

1 **Diurnal variations of NO₂ tropospheric vertical column density over the Seoul**
2 **Metropolitan Area from the Geostationary Environment Monitoring**
3 **Spectrometer (GEMS): seasonal differences and the influence of the *a priori* NO₂**
4 **profile**

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6 Seunghwan Seo¹, Si-Wan Kim^{1,2*}, Kyoung-Min Kim¹, Andreas Richter³, Kezia Lange³,
7 John P. Burrows³, Junsung Park^{4**}, Hyunkee Hong⁵, Hanlim Lee⁴, Ukkyo Jeong⁴, Jung-
8 Hun Woo^{6***}, and Jhoon Kim^{1*}

9

10 ¹Department of Atmospheric Sciences, Yonsei University, Seoul, Republic of Korea

11 ²Irreversible Climate Change Research Center, Yonsei University, Seoul, Republic of
12 Korea

13 ³Institute of Environmental Physics, University of Bremen, Bremen, Germany

14 ⁴Division of Earth Environmental System Science, Major of Spatial Information
15 Engineering, Pukyong National University, Busan, Republic of Korea

16 ⁵National Institute of Environmental Research, Incheon, Republic of Korea

17 ⁶Department of Technology Fusion Engineering, College of Engineering, Konkuk
18 University, Seoul, Republic of Korea

19

20 *To whom correspondence should be addressed:

21 Si-Wan Kim (e-mail: siwan.kim@yonsei.ac.kr) and Jhoon Kim (e-mail:
22 jkim2@yonsei.ac.kr).

23 ** now at: Center for Astrophysics | Harvard & Smithsonian, Cambridge, MA, USA

1 ***now at: Graduate School of Environmental Studies, Seoul National University, Seoul,

2 Republic of Korea

3 Date: 10/13/2024

4

1 **Abstract**

2 The Geostationary Environment Monitoring Spectrometer (GEMS), launched in 2020,
3 provides both temporally and spatially continuous air quality data from geostationary
4 Earth orbit (GEO). This study first investigates the seasonal variations and diurnal
5 behavior of nitrogen dioxide (NO₂) tropospheric vertical column densities (TropVCDs)
6 over the Seoul Metropolitan Area (SMA) using GEMS data, retrieved by the IUP-UB
7 algorithm. We find that the magnitude of the NO₂ TropVCDs and its diurnal behavior
8 have significant seasonal dependences. In January, the highest NO₂ TropVCD values in
9 the range $27.5 - 28.9 \times 10^{15}$ molec. cm⁻² during the four seasons were observed at 15:00
10 local time (LT), and NO₂ TropVCD increases from the first retrieved values at 10:00
11 LT. On the other hand, we find the lowest values ($7.4 - 8.8 \times 10^{15}$ molec. cm⁻²) are at
12 ~14:00 LT in July. The VCD values in July increased up to 10:00 LT, then decreased
13 until 14:00 LT, but then began to increase again. These different diurnal behaviors of
14 the TropVCDs in the different seasons reflect the differences in photochemical and
15 meteorological conditions as well as the emissions of NO_x. Photochemical
16 transformations are typically more rapid in July and slower in January. The absolute
17 values and diurnal behavior of NO₂ TropVCDs are significantly influenced by the wind
18 speed, except in July. Moderate (wind speed ≥ 3 m/s) or strong wind (wind speed > 5 m/s)
19 reduced the magnitude of the diurnal behavior in January, implying that the NO₂ plumes
20 were transported downwind. Finally, we investigate the retrieved NO₂ TropVCDs with
21 that retrieved using different *a priori* NO₂ data simulated by TM5 and WRF-Chem,
22 calculated using the most recent emission inventories. Although simulated VCDs from
23 WRF-Chem and TM5 show differences of up to a factor 2.75, retrieved NO₂ TropVCDs
24 using each *a priori* data have almost identical values and diurnal behaviors, except in
25 July. Notably, the diurnal behavior of the retrieved NO₂ TropVCDs are independent of
26 those from the two chemical transport models, indicating that observations of slant
27 column densities are the dominant factor in determining the diurnal behavior of NO₂
28 TropVCDs. Changes of the model horizontal resolution and volatile organic compounds
29 (VOC) emission inventory do not affect significantly the retrieved NO₂ TropVCDs in

1 this study. However, when the *a priori* NO₂ vertical profile was fixed as the values at
2 13:45 LT, the diurnal patterns of NO₂ TropVCDs showed significant changes with
3 differences of up to -18.3%.

1 **1. Introduction**

2 Nitrogen dioxide (NO₂) is one of the most important trace gases in the photochemical
3 mechanisms, which determine the tropospheric distributions of ozone and secondary
4 aerosol (Milford et al., 1989). Beginning with the launch of the passive remote sensing
5 instrument GOME on ESA ERS-2 (Burrows et al., 1999) in 1995, then followed by
6 SCIAMACHY on ESA Envisat in 2002 (Burrows et al., 1995 and Bovensmann et al.,
7 1999), OMI on NASA AURA (Levelt et al., 2006), GOME-2 on ESA EUMETSAT
8 Metop A, B and C (Callies et al., 2000, Munro et al., 2016), and TROPOMI on the ESA
9 Sentinel 5 Precursor in 2018 (Veefkind et al., 2012), the amounts and distributions of
10 stratospheric and tropospheric NO₂ vertical column densities (TropVCDs) have been
11 retrieved at increasing spatial resolutions from these instruments, which all fly in sun-
12 synchronous low earth orbit (LEO). By using the retrieved NO₂ TropVCDs from the
13 LEO instruments, the tropospheric nitrogen oxide sources have been identified and their
14 NO_x emissions have been estimated, and the chemistry of the troposphere has been
15 studied from the local to the global scale. While instruments on board LEO satellites
16 provide spatially continuous data, observations are obtained only once or twice per day.
17 It was recognized in the late 1990s that instruments similar to SCIAMACHY in
18 geostationary orbit (GEO) would potentially deliver the diurnal variations of key trace
19 gases (see the GeoTROPE concept in Burrows et al., 2004 and references therein). The
20 measurements at the top of the atmosphere of the Geostationary Environment
21 Monitoring Spectrometer (GEMS), launched in 2020, yield the first not only spatially
22 but also temporally continuous air quality data over Asia from the geostationary orbit
23 GEO (see Kim et al., 2020).

24 Mathematical inversion of the GEMS observations provides diurnal variations
25 of the NO₂ TropVCD. These data products enable the seasonal changes not only in
26 pollutant concentration but also in temporal characteristics, such as the times of the
27 maxima and minima and the sources and sinks of NO₂, which vary by diurnally and

1 seasonally, to be studied for the first time from space.

2 As part of the differential optical absorption spectroscopy (DOAS) retrieval of
3 NO₂ TropVCD data, air mass factors (AMF) are used to convert slant column density
4 (SCD) to VCD. The assumptions used in the AMF calculation are explained in
5 Richter and Burrows (2002) and Palmer et al (2001). In agreement with other studies,
6 Lorente et al. (2017) reported that the AMF calculation is the largest source of error or
7 uncertainty in NO₂ satellite retrievals. This is because of the assumption used to
8 determine the ancillary or prior data used in the AMF calculation, such as surface albedo,
9 terrain height, cloud parameters, and trace gas profiles. Consequently, the selection of
10 optimal and appropriate *a priori* data is essential to accurately retrieve NO₂ TropVCDs
11 from the observations of any nadir-sounding satellite spectrometer. This is in addition
12 to the need to separate upper atmospheric NO₂ from that in the troposphere.

13 In this study we investigate two important issues using the GEMS NO₂
14 TropVCD data over the Seoul Metropolitan Area (SMA): (1) the influence of *a priori*
15 profiles on the retrieved GEMS NO₂ TropVCDs and (2) the seasonal variation of the
16 GEMS NO₂ TropVCD. In section 2 we describe the methods and data used.

