the Pandora spectra were processed into vertical column density data for trace gases using the standard NO₂ algorithm in BlickP software provided by PGN (Cede, 2019). The resultant PC-NO₂ data were obtained from the PGN website (https://pandonia-global-network.org) for the 1-year period from Nov. 12, 2020 to Oct. 30, 2021, and were also used as PC-NO₂ statistics for PA₄, in this study.

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169 **3.3 Surface and airborne chemical measurements**

Hourly average data for SI-NO₂ over a period of 1 year were obtained from Ministry of 170 Environment AQM network stations in Seosan: Pandori, Leewon, Taean, Dongmoon, 171 Seongyeon, and Daesan (AQM₁-AQM₆, respectively, in Fig. 1). The Seosan Super Site 172 (PA4/AQM1) provided hourly data for NO and NOy via an NO-DIF-NOy analyzer (42i-Y; 173 174 Thermo Scientific, Waltham, MA, USA), and for PM_{2.5} chemical species using an ambient ion 175 monitor (AIM; URG 9000D, URG Corp., Chapel Hill, NC, USA). Weekly zero and span checks were conducted for NOy calibration, to ensure that differences between checks 176 remained <3%. Water-soluble ions in aerosol and gaseous species were measured hourly using 177 178 an AIM, and ion mass balance was used to ensure data quality under the quality control procedures of the AQM network installation and operation guidelines (NIER, 2021). 179

180 Aircraft measurements were conducted during the GMAP-2020 campaign period, and

nine flights were conducted on 8 days (Nov. 26, 27, and 28 and Dec. 1, 6, 8, 9, and 12, 2020). The horizontal and vertical distributions of NO₂ and O₃ over Seosan were measured during GMAP-2020 using an NO₂ monitor (T500U; Teledyne, Thousand Oaks, CA, USA) and an O₃ analyzer (TEI49C; Thermo Scientific) onboard the Cessna Grand Caravan 208 B. These instruments had response times of <40 and <20 s, and detection limits of 40 ppt and 1 ppb, respectively. The flight paths included a raster mode over all of Seosan at a height of 500–700 m and a profiling mode from 500 m to 1.5 km over PA₁ and PA₄ (Fig. 2).

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189 **3.4 Meteorological measurements**

Ground-based hourly observation data for meteorological variables were obtained from Seosan Automated Synoptic Observing System (ASOS) stations maintained by the KMA, and wind and temperature profile data were obtained twice daily (0000 and 1200 UTC) via a rawinsonde instrument at the Osan World Meteorological Organization upper air measurement station (47122) near Seosan. Due to time constraints of the sonde measurements, information on PBLH variation was obtained from Unified Model (UM) simulation results provided on the KMA website (https://afso.kma.go.kr).

During the GMAP-2020 campaign, a 3D sonic anemometer (CPEC200; Campbell Scientific, Logan, UT, USA) was also installed on the rooftop at PA₄ for turbulent flux measurements at the city–atmosphere interface (Hong *et al.*, 2019). All wind components and sonic temperatures were measured at a 10 Hz sampling rate, and ground-level sensitive heat flux was measured directly using a 30 min averaging period. Quality controls such as double rotation, spike removal, and outlier filtering were also applied.

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204 **3.5 Correlation analyses**

205 We examined the synergy of PC and SI data obtained during the GMAP-2020 campaign, and

combined these measurement data to evaluate air quality in Seosan, South Korea. We attempted
to interpret the meteorological and photochemistry data measured during GMAP-2020, and to
demonstrate that caution is required when attempting to study the vertical structure of air
pollutants using either surface observations or satellite data only, particularly in industrial
areas.

First, we examined the combined use of year-long PC-NO₂ and SI-NO₂ 211 212 measurements, and investigated the factors modulating their correlation. Numerous studies have examined the correlations between chemical species, including aerosols (Thomspon et 213 214 al., 2019; Wang et al., 2019; Kim et al, 2012; Jo et al., 2013; Sanchez et al., 1990; Kim et al, 2018). Wang et al. (2019) reported that aerosols moderately correlate with NO₂ due to the 215 frequent occurrence of lifted layers probably related to the transport of pollutants. Jo et al. 216 217 (2013) differentiated haze types using the trajectory speed and direction and different synoptic conditions. In this background, we hypothesized that their differences in PC-NO₂ and SI-NO₂ 218 were due to meteorological conditions, and performed k-means and agglomerative hierarchical 219 cluster analyses of various meteorological variables. Clustering is the grouping of objects that 220 are alike and are different from the objects belonging to other clusters. As a first step, k-means 221 Clustering was applied to find smaller clusters until each object was classified in one cluster. 222 223 Subsequently, agglomerative hierarchical steps are applied to make up for the shortcomings of k-means clustering, in which once merging (or splitting) is done, it can never be undone. More 224 225 details are found in Venkat Reddy et al. (2017). We used XLSTAT software (Addinsoft, Paris, France) for the cluster analysis with eight meteorological variables representing local and 226 synoptic circulations in the cluster analysis: surface wind speed (Wsfc), 925 hPa temperature 227 228 (T925), sea level pressure (Psfc), pressure tendency (dPsfc/dt), 850 hPa wind speed (W850) and its north-south and east-west components (NS850 and EW850), and 500 hPa geopotential 229 height (GPH500). We subtracted 30-day moving averages from all data to account for typical 230

seasonal variation. Monthly averages were used for PC-NO₂ analysis due to the limited
availability of hourly data.