17 Prior to our geophysical interpretation of the NO₂ TropVCD, in Section 3 we
18 compared three GEMS datasets, retrieved with different *a priori* data from the WRF-
19 Chem model. Thereby we investigated the influence of the inventories of the emissions
20 of NO_x, defined as the sum of nitrogen monoxide (NO) and nitrogen dioxide (NO₂) in
21 an air mass, on the simulated and retrieved NO₂ TropVCD.

22 In Section 4, we utilized two chemical transport models (CTM), the Weather
23 Research and Forecast model combined with Chemistry (WRF-Chem) and the global
24 chemistry transport model TM5 (Tracer Model 5) to analyze both the seasonal
25 variations and the influence of *a priori* NO₂ profiles. The seasonal changes in the
26 magnitudes and the time of the maxima of the diurnal NO₂ TropVCD, which we define
27 as the peak times, were investigated. The differences in the spatial distributions of NO₂

1 TropVCD between the WRF-Chem- and TM5-based GEMS datasets using different *a*
2 *priori* data, were identified for each season and peak time. We also analyzed the
3 influence of wind speed on the variations in the magnitude and diurnal behavior of the
4 retrieved NO₂ TropVCDs.

6 **2. Data and methods**

7 **2.1. GEMS products**

8 GEMS is an ultraviolet-visible (UV-VIS) instrument, measuring contiguously the
9 spectral range from 300 to 500 nm at a spectral resolution of ~ 0.6 nm (Kim et al.,
10 2020). The nominal spatial resolution is 3.5 km × 7.7 km for gases including NO₂ data
11 products. The overall field of regard (FOR) of GEMS covers 75° – 145°E longitude and
12 5°S – 45°N latitude. GEMS measures hourly during the daytime. The number of
13 observations varies depending on the month, as a result of the length of the day and the
14 measurement strategy. For South Korea, observations are least frequent in January, with
15 six observations per day, and most frequent from April to September, with ten
16 observations per day. We utilized GEMS NO₂ TropVCD data with the IUP-UB
17 algorithm (GEMS IUP-UB products) in January, April, July, and October 2021 –
18 detailed explanations of GEMS IUP-UB products are shown in Section 2.1.1.

20 2.1.1. GEMS IUP-UB products v1.0

21 The GEMS NO₂ vertical columns used in this study are from the scientific data product
22 of the University of Bremen, version 1.0 (Lange et al., 2024). NO₂ slant columns are
23 retrieved in the large fitting window 405 – 485nm to reduce noise. In addition to the
24 cross-sections of other absorbing species (O₃, O₄, H₂O and liquid water) pseudo cross-
25 sections for the Ring effect, for GEMS instrument polarization sensitivity and the
26 effects of scene inhomogeneity are included. The stratospheric correction is performed

1 using the STRatospheric Estimation Algorithm from Mainz (STREAM) (Beirle et al.,
2 2016). Conversion to vertical tropospheric columns is based on look-up tables of
3 altitude dependent air mass factors calculated with the radiative transfer model
4 SCIATRAN (Rozanov et al., 2014) using Lambertian equivalent reflectivity (LER)
5 surface reflection values from the TROPOMI climatology (Tilstra et al., 2023). To
6 apply the cloud correction, adjusted cloud fractions and pressure from the GEMS L2
7 cloud product were used. The NO₂ *a priori* data are different in the different model
8 simulations, which we call runs, as explained below.

9

10 **2.2. Experiment designs**

11 To analyze the spatiotemporal characteristics of GEMS NO₂ VCDs and the impacts of
12 different *a priori* data on the retrieved values, we undertook five experiments, called
13 TM5, CTRL, CONST, FINE, and MIXED.

14 The TM5 experiment applies the standard GEMS IUP-UB products v1.0,
15 which use the TM5 model, as their *a priori* data (Huijnen et al., 2010, Williams et al.,
16 2017). The meteorological data for TM5 simulations are obtained from the European
17 Centre for Medium-Range Weather Forecasts (ECMWF) operational forecast data. For
18 the anthropogenic NO_x emission inventory of TM5, the MACCity emission estimates
19 are adopted (Granier et al., 2011), which have no diurnal variation of NO_x emissions.
20 The outputs from TM5 model have a horizontal resolution of 1° × 1° and 34 vertical
21 layers.

22 For the other four numerical experiments (CTRL, CONST, FINE, and MIXED),
23 WRF-Chem version 4.4 was used to generate *a priori* data (Grell et al., 2005,
24 Skamarock et al., 2021). The chemistry scheme follows the Regional Atmospheric
25 Chemistry Mechanism (RACM) with Secondary Organic Aerosol-Volatility Basis Set
26 (SOA-VBS) option (chem_opt = 108) (Ahmadov et al., 2012). The horizontal

1 resolution of WRF-Chem simulation is 28 km × 28 km, except for the FINE run (12 km
2 × 12 km). All simulations have 59 customized vertical layers. To account for the
3 stratospheric vertical profiles, the Whole Atmosphere Community Climate Model
4 (WACCM) model outputs were combined with the WRF-Chem data
5 (ACOM/NCAR/UCAR, 2020, last access: 05 Dec 2022). The combined data comprises
6 a total of 113 vertical layers. Detailed model configuration is described in Kim et al.
7 (2024). For the anthropogenic emission inventories, the Air Quality in Northeast Asia
8 (AQNEA) emission inventory version 2 was adopted. Since the reference year of
9 AQNEA version 2 is 2019, the anthropogenic NO_x emissions decreased by 20% to
10 account for the decreasing trends of NO_x emissions from 2019 to 2021. We applied the
11 normalized diurnal variabilities of NO_x emissions obtained from the Los Angeles Basin
12 in Kim et al. (2016), but shifting the values one hour earlier (**Figure 1**). For the CONST
13 run, only the *a priori* profiles at 13:45 LT were used to retrieve the NO₂ TropVCD. To
14 investigate the impact on the volatile organic compounds (VOC) emissions of the
15 anthropogenic VOC emissions we used the KORUS emission inventory version 5 (Jang
16 et al., 2020, Woo et al., 2012) in the MIXED run. We retrieved four months (January,
17 April, July, and October 2021) for the TM5 and CTRL runs, and one month (July 2021)
18 for the other runs. The experimental designs are summarized in **Table 1**.

19

20 **3. Impacts of different *a priori* data on the retrieved NO₂ TropVCDs**

21 We compared retrieved NO₂ TropVCDs from the five different simulations, or runs, to
22 study the impacts of *a priori* data used in AMF calculations on the retrieved NO₂
23 TropVCD.

24 **3.1. Comparison between the CTRL and TM5 runs**

25 Retrieved NO₂ TropVCDs from the CTRL and TM5 runs exhibit similar diurnal
26 patterns, which are independent of the diurnal patterns of their respective *a priori* data

1 (Figure 2). This suggests that the observed slant column density (SCD) plays a more
2 decisive role in the diurnal pattern of TropVCD than the influence of *a priori* used to
3 determine the AMF. Nevertheless, differences in NO₂ TropVCDs between the two runs
4 were observed, and are particularly noticeable differences in July.

5 **Figure 3** displays spatial distributions of AMF differences between the CTRL
6 and TM5 runs in January, April, July, and October 2021. In urban areas, the AMF in the
7 CTRL run was generally lower (blue) than in the TM5 run, but higher values (red) were
8 observed in the northern and eastern regions of Seoul. As a result, the average values
9 across the SMA domain were similar between CTRL and TM5 – the diurnal patterns of
10 averaged air mass factor over the SMA are shown in **Figure 4**. In July, however, lower
11 values in the CTRL run were observed throughout Seoul and its surrounding areas,
12 leading to lower average AMF values for the SMA region during most of the day. As a
13 result, the TropVCD values in July were higher in the CTRL run (Figure 2c).