233 Correlations between PC-NO₂ and SI-NO₂ were analyzed in each meteorological group 234 and the impact of photochemistry was interpreted based on case-specific features. We also 235 investigated correlations in association with near-surface micrometeorological variables such 236 as PBLH in each meteorological group.

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- 238 4. Results and Discussion
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240 4.1 Correlation analysis results for PC-NO₂ and SI-NO₂

The yearly PC-NO₂ statistics at four Pandora sites (PA₁–PA₄) are summarized in Table 1. The total averaged PC–NO₂ over all sites was 0.45 DU during GMAP-2020, which is well above the typical values (0.1–0.2 DU) for Anmyeondo (the location is shown in Figure 1), a representative background site (Herman *et al.*, 2018). Although site PA₃ is located in a rural area, it nevertheless exhibited the highest PC-NO₂ amounts, suggesting that plumes were frequently transported from nearby point sources and/or urban areas.

Scatter diagrams of hourly PC-NO₂ and SI-NO₂ measurements from Pandora sites PA₁-247 PA₃ (GMAP-2020) and PA₄ (yearly measurement; November 12, 2020–October 30, 2021) are 248 shown in Fig. 3a. These hourly data had a fair 1:1 linear relationship (R = 0.41), implying the 249 overall uniformity of NO₂ profiles, whereas the linear relationship with PC-NO₂ weakened as 250 SI-NO₂ levels increased (Fig. 3a). It appears that the SI-NO₂ has a distinct diurnal change 251 despite the same PC-NO₂, and higher variable surface NO₂ levels may result from the relatively 252 weaker linear relationship between PC-NO₂ and SI-NO₂. To explore these anti-correlation 253 cases further, we selected the lower and upper bounds of the tendencies; these are plotted in 254 Fig. 3b, which shows that PC-NO₂ was positively correlated with SI-NO₂ on February 24, 2021 255

(R = 0.88), while a negative correlation occurred on April 21, 2021 (R = -0.88), indicating a 256 wide range of case-specific correlations. The negative correlation on April 21 (Fig. 3b) implied 257 that the nonhomogeneous NO₂ distributions vertically were partially due to the photochemical 258 process. For example, the decrease in PC-NO₂ despite an increase in SI-NO₂ might have 259 occurred because NO₂ is removed by photochemical loss; it can occur more severely in the 260 upper atmosphere with high OH concentrations. Another possible reason is the occurrence of 261 262 lifted layers related to pollutant transport, yielding sharp changes in vertical concentration from the surface to the upper layer. The case-specific discussion follows. 263

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4.2 Impacts of meteorological conditions on correlations between PC-NO₂ and SI-NO₂

Our k-means cluster analysis distinguished three groups with the lowest within-group variance 266 267 and largest among-group variance. Among the total of 141 cases, 47, 66, and 28 were classified into groups 1–3, respectively. Thus, group 2 had the largest proportion of cases (47%) and 268 group 3 had the smallest (20%). The combination of meteorological components in group 1 269 270 indicated the end of a high-pressure system (Psfc > 0, dPsfc/dt < 0), with southerly winds (NS850 > 0) bringing warmer air (T925 > 0) to the region, leading to stable atmospheric 271 stratification and weak surface winds (Fig. 4). This group 1 meteorological mode appeared to 272 result in very weak NO₂ ventilation, which produced the highest PC-NO₂ and SI-NO₂ values. 273 274 Group 3 showed the opposite trend, with strong northerly winds bringing colder air into the 275 region, leading to an unstable atmosphere and stronger surface winds, and ultimately decreasing PC-NO₂ and SI-NO₂ to their lowest levels. 276

SI-NO₂ was approximately twice as high in group 1 than group 3, whereas PC-NO₂ showed no significant difference (Fig. 4a). We hypothesized that PBLH might also differ significantly under these micrometeorological conditions; therefore, we further explored daily maximum PBLH simulated by the Global Forecast System (GFS) and Lagrangian backward

trajectories obtained from Hybrid Single-Particle Lagrangian Integrated Trajectory 281 (HYSPLIT) and GFS system for the 141 cases. The mean simulated PBLH in Seosan, our study 282 area, was 942.1 \pm 405.3 m for 2020, which was similar to the annual mean daily maximum 283 PBLH (1,013.6 m) in Osan (Lee et al., 2013). However, the simulated PBLH differed 284 significantly among the three groups (767.0 \pm 304.8, 923.2 \pm 335.3, and 1,280.6 \pm 501.2 m for 285 groups 1–3, respectively). The PBLH for group 3 was 1.7-fold higher than that for group 1 286 287 (Fig. 4k). We also detected significant differences among the three groups in synoptic components of the lower troposphere including W850, as well as in local meteorological 288 289 parameters such as the sea breeze index (SBI) suggested by Biggs and Graves (1962), which

is defined as, $SBI = \frac{U^2}{C_p \Delta T}$, where *U* is Wsfc (Fig. 4c), C_p is specific heat, and ΔT is the temperature difference between T925 and the sea surface temperature. Thus, the SBI represents the ratio between inertial ($\rho U^2/2$) and buoyance forces ($\rho g C_p \Delta T$), where ρ is air density and *g* is gravity, and its value provides an indication of the likelihood of local circulation events such as sea breezes; at higher SBIs (i.e., SBI > 3), sea breezes cannot overcome the prevailing wind, whereas lower SBIs (i.e., 0 < SBI < 3) can indicate strong sea breezes.

In the example shown in Fig. 41, the SBI for groups $1-3 \text{ was } 0.1 \pm 4.5$, 0.1 ± 9.2 , and -0.2 ± 12.5 , respectively. Most SBIs in group 1 ranged from 0 to 3, indicating that group 1 corresponded to the dominant local circulation (LD), whereas the SBIs in group 3 had the lowest frequencies comparing 1 and 3, which corresponded to a dominant synoptic-scale circulation (SD). Group 2 can be considered a mixture of local and synoptic-scale circulation (MD). These results indicate that Seosan may experience frequent LD conditions (with sun on one third of the days of the year), with infrequent SD conditions (one fifth of all days).

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304 4.2.1 Relationship between daily mean PC-NO₂ and SI-NO₂ under LD, MD, and SD

305 conditions

Scatter diagrams of daytime mean PC-NO₂ and SI-NO₂ measurements at Seosan over the entire 306 1-year period are shown in Fig. 5. Based on the 141 cases, daytime mean values averaged 307 between 1100 and 1700 KST were used to reduce the effect of nocturnal PBLH variation. Other 308 data selection criteria included concurrent PC-NO2 and SI-NO2 measurements, with data 309 acquisition rates of >80% per day. Overall, PC-NO₂ and SI-NO₂ were strongly correlated (R =310 311 0.73; Fig. 5), suggesting that the vertical profiles were generally uniform in the PBL throughout all four seasons. The slope of the linear regression curve shown in Fig. 5a was 0.02 DU/ppb (= 312 0.53×10^{15} molecules cm⁻²/ppb), which is comparable to values (0.3–0.59 × 10¹⁵ molecules 313 cm⁻²/ppb) obtained previously in a study of surface and OMI-NO₂ measurements downwind 314 of strong point sources in Israeli cities (Boersma et al., 2009). The intercept (0.17 DU) was 315 316 within the range of previous Anmyeondo Pandora measurements, suggesting that intercepts of 0.15–0.2 DU may represent the local background PC-NO₂ amount (including the stratospheric 317 NO₂), rather than the influence of local anthropogenic NO₂ emissions. 318

We classified daily averaged PC-NO₂ and SI-NO₂ data according to the three meteorological conditions (LD, MD, and SD) and detected a weak correlation under LD conditions (Fig. 5b); the lowest coefficient of determination for the LD condition ($R^2 = 0.34$) was approximately half of those for the MD (0.359) and SD (0.64) conditions, suggesting that NO₂ vertical profiles were more complex under LD conditions, with anomalous layers.

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4.2.2 Diurnal variation in column-surface NO₂ under LD, MD, and SD conditions

Diurnal patterns of PC-NO₂, SI-NO₂, and O₃ under SD, MD, and LD conditions are shown in Fig. 6. Under LD conditions, PC-NO₂ increased from morning to afternoon (Fig. 6a), whereas under SD conditions, it had a weak morning peak and subsequent decrease until late afternoon (Fig. 6c). Under MD conditions, PC-NO₂ had one large peak in the morning and a shoulder peak in the late afternoon (Fig. 6b). However, SI-NO₂ showed nearly identical diurnal patterns among the three meteorological conditions, with an early morning peak followed by a second peak in the late afternoon (Fig. 6d–f). Diurnal patterns of O_3 were strongly associated with O_3 -NO₂ photochemical reactions under both LD and MD conditions (Fig. 6g–h), whereas no particular photochemical effects were detected under SD conditions (Fig. 6i).

A simple linear regression was applied to daytime average (1100-1700 LST) 335 336 measurements of both PC-NO₂ and SI-NO₂ under the three meteorological conditions, and yielded correlation coefficients (*R*) of 0.51 and 0.41 for SD and MD conditions, respectively; 337 338 however, LD conditions produced a significantly lower R (0.27). Thus, under SD conditions, strong synoptic winds suppressed PC-NO₂ and SI-NO₂ diurnal fluctuations, rendering them 339 similar to each other. Strong winds also inhibited local effects of O₃ formation on the diurnal 340 variation in PC-NO₂, and the smaller impact of chemical conversion from local NO₂ to O₃ 341 lowered R values during the day. Under MD conditions, both PC-NO₂ and SI-NO₂ exhibited 342 distinctive peaks in the morning with a degree of time lag; both subsequently declined toward 343 noon, and showed higher *R* values than those obtained under SD conditions. By contrast, under 344 MD conditions, correlations were enhanced due to a minimum around 1500 KST for both PC-345 NO₂ and SI-NO₂, despite time lags in both peaks in the morning and afternoon. 346

Previous studies of the Megacity Air Pollution Seoul (MAPS-Seoul) and KORUS-AQ 347 campaigns reported a typical pattern of continuously increasing PC-NO₂ over the Seoul 348 349 metropolitan area (Chong et al., 2018; Herman et. al., 2018). However, in the current campaign, we found similar results only under LD conditions. The diurnal patterns reported in 350 previous studies were mainly caused by the dominance of NO₂ emissions sources over NO₂ 351 352 losses (Chong et al., 2018; Herman et al., 2018) among several processes associated with NO₂ photochemical loss, including transport and deposition, which were also investigated in 353 specific cases in the current study. 354