14 In **Figure 5**, we compare NO₂ vertical profiles at 08, 10, 12, 14, and 16 LT from
15 the CTRL and TM5 runs. NO₂ values in the lower atmosphere in the CTRL run are
16 much higher than those in the TM5 run in July, which lead to lower AMF and thus
17 higher NO₂ TropVCD.

18
19 **3.2. Comparisons between the CTRL and CONST, FINE, and MIXED runs**

20 In **Figure 6**, the diurnal patterns of retrieved and *a priori* NO₂ TropVCDs in July 2021
21 over the SMA region from the CTRL run and the CONST, FINE, and MIXED runs, are
22 shown. Despite some changes in model resolution and VOC emissions, the FINE and
23 MIXED runs did not show significant differences compared to the CTRL run. In
24 particular, the MIXED run resulted in almost no difference in the *a priori* TropVCD ,
25 resulting in nearly identical retrieved NO₂ TropVCD.

26 On the other hand, the CONST run, which used only the *a priori* vertical profile

1 from 13:45 LT in the retrieval process, exhibited clear differences to the CTRL run.
2 Specifically it had lower values than the CTRL run before ~14:00 LT, but higher values
3 after. These differences are explained by comparisons of vertical profiles from each run,
4 which are displayed in **Figure 7**. The vertical profile shapes of the CTRL, FINE, and
5 MIXED runs are identical, indicating that AMF of each runs have similar values. On
6 the other hand, clear differences of vertical profile shape are apparent between the
7 CTRL and CONST runs. Before 14:00 LT, the CTRL run showed lower sensitivity in
8 the upper layers compared to the CONST run. This indicates a smaller AMF and thus
9 higher VCD values. In contrast, after 14:00 LT, the CTRL run exhibited higher
10 sensitivity to NO₂ in the upper layers of the troposphere, leading to a larger AMF and
11 consequently lower VCD values compared to the CONST run. These differences in the
12 vertical profile arise from effects such as the development of the mixing layer and
13 variations in emissions throughout the day. This implies that providing optimal time-
14 dependent *a priori* data for the AMF calculation will improve the accuracy of the
15 retrieved NO₂ TropVCD.

16

17 **4. Spatiotemporal characteristics of GEMS NO₂ TropVCD**

18 We report on our investigation of the spatiotemporal characteristics of GEMS NO₂
19 TropVCD. We use the retrieved NO₂ TropVCD and those simulated by the TM5 and
20 CTRL runs to assess two geophysically important influences on the NO₂ TropVCD the
21 SMA region (126.5 – 127.3°E, 37.2 – 37.8°N) in 2021: (1) the identification,
22 quantification and origin of the seasonal changes; and (2) advection and convection of
23 air masses.

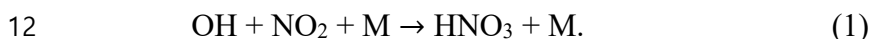
24 **4.1. Seasonal variations**

25 **Figure 2** displays diurnal patterns of retrieved and *a priori* NO₂ TropVCDs during
26 weekdays in January, April, July, and October 2021 over the SMA region from the TM5

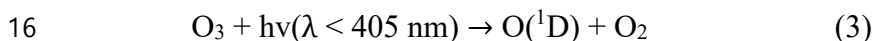
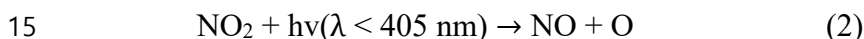
1 and CTRL runs. The scenes with wind speed faster than 3m/s are excluded to remove
2 the transport impacts. The effects of transport on NO₂ columns are analyzed in **Section**
3 **4.2.**

4 In January, NO₂ TropVCDs continuously increase from 10:00 local time (LT)
5 to 15:00 LT. During the winter, NO₂ in the urban region accumulates particularly in the
6 boundary layer. Qualitatively, this is explained as follows.

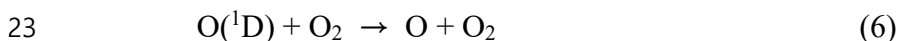
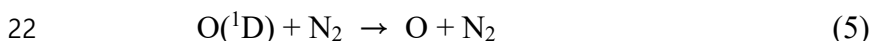
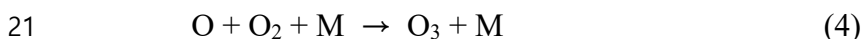
7 As tropospheric solar UV radiation is low in winter and the atmosphere is cold,
8 photolysis frequencies are small. Similarly, the rate coefficients of many reactions are
9 smaller at the lower winter temperatures compared to those of the other seasons. In
10 winter, the relatively slow loss of NO_x occurs through the three body reaction of
11 hydroxyl (OH) with NO₂ to form nitric acid (HNO₃):

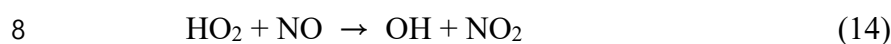
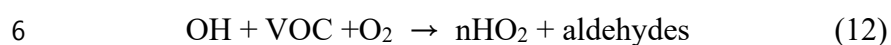
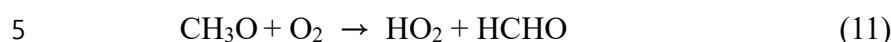
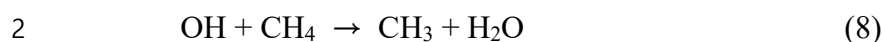


13 The smaller photolysis frequencies of reactions following photoexcitation in the
14 reactions:



17 lead to slower production of i) the first excited state of oxygen (O(¹D)) from the
18 photolysis of ozone (O₃), ii) the hydroxyl radical (OH), and iii) the production of
19 organic peroxy radicals (RO₂), and hydroperoxy (HO₂) through the oxidation of
20 methane (CH₄) and VOC. Some of the following reactions are involved:





9

10 Overall at low solar insolation, the low levels of actinic radiation result in
11 smaller amounts of OH and HO₂. The oxidation process is slow and HO₂ and OH
12 chemistry are coupled with NO_x chemistry and controlled by rate of oxidation of VOC
13 and CH₄ and the rate of HO₂ to OH through the rate of reaction (14) and the rate of loss
14 of HO_x and NO_x for example through reaction (1).

15 In January the maximum values of retrieved TropVCDs are 27.5×10^{15} molec.
16 cm^{-2} (TM5) and 28.9×10^{15} molec. cm^{-2} (CTRL) at 15:00 LT, whereas the *a priori* NO₂
17 TropVCDs have maxima of 11.2×10^{15} molec. cm^{-2} (TM5) and 21.9×10^{15} molec. cm^{-2}
18 (CTRL) at the same time. These higher values of retrieved NO₂ TropVCDs relative to
19 the model NO₂ TropVCDs are explained by the following inadequate knowledge of the
20 bottom-up diurnal NO_x emissions in January and/or the dilution during the transport of
21 plumes, which is dependent on the model horizontal resolution.

1 For other months, the maxima of NO₂ TropVCDs occur at earlier times of the
2 day in April at 12:00 LT, in July at 10:00 LT and in October at 11:00 LT. There is also
3 a second maximum at 15:00 LT in October. The behavior of NO₂ TropVCD in April,
4 July and October, when compared to that in January, is explained by the following
5 effects: i) faster tropospheric photolysis frequencies, as a result of higher levels of
6 tropospheric solar insolation and actinic radiation accelerating the photochemical
7 oxidation of CH₄ and VOC in April, July and October compared to January; ii)
8 generally faster reaction rate coefficients of the free radical reactions at the higher
9 temperatures, the rate coefficient of reaction (4) being an exception; iii) the different
10 diurnal emissions of NO_x compared to those in January. **Figure 8** shows the diurnal
11 variations of OH concentrations averaged across boundary layer height in each month,
12 calculated by the CTRL model run. The OH concentration in January is about an order
13 of magnitude smaller than that in July.