In this study, we extended the correlation analysis, and investigated the correlation 355 between hourly PC-NO₂ and SI-NO₂ data. The results show a lower correlation in the morning, 356 and a higher correlation in the afternoon (Fig. S1). The respective median correlation 357 coefficients for the LD, MD, and SD meteorological conditions were -0.71, 0.18, and 0.22 in 358 the morning (0900–1200 LST), and 0.84, 0.77, and 0.79 in the afternoon (1200–1400 LST). 359 These values may reflect PBL development. SI-NO₂ decreases in the morning due to the rapid 360 361 growth of the PBL, while PC-NO₂ increases due to the accumulation of NO₂ in the atmosphere, deriving a lower correlation. However, there is very little change in the PBL in the afternoon, 362 363 and PC-NO₂ and SI-NO₂ show similar changes, yielding a positive correlation each other during the GMAP-2020 campaign. 364

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366 4.3 Aircraft measurements collected during GMAP-2020

Data collected via aircraft during GMAP-2020 are summarized in Table 2. A total of nine
aircraft measurements were conducted during the campaign period (November 12, 2020–
January 20, 2021). Four of nine flights were conducted under LD conditions, and the remaining
flights (except that on November 27, 2020) were conducted under MD conditions. No aircraft
measurements were consistent with SD conditions during the GMAP-2020 campaign.

We examined spiral segments from each flight over Seosan during 1100–1700KST to exclude marginal effects of diurnal variation in NO₂ (Fig. 2). The overall results indicated that the vertical O₃ profiles were relatively constant in the PBL, whereas NO₂ profiles appeared to be highly dependent on meteorological conditions. We compared data collected during flights conducted under LD (one flight) and MD conditions (two flights) during the GMAP-2020 campaign, to examine differences in the vertical structures of the PA and SI observations.

Aircraft measurements of vertical NO₂ and O₃ profiles for flights FL-5 (December 6) and FL-

379 6 (December 8) under LD conditions are shown in Fig. 7, along with 24 h backward trajectories

starting at different altitudes (100, 500, and 1,000 m). All observed NO₂ profiles shown in Fig. 380 7 appeared to have generally exponential curves, with anomalous features at higher altitudes. 381 For example, when vertical turbulent mixing prevailed within the PBL (O₃ profile, Fig. 7b), 382 the data were fitted with an exponential vertical curve, and the anomalous NO₂ layer aloft was 383 found to have a height of 1.5 km, which was higher than the estimated PBLH of 1.2 km. 384 HYSPLIT 24 h backward trajectories starting at 1200 KST showed that all air mass from the 385 386 surface to the lower free atmosphere was transported over the Yellow Sea via the Shandong Peninsula (Fig. 7c). This finding suggests that the anomalous NO₂ layer aloft was not produced 387 388 locally (i.e., from local LPS emissions), but instead traveled via long-range regional-scale transport. This transport of NO₂ across the region was also discussed and might be particularly 389 high during the winter when the NO_x lifetime is relatively longer (Stohl et al., 2002; Wenig et 390 391 al., 2003; Lee et al., 2013). According to Anmyeondo Lidar measurements for December 6 (http://kalion.kr), the anomalous NO₂ layer aloft corresponded well to an aerosol layer that 392 appeared at ~1.0 km at approximately 1200 KST, persisting until 2200 KST. However, based 393 on a cross-comparison of our data, high surface levels of SI-NO₂ (> ~4 ppb; Fig. 7a) were 394 influenced more by local LPS than by that in the atmosphere aloft due to long-range transport 395 (Fig. 7a). 396

Aircraft measurements for flight FL-7 (December 9) under LD conditions are shown in 397 Fig. 7b. The NO₂ vertical profile exhibited an exponential curve, with an anomalous peak at 398 399 ~600 m immediately above the top of the simulated PBL. HYSPLIT backward trajectory data starting at 1200 KST showed that the non-surface air had a different origin from the surface air 400 (Fig. 6d), indicating that the anomalous NO₂ plume likely traveled from coal-fired power plants 401 402 in a nearby industrial city (Taean) northwest of Seosan. This finding indicates a distinct vertical structure of higher NO₂ at the surface due to strong local emissions, whereas lower NO₂ levels 403 were observed at higher altitudes, with anomalously high NO₂ levels in some layers aloft due 404

to medium-range transport from nearby areas. Thus, despite the limited number of aircraft measurements, the elevated anomalous NO₂ structure that was observed intermittently led to a negative correlation between PA-NO₂ and SI-NO₂. The discrepancies imply that vertical profile distribution study should proceed cautiously when only surface measurements are obtained under LD meteorological conditions.

Aircraft measurements were conducted under MD conditions on flights FL-1 (November 410 411 26), FL-3 (November 28), and FL-8 (December 12) (Fig. 8). We applied several regression models (linear, exponential, and polynomial) to three vertical structures, and obtained two 412 413 distinct NO₂ vertical profile patterns from the surface to the PBLH: decreasing linearly for FL-1 and FL-8 (Fig. 8), and constant with altitude for FL-3 (Fig. 8b). None of the three cases 414 showed anomalous layers above the PBLH, similar to the exponentially declining profiles 415 416 obtained under LD conditions (Fig. 7). These vertical structures observed under MD conditions may have been induced by strong vertical mixing within the PBL, supplemented by prominent 417 surface photochemical losses at the same time. The vertical O₃ profile during FL-1 showed a 418 decoupled structure, with different patterns within and above the PBL (Fig. 8d); however, the 419 other 2 days showed uniform distributions, with no particular anomalous features between the 420 upper PBL and surface atmosphere (Fig. 8b, c, e, f). The observed daily maximum sensible 421 heat fluxes measured at Seosan (Fig. S3) were much higher for FL-3 (175.9 Wm⁻²) than FL-1 422 and FL-8 (118.9 and 102.0 Wm⁻²), suggesting that vertical turbulent mixing was much more 423 424 prominent during FL-3. These chemical and physical characteristics are all related to MD conditions. Thus, the higher coefficient of determination ($R^2 = 0.64$) obtained under MD 425 conditions (Fig. 5b) has an important bearing on the absence of irregular or anomalous layers 426 427 aloft, with little variation regardless of the shape of the curve (Figs. 7 and 8).