14 In April, NO₂ TropVCDs increased until 12:00 LT. It then maintains similar
15 levels until 17:00 LT. The maximum NO₂ TropVCD occurred at 12:00 LT for the CTRL
16 run (21.4×10^{15} molec. cm⁻²). The maximum NO₂ TropVCD for the TM5 run appeared
17 at 17:00 LT, being 21.9×10^{15} molec. cm⁻². However, the retrieved NO₂ TropVCDs
18 from the TM5 and CTRL runs have almost identical behavior up to 15:00 LT. There is
19 a difference of 1.6×10^{15} molec. cm⁻² (8.1%) between the two runs at 17:00 LT, when
20 *a priori* NO₂ TropVCD value sharply increased from the CTRL run.

21 In July, both the TM5 (12.2×10^{15} molec. cm⁻²) and CTRL (13.9×10^{15} molec.
22 cm⁻²) runs show maxima at 10:00 LT, i.e. the earliest for the four months investigated.
23 After the peak, NO₂ TropVCDs decrease, most likely due to more rapid photochemical
24 loss processes e.g. reaction (1) until 14:00 LT, and then increase. In other seasons, the
25 minimum values were observed in the morning. However, in July, the minimum
26 occurred at 14:00 LT. This unique pattern of behavior is explained by the more rapid
27 photochemical production and removal reactions in summer. We infer that the chemical

1 removal becomes relatively more rapid than the emission and production of NO₂ (see
2 Figure 8). The two types of run show similar diurnal behavior, but the retrieved NO₂
3 TropVCD of the CTRL runs between 10:00 and 14:00 LT rise to 2.1×10^{15} molec. cm⁻²
4 i.e. higher than those of the TM5 runs. The diurnal change of *a priori* NO₂ TropVCDs
5 from the CTRL runs shows a similar behavior to that of the retrieved NO₂ TropVCDs,
6 despite the magnitude of *a priori* NO₂ TropVCD being $3.9 - 8.2 \times 10^{15}$ molec. cm⁻²
7 higher than those retrieved. On the other hand, the *a priori* NO₂ TropVCD from the
8 TM5 runs decreases between 08:00 and 14:00 LT, reflecting diurnally varying
9 photochemistry with similar levels of NO_x emissions throughout the day.

10 In October, there are broad maxima of NO₂ TropVCD between 12:00 LT and
11 15:00 LT. Overall diurnal behavior comprises increase up to 12:00 LT, followed by
12 broad maxima, after which the NO₂ TropVCD are similar to those in April.

13 The highest retrieved values are in the range 25.1×10^{15} to 25.5×10^{15} molec.
14 cm⁻² for both the TM5 and CTRL runs. As expected, the NO₂ TropVCD are the highest
15 in January and lowest in July.

16 **Figures 9 and 10** show the spatial distributions of retrieved NO₂ TropVCDs in
17 January, April, July, and October 2021 from the TM5 and CTRL runs, respectively. In
18 January, a plume over the SMA region developed as a function of time. Consequently,
19 the suburban areas, which surround the SMA region, experience relatively high NO₂
20 TropVCD ($> 10 \times 10^{15}$ molec. cm⁻²) compared to that retrieved in the other months. In
21 April and October, the plumes over the SMA are saturated prior to 12:00 LT and then
22 decrease. In contrast, the NO₂ TropVCD of the surrounding regions are relatively
23 constant or even increase. In July, the overall low values cover the SMA and nearby
24 regions for whole days. The maximum values appear at 10 LT, and then decreased until
25 14 LT. However, the NO₂ VCD rebounded at 16 LT. **Figure 11** displays the differences
26 between the TM5 and CTRL runs – red color indicates the CTRL run has higher values
27 than the TM5 run; blue means opposite. The CTRL run shows higher VCD values than

1 the TM5 run for all times in July. The largest differences over the SMA region are found
2 at 12 LT in July with differences of 2.1×10^{15} molec. cm^{-2} . In other months, the CTRL
3 run generally have higher values of VCD than the TM5 run over the Seoul and urban
4 regions, while there are lower values of VCD from the CTRL run over rural regions.

5

6 **4.2. Impacts of horizontal transport**

7 **Figure 12** shows the diurnal behavior of the retrieved NO_2 TropVCDs from the CTRL
8 run for different wind conditions. The black lines indicate calm runs (wind speed lower
9 than 3m/s), the green line is a strong-wind run (wind speed faster than 5m/s), and the
10 blue lines are the average values with no wind filters. In January (Figure 12a), the
11 diurnal behavior of the NO_2 TropVCDs change significantly with the wind conditions.
12 In the calm run (black solid), NO_2 TropVCD steadily increases due to a combination of
13 the emissions increasing and the slow chemical loss in this month. In windy runs,
14 however, diurnal changes in the retrieved NO_2 TropVCD are negligible.

15 Although the chemical loss is slow during wintertime, the accumulation of NO_2
16 was mitigated as strong winds transported large concentrations of NO_2 to downwind
17 regions. The differences between calm and other runs were most significant at 15:00
18 LT, further indicating that continuous outflow due to transport suppressed the
19 accumulation. As the wind speed increased, there was a noticeable reduction in NO_2
20 TropVCD values, which indicates a clear inverse relationship between wind speed and
21 VCDs, as shown in Edwards et al. (2024). The values of calm, average (blue solid), and
22 strong wind (green solid) are $19.0 - 28.9$, $17.2 - 19.8$, and $12.1 - 13.4 \times 10^{15}$ molec.
23 cm^{-2} , respectively.

24 In April (Figure 12b) and October (Figure 12d), the averaged values with no
25 wind filters (blue solid) have different diurnal behavior, but the maximum NO_2
26 TropVCD appear almost simultaneously with that of the calm low wind speed run. In

1 July (Figure 7c), however, the diurnal behavior from the calm run and no wind filters
2 are nearly identical, implying that the wind speeds are overall slow in July.

3 In summary, the transport effect is maximized in wintertime, changing not only
4 the absolute values but also diurnal behavior of NO₂ Trop VCDs. Consequently,
5 transport must be taken into account when analyzing NO₂ TropVCDs and when
6 estimating top-down NO_x emissions. The role of transport needs to be taken into
7 account even for cases, where the wind speed is relatively slow during summertime
8 (Yang et al., 2024).

9

10 **5. Conclusions**

11 In this study, we analyzed the seasonal variations and diurnal behavior of the retrieved
12 GEMS IUP-UB NO₂ TropVCD, using the monthly mean data in January, April, July
13 and October. The effects of wind speed, and the impact of *a priori* NO₂ profiles on the
14 retrieval. Both in the CTRL and TM5 runs, the GEMS NO₂ product showed significant
15 changes in quantity, diurnal pattern, and peak time as the seasons changed. In winter,
16 the values were the highest, with a gradual increase over time, whereas in summer, the
17 values were the lowest, reaching a minimum in the afternoon. This is consistent with
18 previous studies, which have shown that atmospheric chemical reactions are more
19 active in summer. Furthermore, we confirmed that wind-driven transport significantly
20 influences the diurnal patterns, clearly demonstrating that advection and possibly
21 convection need to be taken into account when top-down NO_x emissions are estimated
22 from an urban agglomeration such as SMA.

23 On the other hand, when using different *a priori* data to calculate VCD values,
24 more complex results emerged. A comparison between the CTRL and TM5 runs
25 revealed that, despite different spatial resolution and emission characteristics, the
26 retrieved NO₂ TropVCDs exhibited similar diurnal patterns, with significant differences

1 only in July. Additionally, we found that the retrieved NO₂ TropVCDs had diurnal
2 behaviors independent of the *a priori* data in both runs. We infer that the observed SCD
3 has a stronger influence on the retrieved diurnal patterns than *a priori* profiles.
4 Adjusting the horizontal resolution of the model (FINE run) or changing the VOC
5 emissions data (MIXED run) also resulted in no significant differences. However, in
6 the CONST run, where only the vertical profile at 14:00 LT was used in the retrieval
7 process throughout the day, there were significant differences in both the NO₂ Trop
8 VCD values and diurnal patterns. This reaffirms that the vertical shape factor of *a priori*
9 data plays a critical role in NO₂ TropVCD retrievals.