428

429 **4.4 Analyses of column–surface relationships for specific GMAP-2020 cases**

Figure 9 shows examples of PC-NO₂ and SI-NO₂ diurnal variation under LD (FL-5 and FL-7) 430 and MD (FL-1 and FL-8) conditions, and Fig. 10 shows latitudinal mean distributions for FL-431 5 and FL-7, based on the aircraft measurement data shown in Figs. 7 and 8. PC-NO₂ was 432 decoupled from SI-NO₂ on 2 days, FL-5 and FL-7, which were both classified as having LD 433 conditions (Fig. 9a, b), whereas good vertical mixing and uniform NO₂ distribution were 434 observed on the remaining 2 days, FL-1 and FL-8, which showed MD conditions (Fig. 9c, d). 435 436 According to our analysis of the aircraft measurements (Fig. 7), the poor correlations between PC-NO₂ and SI-NO₂ captured by FL-5 and FL-7 were mainly due to an NO₂ polluted layer 437 438 transported aloft, as described in Section 4.3.

439

440 **4.4.1 LD conditions**

Several cases showed poor correlations between PC-NO2 and SI-NO2 under LD conditions 441 within the study period. When we examined the results of previous studies (Thompson et al., 442 2019; Chong et al., 2018; Herman et al., 2018; Kim et al., 2021), we first considered the 443 possibility that LPS emissions influenced downwind regions under LD conditions, because the 444 increase in PC-NO₂, but not SI-NO₂, may have required an additional source of NO₂ apart from 445 early afternoon traffic emissions. The FL-5 data for December 6 represent an example of this, 446 showing a poor correlation between PC-NO₂ and SI-NO₂ ($R^2 = 0.06$; Fig. 9a). On the same day, 447 Anmyeondo LIDAR detected two elevated aerosol layers at 1200 and 1600-2200 KST 448 (http://kalion.kr); the first aerosol layer may reflect a PC-NO₂ peak, as shown in Fig. 9a. The 449 HYSPLIT backward trajectories, starting at different altitudes from the surface to the lower 450 troposphere, revealed that all air parcels moved eastward from China to Anmyeondo and 451 Seosan (Figure 1); thus, other NO₂ plumes may have begun to pass over Seosan at 1600KST 452 (Fig. 7c). Longitudinal SI-NO₂ distributions (Fig. 10) exhibited 5.2 ppb at 126.1°E, 8.1 ppb at 453 126.3°E, and 7.3 ppb at 126.4°E, averaged between 1300 and 1600 KST by longitude (Table 454

S1), whereas they were nearly constant at a height of 500–600 m on December 6. Therefore,
westerly winds advected cleaner air from Padori (AQM₁) to Seosan at the surface, but not at a
height of 500–600 m, contributing to low SI-NO₂ levels in the afternoon (Fig. 9a).

Another example of a weak correlation was obtained by flight FL-7 (December 9), as 458 shown in Fig. 9b. Time series PC-NO₂ data exhibited several peaks during 1200–1400 KST 459 (Fig. 9b), whereas SI-NO₂ showed less temporal variation, resulting in a weak correlation (R =460 -0.24) compared to the overall daytime (1100-1700 KST) correlation ($R^2 = 0.53$; Fig. 5a). 461 Latitudinal NO₂ levels at high altitudes of ~600 m (Fig. 10b) gradually increased northward, 462 463 whereas surface NO₂ was minimal at the midpoint. For example, at high altitudes, the latitudinal mean NO₂ levels were 1.4 ppb (36.8°N), 4.1 ppb (36.9°N), and 5.1 ppb (37.0°N), 464 whereas the SI-NO₂ levels at the same sites were 18.0 ppb (36.8°N), 14.3 ppb (36.9°N), and 465 16.8 ppb (37.0°N), respectively, averaged during 1200–1400 KST by latitude (Table S1). This 466 finding is attributable to a prevailing north wind that transported NO₂ southward at high 467 altitudes, while simultaneously ventilating SI-NO₂ toward outer Seosan, resulting in the 468 development of several PC-NO₂ peaks. By contrast, SI-NO₂ decreased slowly (Figs. 9b and 469 10b). 470

471

472 **4.4.2 MD and SD conditions**

We obtained higher PC–SI correlation coefficients under MD and SD conditions than LD conditions (Figs. 5b and 9c, d). Under MD and SD conditions, diurnal variation in PC-NO₂ and SI-NO₂ showed simultaneous declines from early morning until noon (Fig. 6). Notably, PC-NO₂ showed a continuously decreasing trend, particularly during the morning hours, in the period of approximately 0900–1200 KST under both MD and SD conditions (Fig. 6b, c). These diurnal patterns of decreasing PC-NO₂ in the study area were opposite to those reported in previous studies (Chong *et al.*, 2018; Herman *et al.*, 2018) that observed increasing PC-NO₂ in large urban areas during the daytime, caused by higher NO₂ emissions even during
photochemical NO₂ losses to form O₃.

We hypothesized that decreasing PC-NO₂ can occur due to photochemical loss and 482 surface wind transport, which both intensify with increasing solar radiation in the morning. 483 Photochemically, NO₂ is converted into photochemical oxidants such as PAN, HNO₃, and 484 nitrate under sunlight, thereby disrupting the NOx-VOC-O₃ cycle. Concurrently, Wsfc 485 486 intensified due to thermal turbulence transport of NO₂ emissions away from Seosan during the day. Thus, PC-NO₂ decreases under MD conditions as a result of ventilation effects caused by 487 488 stronger wind speeds. There are two possible mechanisms for this: sea breeze penetration (because the study area is adjacent to the northern coast of the Taean Peninsula; Fig. 1) and 489 vigorous turbulent mixing (which leads to vertical mixing of surface NO₂ during PBL growth; 490 Sun et al., 2013). We investigated these factors in detail for specific cases. 491

Figure 11 shows the diurnal variation in selected meteorological and chemical variables 492 measured under MD (November 25) and SD conditions (December 14). Under MD conditions 493 (Fig. 11a-c), declines in PC-NO₂ and SI-NO₂ were observed toward noon. In particular, 494 decreasing PC-NO₂ was accompanied by increased Wsfc (Fig. 11b); therefore, we examined 495 GMAP-2020 campaign measurements of sea breeze penetration. Figure S2a shows diurnal 496 variation in observed air temperatures at site Met₁ and measured sea surface temperatures at 497 nearby site Met₂ (37.14°N, 126.01°E), located 55 km from PA₄. The thermal meteorological 498 499 observations were used to calculate SBI (+0.37), which was greater than +3 (the threshold for sea breeze occurrence; Brigges and Graves, 1962). Sea breeze disturbances with a sharp 500 decrease (increase) in temperature (humidity) were observed at site Met₃ (Fig. S3b), which is 501 502 located on the northern coastline of Taean Peninsula (Fig. 1). However, sea breezes did not progress inland at the Met₁ Seosan Meteorological Automated Surface Observing System 503 (ASOS) site, which is closer to the Pandora sites; sea breezes did not correlate with NO₂ 504

505 ventilation to offset its high emission.

We further detected a strong positive correlation between wind speed and sensible heat 506 flux (Fig. 11b). We speculated that thermal and momentum turbulences caused by a vertical 507 temperature gradient and surface friction entrained surface turbulence, thus increasing 508 momentum in the free atmosphere downward to the surface due to strong turbulent mixing 509 within the PBL, in turn leading to a uniform vertical NO₂ profile with a positive correlation 510 511 between PC-NO₂ and SI-NO₂. Figure S3 shows a comparison of daily maximum sensible heat and momentum fluxes under LD, MD, and SD conditions during the GMAP-2020 campaign. 512 513 SD conditions showed the highest mean heat flux, followed by MD and LD, indicating that downward momentum transport led by both heat and momentum fluxes plays a greater role in 514 Wsfc enhancement under MD than LD conditions within the PBL. 515

516 Photolytic NO₂ loss was detected as temporal variation in NO₂, NO₃⁻, and CO at PA₄. Because no NO₂ analyzer was installed at PA₄, NO₂^{*}(= NO_y-NO) was used instead of NO₂ 517 under the assumption that NO_z is negligible in winter. Figure 11c shows the diurnal variation 518 in NO₂, O₃, and NO₃⁻ under MD conditions, normalized by CO to reduce the effect of PBL 519 evolution. The results showed that NO₂/CO decreased after the morning peak; however, NO₃⁻ 520 /CO and O₃/CO increased toward midday, indicating that photolytic activity also contributed 521 considerably to the concurrent decline of SI-NO2 and PC-NO2 (Fig. 11a). In turn, this indicated 522 that photochemistry can contribute to higher correlation coefficients under MD conditions. 523

524 Under SD conditions (Fig. 11d–f), PA-NO₂ and SI-NO₂ exhibited weak diurnal variability 525 compared to LD and MD conditions. SD conditions on December 14 produced significantly 526 stronger winds (i.e., wind speed > 6 m s⁻¹ at 1300 KST), with generally higher PBLHs (Fig. 527 11e). Meteorological features, such as strong wind at both 850 hPa (18.0 m s⁻¹) and 10 m height 528 (4.26 m s⁻¹), suppressed both PC-NO₂ and SI-NO₂ (7.3 ppb and 0.31 DU, respectively) to 529 below the average, producing a strong correlation (R = 0.9 at AQM₅) and nearly flattening their temporal curves during the day (Fig. 11d). Thus, under SD conditions, wind speed and turbulent fluxes such as sensible heat flux had larger values, and NO_2 and NO_3^- decreased or increased at the same time during the day (Fig. 11f), indicating that the transport effect was much greater than that of local photochemical loss over the study area.