10 Additionally, given that vertical as well as horizontal model resolution can
11 influence retrievals (Liu et al., 2020), future studies should analyze the results when the
12 vertical resolution is adjusted. Furthermore, as highlighted by previous studies, such as
13 Lorente et al. (2017) and Hong et al. (2017), which emphasized the importance of cloud
14 parameters, aerosol characteristics, and surface albedo, uncertainties arising from
15 factors in addition to the *a priori* NO₂ profile should further be investigated in the
16 retrieval of NO₂ TropVCD for both diurnal GEO observation and those from LEO.

17

18 **Data availability**

19 GEMS measurement data retrieved by the IUP algorithm are available on request from
20 Andreas Richter (richter@iup.physik.uni-bremen.de). WRF-Chem v4.4 is available in
21 GitHub (wrf-model, 2022).

22

23 **Author contributions**

24 SWK initiated this study and secured funding. SS and SWK analyzed the satellite and
25 model data. SS, KMK, and SWK conducted the model simulations. AR, KL, and JPB
26 provided GEMS IUP products and analyzed the data. JK, JP, HH, HL, UJ retrieved and

1 analyzed the GEMS observations and discussed the results. JHW provided AQNEA
2 version 2 emission inventory. SS and SWK wrote the paper, with contributions from all
3 co-authors.

4

5 **Competing interests**

6 At least one of the authors is a member of the editorial board of Atmospheric
7 Measurement Techniques.

8

9 **Acknowledgement**

10 This work was supported by the National Research Foundation of Korea (NRF) grant
11 funded by the Korea government (MSIT) (No. 2020R1A2C2014131). All the
12 computing resources are provided by National Center for Meteorological
13 Supercomputer. Th contributions from the University of Bremen were supported by the
14 State and University of Bremen and the DLR.

15

16 **References**

17 Ahmadov, R., McKeen, S. A., Robinson, A. L., Bahreini, R., Middlebrook, A. M., de
18 Gouw, J. A., Meagher, J., Hsie, E.-Y., Edgerton, E., Shaw, S., and Trainer, M.: A
19 volatility basis set model for summertime secondary organic aerosols over the eastern
20 United States in 2006, *J. Geophys. Res. Atmos.*, 117, D06301,
21 <https://doi.org/10.1029/2011JD016831>, 2012.

22 Atmospheric Chemistry Observations & Modeling/National Center for Atmospheric
23 Research/University Corporation for Atmospheric Research: Whole Atmosphere
24 Community Climate Model (WACCM) Model Output, Research Data Archive at the
25 National Center for Atmospheric Research, Computational and Information System

1 Laboratory, <https://doi.org/10.5065/G643-Z138>, (last access: 05 December 2022),
2 2020.

3 Beirle, S., Hörmann, C., Jöckel, P., Liu, S., Penning De Vries, M., Pozzer, A., Sihler, H.,
4 Valks, P. and Wagner, T.: The STRatospheric Estimation Algorithm from Mainz
5 (STREAM): Estimating stratospheric NO₂ from nadir-viewing satellites by weighted
6 convolution, *Atmos. Meas. Tech.*, 9(7), 2753–2779, doi:10.5194/amt-9-2753-2016,
7 2016.

8 Bovensmann, H., Burrows, J. P., Buchwitz, M., Frerick, J., Noël, S., Rozanov, V. V.,
9 Chance, K. V., and Goded, A. P. H.: SCIAMACHY: Mission Objectives and
10 Measurement Modes, *J. Atmos. Sci.*, 56, 127-150, [https://doi.org/10.1175/1520-
11 0469\(1999\)056<0127:SMOAMM>2.0.CO;2](https://doi.org/10.1175/1520-0469(1999)056<0127:SMOAMM>2.0.CO;2), 1999.

12 Burrows J. P., Hölzle E., Goede A. P. H., Visser H. and Fricke, W.: SCIAMACHY -
13 Scanning Imaging Absorption Spectrometer for Atmospheric Cartography, *Acta
14 Astronautica*, 35, 445-461, [https://doi.org/10.1016/0094-5765\(94\)00278-T](https://doi.org/10.1016/0094-5765(94)00278-T), 1995.

15 Burrows, J. P., Weber, M., Buchwitz, M., Rozanov, V., Ladstätter-Weissenmayer, A.,
16 Richter, A., DeBeek, R., Hoogen, R., Bramstedt, K., Eichmann, K.-U., Elsinger, M.,
17 and Perner, D.: The Global Ozone Monitoring Experiment (GOME): Mission
18 Concept and First Scientific Results, *J. Atmos. Sci.*, 56, 151-175,
19 [https://doi.org/10.1175/1520-0469\(1999\)056<0151:TGOMEG>2.0.CO;2](https://doi.org/10.1175/1520-0469(1999)056<0151:TGOMEG>2.0.CO;2), 1999.

20 Burrows, J. P., Bovensmann, H., Bergametti, G., Flaud, J. M., Orphal, J., Noël, S.,
21 Monks, P. S., Corlett, G. K., Goede, A. P. H., von Clarmann, T., Steck, T., Fischer,
22 H., and Friedl-Vallon, F.: The geostationary tropospheric pollution explorer
23 (GeoTROPE) missions: objects, requirements and mission concept, *Advances in
24 Space Research*, 34, 682-687, <https://doi.org/10.1016/j.asr.2003.08.067>, 2004.

25 Callies, J., Corpaccioli, E., Eisinger, M., Hahne, A., and Lefebvre, A.: GOME-2-
26 Metop's second-generation sensor for operational ozone monitoring, *ESA Bulletin*,

1 102, 28–36, 2000.

2 Edwards, D. P., Martínez-Alonso, S., Jo, D.-S., Ortega, I., Emmons, L. K., Orlando, J.
3 J., Worden, H. M., Kim, J., Lee, H., Park, J., and Hong, H.: Quantifying the diurnal
4 variation of atmospheric NO₂ from observations of the Geostationary Environment
5 Monitoring Spectrometer (GEMS), *Atmos. Chem. Phys.*, 24, 8943-8961,
6 <https://doi.org/10.5194/acp-24-8943-2024>, 2024.

7 Granier, C., Bessagnet, B., Bond, T., D'Angiola, A., van der Gon, H. D., Frost, G. J.,
8 Heil, A., Kaiser, J. W., Kinne, S., Klimont, Z., Kloster, S., Lamarque, J.-F., Liousse,
9 C., Masui, T., Meleux, F., Mieville, A., Ohara, T., Raut, J.-C., Riahi, K., Schultz, M.
10 G., Smith, S. J., Thompson, A., van Aardenne, J., van der Warf, G. R., and van Vuuren,
11 D. P.: Evolution of anthropogenic and biomass burning emissions of air pollutants at
12 global and regional scales during the 1980 – 2010 period, *Climatic Change*, 109, 163,
13 <https://doi.org/10.1007/s10584-011-0154-1>, 2011.

14 Grell, G. A., Peckham, S. E., Schmitz, R., McKeen, S. A., Frost, G., Shamarock, W. C.,
15 and Eder, B.: Fully coupled “online” chemistry within the WRF model, *Atmos.*
16 *Environ.*, 39, 6957-6975, <https://doi.org/10.1016/j.atmosenv.2005.04.027>, 2005.

17 Hong, H., Lee, H., Kim, J., Jeong, U., Ryu, J., Lee, D. S.: Investigation of Simultaneous
18 Effects of Aerosol Properties and Aerosol Peak Height on the Air Mass Factors for
19 Space-Borne NO₂ Retrievals, *remote sens.*, 9(3), 208,
20 <https://doi.org/10.3390/rs9030208>, 2017.

21 Huijnen, V., Williams, J., van Weele, M., van Noije, T., Krol, M., Dentener, F., Segers,
22 A., Houweling, S., Peters, W., de Laat, J., Boersma, F., Bergamaschi, P., van
23 Velthoven, P., Le Sager, P., Eskes, H., Alkemade, F., Scheele, R., Nédélec, P., and
24 Pätz, H.-W.: The global chemistry transport model TM5: description and evaluation
25 of the tropospheric chemistry version 3.0, *Geosci. Model Dev.*, 3, 445-473,
26 <https://doi.org/10.5194/gmd-3-445-2010>, 2010.

1 Jang, Y., Lee, Y., Kim, J., Kim, Y., and Woo, J.-H.: Improvement China point source for
2 improving bottom-up emission inventory, *Asia-Pac. J. Atmos. Sci.*, 56, 107-118,
3 <https://doi.org/10.1007/s13143-019-00115-y>, 2020.

4 Kim, J., Jeong, U., Ahn, M.-H., Kim, J. H., Park, R. J., Lee, H., Song, C. H., Choi, Y.-
5 S., Lee, K.-J., Yoo, J.-M., Jeong, M.-J., Park, S. K., Lee, K.-M., Song, C.-K., Kim,
6 S.-W., Kim, Y. J., Kim, S.-W., Kim, M., Go, S., Liu, X., Chance, K., Miller, C. C.,
7 Al-Saadi, J., Veihelmann, B., Bhartia, P. K., Torres, O., González Abad, G., Haffner,
8 D. P., Ko, D. H., Lee, S. H., Woo, J.-H., Chong, H., Park, S. S., Nicks, D., Choi, W.
9 J., Moon, K.-J., Cho, A., Yoon, J., Kim, S.-K., Hong, H., Lee, K., Lee, H., Lee, S.,
10 Choi, M., Veekfind, P., Levelt, P. F., Edwards, D. P., Kang, M., Eo, M., Bak, J., Baek,
11 K., Kwon, H.-A., Yang, J., Park, J., Han, K. M., Kim, B.-R., Shin, H.-W., Choi, H.,
12 Lee, E., Chong, J., Cha, Y., Koo, J.-H., Irie, H., Hayashida, S., Kasai, Y., Kanaya, Y.,
13 Liu, C., Lin, J., Crawford, J. H., Carmichael, G. R., Newchurch, M. J., Lefter, B. L.,
14 Herman, J. R., Swap, R. J., Lau, A. K. H., Kurosu, T. P., Jaross, G., Ahlers, B., Dobber,
15 M., McElroy, C. T., and Choi, Y.: New Era of Air Quality Monitoring from Space:
16 Geostationary Environment Monitoring Spectrometer (GEMS), *Bull. Amer. Meteor.*
17 *Soc.*, 101, E1-E22, <https://doi.org/10.1175/BAMS-D-18-0013.1>, 2020.

18 Kim, K.-M., Kim, S.-W., Seo, S., Blake, D. R., Cho, S., Crawford, J. H., Emmons, L.
19 K., Fried, A., Herman, J. R., Hong, J., Jung, J., Pfister, G. G., Weinheimer, A. J., Woo,
20 J.-H., and Zhang, Q.: Sensitivity of the WRF-Chem v4.4 simulations of ozone and
21 formaldehyde and their precursors to multiple bottom-up emission inventories over
22 East Asia during the KORUS-AQ 2016 field campaign, *Geosci. Model Dev.*, 17,
23 1931-1955, <https://doi.org/10.5194/gmd-17-1931-2024>, 2024.

24 Kim, S.-W., McDonald, B. C., Brown, S. S., Dube, B., Ferrare, R. A., Frost, G. J., Harley,
25 R. A., Holloway, J. S., Lee, H.-J., McKeen, S. A., Neuman, J. A., Nowak, J. B., Oetjen,
26 H., Ortega, I., Pollack, I. B., Roberts, J. M., Ryerson, T. B., Scarino, A. J., Senff, C.
27 J., Thalman, R., Trainer, M., Volkamer, R., Wagner, N., Washenfelder, R. A., Waxman,

1 E., and Young, C. J.: Modeling the weekly cycle of NO_x and CO emissions and their
2 impacts on O₃ in the Los Angeles-South Coast Air Basin during the CalNex 2010
3 field campaign, *J. Geophys. Res. Atmos.*, 121, 1340-1360,
4 <https://doi.org/10.1002/2015JD024292>, 2016.

5 Lange, K., Richter, Bösch, T., Zilker, B., Latsch, M., Behrens, L. K., Okafor, C. M.,
6 Bösch, H., Burrows, J. P., Merlaud, A., Pinardi, G., Fayt, C., Friedrich, M. M.,
7 Dimitropoulou, E., Van Roozendael, M., Ziegler, S., Ripperger-Lukosiunaite, S.,
8 Kuhn, L., Lauster, B., Wagner, T., Hong, H., Kim, D., Chang, L.-S., Bae, K., Song,
9 C.-K., and Lee, H.: Validation of GEMS tropospheric NO₂ columns and their diurnal
10 variation with ground-based DOAS measurements, *EGUsphere [preprint]*,
11 <https://doi.org/10.5194/egusphere-2024-617>, 2024.

12 Levelt, P. F., van den Oord, G. H. J., Dobber, M., R., Mälkki, A., Visser, H., de Vries,
13 J., Stammes, P., Lundell, J. O. V., and Saari, H.: The Ozone Monitoring Instrument,
14 *IEEE Trans. Geosci. Remote Sens.*, 44(5), 1093-1101,
15 <https://doi.org/10.1109/TGRS.2006.872333>, 2006.

16 Liu, S., Valks, P. Pinardi, G., Xu, J., Argyrouli, A., Lutz, R., Tilstra, G., Huijnen, V.,
17 Hendric, F., and Van Roozendael, M.: An improved air mass factor calculation for
18 nitrogen dioxide measurements from the Global Ozone Monitoring Experiment-2
19 (GOME-2), *Atmos. Meas. Tech.*, 13, 755-787, [https://doi.org/10.5194/amt-13-755-](https://doi.org/10.5194/amt-13-755-2020)
20 [2020](https://doi.org/10.5194/amt-13-755-2020), 2020.

21 Lorente, A., Boersma, K. F., Yu, H., Dörner, S., Hilboll, A., Richter, A., Liu, M., Lamsal,
22 L. N., Barkley, M., De Smedt, I., Van Roozendael, M., Wang, Y., Wagner, T., Beirle,
23 S., Lin, J.-T., Krotkov, N., Stammes, P., Wang, P., Eskes, H. J., and Krol, M.:
24 Structural uncertainty in air mass factor calculation for NO₂ and HCHO satellite
25 retrievals, *Atmos. Mech. Tech.*, 10, 759-782, [https://doi.org/10.5194/amt-10-759-](https://doi.org/10.5194/amt-10-759-2017)
26 [2017](https://doi.org/10.5194/amt-10-759-2017), 2017.