In conclusion, in this case-specific study, we assessed the correlations between PC-NO₂ 534 and SI-NO₂, and explored their mechanisms by investigating the impact of meteorological and 535 536 photochemical conditions. A weak correlation between PC-NO2 and SI-NO2 occurred when anomalously high concentrations remained, with ragged fragments of NO₂ plumes in the upper 537 538 or middle layers. We also found that a negative correlation occurred intermittently under LD conditions, with generally lower PBLH. In particular, elevated pollutant levels due to regional-539 scale transport or decoupled NO₂ plumes advected within the PBL may have also caused the 540 541 weak correlation between PC-NO₂ vs. SI-NO₂. These phenomena were detected only from the PA-SI coupled measurements in this study. Thus, when either PC or SI observations are 542 applied alone for understanding the vertical structure of air pollutants, undetected bias can 543 occur under LD conditions, particularly where transport processes prevail, and these results 544 can be also applicable to GEMS observations analysis. 545

546

547 **5. Conclusion**

We explored the potential applicability of combined PC-NO₂ and SI-NO₂ measurements collected at Seosan during the GMAP-2020 campaign. We characterized the correlation between PC-NO₂ and SI-NO₂ under various conditions to understand the complex air quality of Seosan, which appears to be vulnerable to LPS emissions from surrounding areas. We hypothesize that correlations between PC-NO₂ and SI-NO₂ are closely related to NO₂ vertical profiles, which also depend on meteorological conditions. We performed statistical analyses of a year-long PC-NO₂ dataset (November 12, 2020–October 30, 2021) combined with 555 meteorological data, *in situ* ground data, and airborne chemical data measured during the 556 GMAP-2020 campaign in the same period.

557 Our results show that hourly PC-NO₂ and SI-NO₂ over the 1-year period exhibited a linear 558 relationship with a fair correlation (R = 0.41), and daily mean PC-NO₂ vs. SI-NO₂ exhibited a 559 good linear correlation (R = 0.73), supporting the overall uniformity of NO₂ profiles in the PBL 560 over Seosan despite the continuous impact of LPS emissions.

561 The impact of meteorological conditions on the relationship between PC-NO₂ and SI-NO₂ was investigated through agglomerative hierarchical clustering, which indicated three 562 563 meteorological conditions: LD, MD, and SD. Under LD conditions, southerly winds advect warm air under the upper ridge, forming stable and short PBLs and weak surface winds. By 564 contrast, under SD conditions, cold northerly winds induce unstable and high PBLs with strong 565 surface winds. The correlations between daily mean PC-NO2 and SI-NO2 levels, and their 566 variation during 1100–1700 KST, weakened under LD conditions, suggesting that the shape of 567 the NO₂ profile typically deviates from a uniform profile under SD and MD conditions. Aircraft 568 measurements under LD conditions demonstrated NO₂ plumes aloft, with anomalous vertical 569 structures and different horizontal (latitudinal) gradients of surface NO₂ at higher altitudes, 570 such as 600 m over Seosan. 571

Thus, the relationship between PC-NO₂ and SI-NO₂ depends on the presence of NO₂ 572 plumes aloft under LD conditions, which provide a favorable environment for LPS plumes 573 574 decoupled from the surface at Seosan. Our findings suggest that the correlation between PC-NO₂ and SI-NO₂ may serve as an indicator of the degree of complexity of urban air quality. 575 This correlation can be optimally applied for air quality evaluation and vertical analysis by 576 577 combining the Pandora Asia Network with AQM networks, and the results can be also applied to environmental GEMS observation analysis in combination with SI observations. More 578 detailed studies on urban air pollution evaluation will be undertaken based on PC, DOAS, 579

580	aircraft, SI air quality, and surface turbulence observation data, as well as modeling studies of
581	data collected during the GMAP-2021 campaign.
582	
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591	
592	Data Availability.
593	The measurements can be accessed by contacting the corresponding authors.
594	
595	Competing interest.
596	The authors declare that they have no conflict of interest.
597	
598	Author Contributions.
599	Lim-Seok Chang: Conceptualization, Formal analysis, Visualization, Investigation, Writing -
600	Original draft; Donghee Kim, Hyunkee Hong, Deok-Rae Kim, Jeonga Yu, and Daewon Kim; Data
601	curation; Hanlim Lee, Kwangyul Lee, and Jinkyu Hong: Methodology and formal analysis; Hyun-
602	Young Jo: Formal analysis and Visualization; Cheol-Hee Kim ; Writing—original draft preparation,
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736	Table 1. Summary of NO ₂ column data from four Pandora (PA) measurement sites.

	Site name	Site location						Number of	
Site		Longitude (°E)	Latitude (°N)	Mean (DU)	SD (DU)	Minimum (DU)	Maximum (DU)	data points (days)	Operating period
PA ₁	Seosan-DHJ	126.502	36.900	0.50	0.22	0.20	1.60	838 (11)	GMAP-2020 campaign
PA ₂	Seosan-DM	126.458	36.778	0.43	0.19	0.18	1.62	1241 (13)	GMAP-2020 campaign
PA ₃	Seosan-CC	126.449	36.785	0.40	0.14	0.18	0.97	1242 (13)	GMAP-2020 campaign
PA ₄	Seosan-SS	127.492	36.777	0.39	0.16	0.17	1.79	8753 (141)*	1 year (Nov. 12, 2020–Oct. 30, 2021)

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* The Pandonia Global Network (PGN) retrieval algorithm was applied to yearly measurements.

744	Table 2. Summary of aircraft measurements collected during the Geostationary Environment
745	Monitoring Spectrometer (GEMS) Map of Air Pollution (GMAP)-2020 campaign period
746	(November 12, 2020–January 20, 2021).