- 1 Milford, J. B., Russell, A. G., and McRae, G. J.: A new approach to photochemical
2 pollution control: implications of spatial patterns in pollutant responses in nitrogen
3 oxides and reactive organic gas emissions, *Environ. Sci. Technol.*, 23, 1290-1301,
4 <https://doi.org/10.1021/es00068a017>, 1989.
- 5 Munro, R., Lang, R., Klaes, D., Poli, G., Retscher, C., Lindstrot, R., Huckle, R., Lacan,
6 A., Grzegorski, M., Holdak, A., Kokhanovsky, A., Livschitz, J., and Eisinger, M.:
7 The GOME-2 instrument on the Metop series of satellites: instrument design,
8 calibration, and level 1 data processing – an overview, *Atmos. Meas. Tech.*, 9, 1279–
9 1301, <https://doi.org/10.5194/amt-9-1279-2016>, 2016.
- 10 Palmer, P. I., Jacob, D. J., Chance, K., Martin, R. V., Spurr, R. J. D., Kurosu, T. P., Bey,
11 I., Yantosca, R., Fiore, A., and Li, Q.: Air mass factor formulation for spectroscopic
12 measurements from satellites: Application to formaldehyde retrievals from the
13 Global Ozone Monitoring Experiment, *J. Geophys. Res.*, 106, 14539-14550,
14 <https://doi.org/10.1029/2000JD900772>., 2001.
- 15 Richter, A. and Burrows, J. P.: Tropospheric NO₂ from GOME Measurements,
16 *Advances in Space Research*, 29, 1673-1683, [https://doi.org/10.1016/S0273-
17 1177\(02\)00100-X](https://doi.org/10.1016/S0273-1177(02)00100-X), 2002.
- 18 Rozanov, V. V., Rozanov, A. V., Kokhanovsky, A. A. and Burrows, J. P.: Radiative
19 transfer through terrestrial atmosphere and ocean: Software package SCIATRAN, *J.*
20 *Quant. Spectrosc. Radiat. Transf.*, 133, 13–71,
21 <https://doi.org/10.1016/j.jqsrt.2013.07.004>, 2014.
- 22 Skamarock, W. C., Klemp, J. B., Dudhia, J., Gill, D. O., Liu, Z., Berner, J., Wang, W.,
23 Powers, J. G., Duda, M. G., Barker, D. M., Huang, X.-Y.: A Description of the
24 Advanced Research WRF Model Version 4 (No. NCAR/TN-556+STR),
25 <https://doi.org/10.5065/1dfh-6p97>, 2021.
- 26 Tilstra, L. G., de Graaf, M., Trees, V. J. H., Litvinov, P., Dubovik, O., and Stammes, P.:

1 A directional surface reflectance climatology determined from TROPOMI
2 observations, *Atmos. Meas. Tech.*, 17, 2235-2256, [https://doi.org/10.5194/amt-17-](https://doi.org/10.5194/amt-17-2235-2024)
3 [2235-2024](https://doi.org/10.5194/amt-17-2235-2024), 2024.

4 Veefkind, J. P., Aben, I., McMullan, K. Förster, H., de Vries, J., Otter, G., Claas, J.,
5 Eskes, H. J., de Haan, J. F., Kleipool, Q., van Weele, M., Hasekamp, O., Hoogeveen,
6 R., Landgraf, J., Snel, R., Tol, P., Ingmann, P., Voors, R., Kruizinga, B., Vink, R.,
7 Visser, H., and Levelt, P. F.: TROPOMI on the ESA Sentinel-5 Precursor: A GMES
8 mission for global observations of the atmospheric composition for climate, air
9 quality and ozone layer applications, *Remote Sens. Environ.*, 120, 70-83,
10 <https://doi.org/10.1016/j.rse.2011.09.027>, 2012.

11 Williams, J. E., Boersma, K. F., Le Sager, P., and Verstraeten, W. W.: The high-
12 resolution version of TM5-MP for optimized satellite retrievals: description and
13 validation, *Geosci. Model Dev.*, 10, 721–750, [https://doi.org/10.5194/gmd-10-721-](https://doi.org/10.5194/gmd-10-721-2017)
14 [2017](https://doi.org/10.5194/gmd-10-721-2017), 2017.

15 Woo, J.-H., Choi, K.-C., Kim, H. K., Baek, B. H., Jang, M., Eum, J.-H., Song, C. H.,
16 Ma, Y.-I., Sunwoo, Y., Chang, L.-S., and Yoo, S. H.: Development of an
17 anthropogenic emissions processing system for Asia using SMOKE, *Atmos. Environ.*,
18 58, 5-13, <https://doi.org/10.1016/j.atmosenv.2011.10.042>, 2012.

19 wrf-model: WRF, Github [code], <https://github.com/wrf-model/WRF/release/tag/v4.4>,
20 last access: 18 May 2022.

21 Yang, L. H., Jacob, D. J., Dang, R., Oak, Y. J., Lin, H., Kim, J., Zhai, S., Colombi, N.
22 K., Pendergrass, D. C., Beaudry, E., Shah, V., Feng, X., Yantosca, R. M., Chong, H.,
23 Park, J., Lee, H., Lee, W.-J., Kim, S., Kim, E., Travis, K. R., Crawford, J. H., Liao,
24 H., Interpreting GEMS geostationary satellite observations of the diurnal variation
25 of nitrogen dioxide (NO₂) over East Asia, *Atmos. Chem. Phys.*,
26 <https://doi.org/10.5194/acp-24-7027-2024>, 2024.

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8 **Figure 3.** Spatial distributions of air mass factor (AMF) differences (CTRL – TM5) in
9 January, April, July, and October 2021. The pixels with wind speed faster than 3m/s are
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11 **Figure 4.** Diurnal patterns of the air mass factor during weekdays in (a) January, (b)
12 April, (c) July, and (d) October 2021 over the SMA region. Gray lines indicate the TM5
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14 are excluded.

15 **Figure 5.** Vertical profiles of *a priori* NO₂ mixing ratios at 08, 10, 12, 14, and 16 LT
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18 **Figure 6.** Diurnal patterns of retrieved (solid) and *a priori* (dashed) NO₂ TropVCDs in
19 July 2021 over SMA region from the CTRL run (black) and (a) CONST run (red), (b)
20 FINE run (pink), and (c) MIXED run (yellow). The pixels with wind speed faster than
21 3m/s are excluded. Note that diurnal changes of *a priori* NO₂ TropVCDs in the CONST
22 run occur during calculating domain-averaged values – the location and number of
23 pixels excluded during the collocation with satellite data vary over time during the day.

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6 wind speed faster than 3m/s are excluded to minimize the impact of rapid transport.

7 **Figure 10.** Same as Figure 9, except that *a priori* values for the AMF calculation are
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9 **Figure 11.** Similar to Figure 9, but for the differences of NO₂ TropVCD between CTRL
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11 **Figure 12.** Diurnal patterns of retrieved NO₂ TropVCDs from the CTRL run in (a)
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13 indicate the NO₂ TropVCD values with wind-filtered data; only the scenes with wind
14 speed lower than 3m/s are utilized. Blue lines are the averaged values without any wind
15 filters. The green line is for case of strong-wind run with the NO₂ TropVCD being
16 selected and averaged for wind speeds faster than 5m/s in January.

17

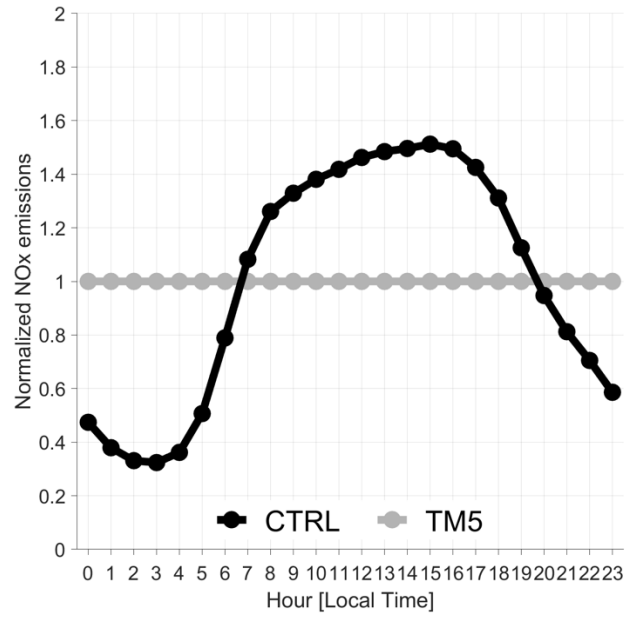
1 **Table 1.** Description of the experimental designs. MACCity provides hourly-constant
 2 emissions, while the others provide hourly-varying emissions.