Flight no. Date		Meteorological classification		
FL-1	Nov. 26, 2020	MD ¹⁾		
FL-2	Nov. 27, 2020	No Pandora measurements		
FL-3	Nov. 28, 2020	MD		
FL-4	Dec. 1, 2020	LD ²⁾		
FL-5	Dec. 6, 2020	LD		
FL-6	Dec. 8, 2020	LD		
FL-7	Dec. 9, 2020	LD		
FL-8	Dec. 12, 2020 (am)	MD		
FL-9	Dec. 12, 2020 (pm)	MD		

747 ¹⁾ LD: local wind-dominant conditions; ²⁾ MD: mixed conditions.

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Figure Captions

753 Figure 1. Map of sites used for Geostationary Environment Monitoring Spectrometer (GEMS) Map of 754 Air Pollution (GMAP) campaigns conducted in (left) Seosan, South Korea in November 2020 to January 2021 (GMAP-2020), and (right) the Seoul metropolitan area from October 2021 to November 755 2021 (GMAP-2021). (Left) Measurement sites around Seosan, the study area for the GMAP-2020 756 757 campaign. Red circles indicate Pandora column measurement sites including (left) Seosan Daehoji (PA₁), Seosan Dongmun (PA₂), Seosan City Council (PA₃), and Seosan Super Site (PA₄). Blue triangles 758 759 indicate large point sources (LPSs) including the Taean and Dangjin thermal power stations (LPS₁ and 760 LPS₂, respectively), Hyundai steelworks (LPS₃), and Daesan petrochemical complex (LPS₄). Yellow 761 squares indicate Automated Synoptic Observing System meteorological sites in Seosan (Met₁), AWS 762 (Met₂), and buoy (Met₃). Green squares indicate air quality monitoring (AQM) network stations 763 including Padori (AQM1), Leewon (AQM2), Taean (AQM3), Daesan (AQM4), Seongyeon (AQM5), and Dongmoon (AQM₆). In the right panel, the black line indicates the route used for car-based differential 764 765 optical absorption spectroscopy (Car-DOAS) measurements and the blue dotted line indicates the 766 horizontal domain of Geo-CAPE airborne simulator (GCAS) measurements taken during the GMAP-767 2021 campaign.

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Figure 2. Flight tracks for two Cessna Grand Caravan 208 B aircraft over Pandora sites (left)

PA₄ and (right) PA₁ during the GMAP-2020 campaign. Colored circles indicate airborne NO₂

concentration observations. Stacked circles indicate spiral flights conducted over two sites.

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Figure 3. (a) Pandora column (PC) NO₂ measurements as a function of surface *in situ* (SI) NO₂
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Figure 4. K-means clustering yielded three groups of cases for (a) surface NO₂ and (b) PCNO₂, associated with eight meteorological variables: (c) surface wind speed (Wsfc), (d) Psfc,
(e) Psfc tendency (dPsfc/dt), (f) 925-hPa air temperature (T925), (g) 850-hPa wind speed
(W850), (h) 850-hPa north–south wind component (NS850), (i) 850-hPa east–west wind
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Figure 5. (a) Scatterplots of daytime measurements at site PA_4 (a) PC-NO₂ vs. SI-NO₂ under all meteorological conditions and (b) PC-NO₂ vs. Surface $\triangle NO_2$ in each meteorological condition over a 1-year period (November 12, 2020–October 30, 2021). Here Surface $\triangle NO_2$ = SI-NO₂–(30-day moving average) SI-NO₂.

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Figure 7. Box and whisker plots of the vertical NO₂ and O₃ profiles measured by GMAP aircraft superposed with *in situ* AQMS₁ measurements during flights (a, b) FL-5 (December 6) and (d, e) FL-7 (December 6 and 9). Blue dashed lines are linear regression lines fitted to NO₂ and O₃ profiles within the planetary boundary layer (PBL). Black arrows indicate the simulated PBL height (PBLH) obtained from the Korea Meteorological Administration (KMA). HSYPLIT 24-h backward trajectories in Seosan are shown at altitudes of 100, 500, and 1,000 m, starting at 1600 KST on November 26 and 1200 KST on December 12.

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with *in situ* AQMS measurements for (1) NO₂ and (2) O₃ for flights (a) FL-1 (November 26),
(b) FL-3 (November 28), and (c) FL-8 (December 12). Blue dashed lines are linear regression
lines fitted to NO₂ and O₃ in the PBL. Black arrows indicate PBLH simulated by the Hybrid
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- Figure 9. Time series and scatterplots of PC-NO₂ and SI-NO₂ at PA₂ on (a) December 6, (b)
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- on December 6 (blue), December 9 (red), November 26 (gray), and December 12 (black). (f)
- 815 Vertical potential temperature profiles on December 6, 9, and 12, 2020. Radiosonde data for
- 816 November 26, 2020 are missing.

- Figure 10. Latitudinal NO₂ distribution at the surface and 600 m over PA₄ (Seosan Super Site),
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 KST on December 9 (FL-7) by latitude, obtained from airborne (blue) and surface
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- 824 f). (a, b) Column NO₂ at sites PA_1 – PA_4 and surface NO₂ at air quality monitoring sites AQM₄
- and AQM₆. (c, d) Sensible heat fluxes and surface wind speed at PA₄. (e, f) Diurnal variation in NO₂, NO₂⁻, and O₃ normalized by CO. Figure 1 shows the locations of the measurement sites.

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