Run name	Model	Horizontal resolution	Emission inventory
TM5	TM5	1° × 1°	MACCity
CTRL		28 × 28 km ²	2021AQNEA
CONST ^{a)}	WRF-Chem v4.4	28 × 28 km ²	2021AQNEA
FINE		12 × 12 km ²	2021AQNEA
MIXED		28 × 28 km ²	(VOC) KORUSv5 (others) 2021AQNEA

3

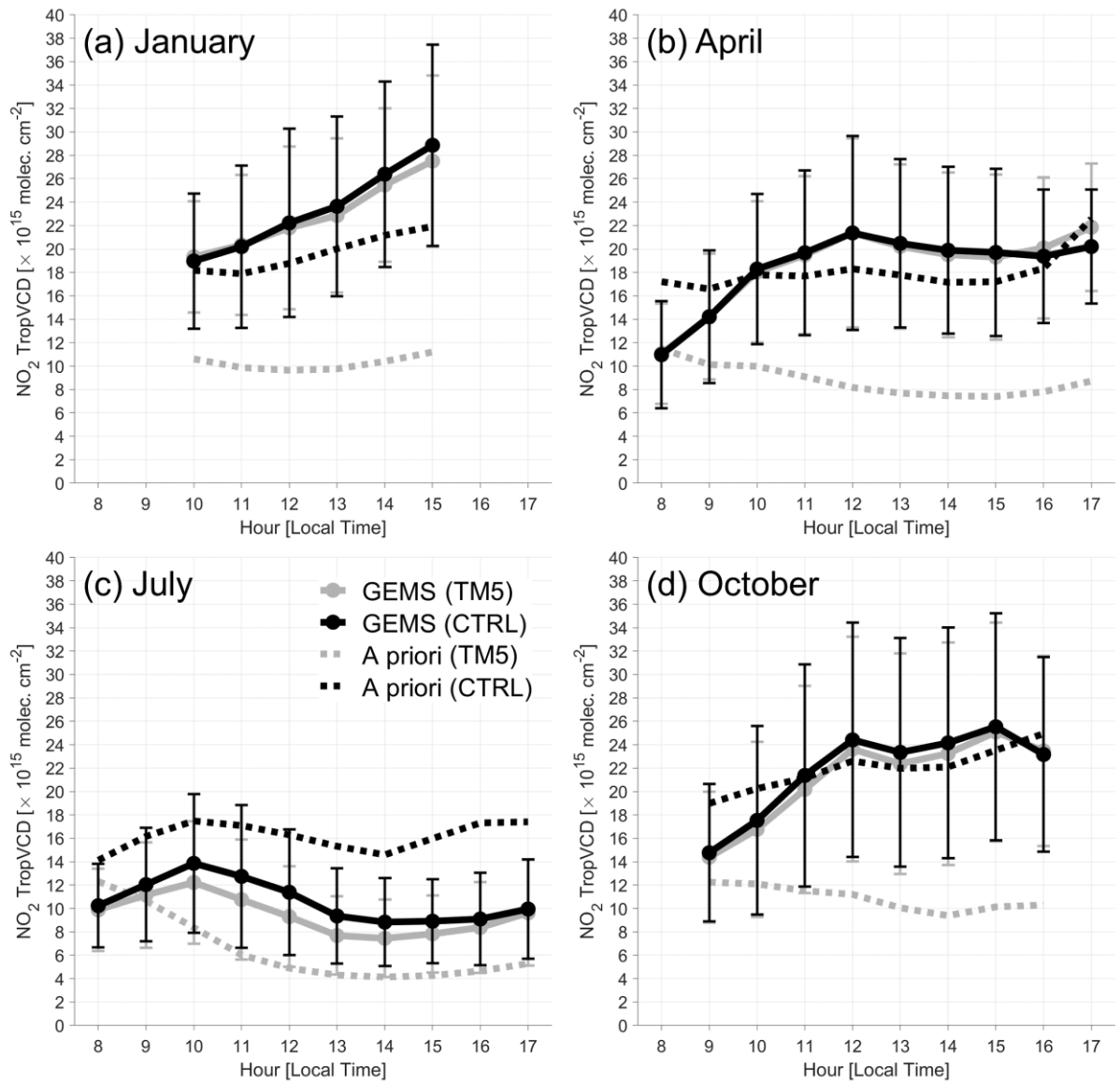
4 ^{a)} CONST run uses hourly-varying emission inventory, but only data of 13:45 LT were
 5 utilized to compute AMF.

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2 **Figure 1.** Diurnal variabilities of normalized NOx emissions for CTRL (black) and
3 TM5 (gray) runs over the SMA region.

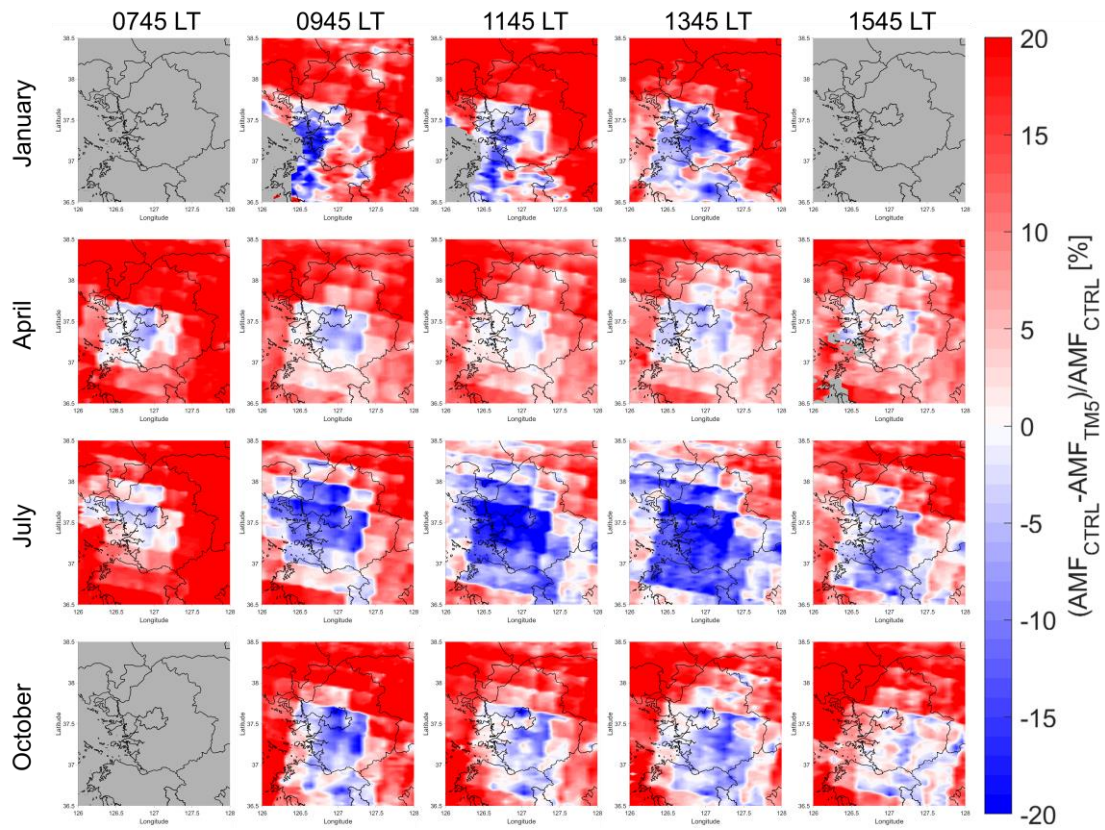


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2 **Figure 2.** Diurnal behavior of retrieved (solid) and *a priori* (dashed) NO₂ TropVCDs
 3 during weekdays in (a) January, (b) April, (c) July, and (d) October 2021 over the SMA
 4 region. Gray lines identify the TM5 run, while black lines represent the CTRL run. The
 5 pixels with wind speed faster than 3m/s are excluded.

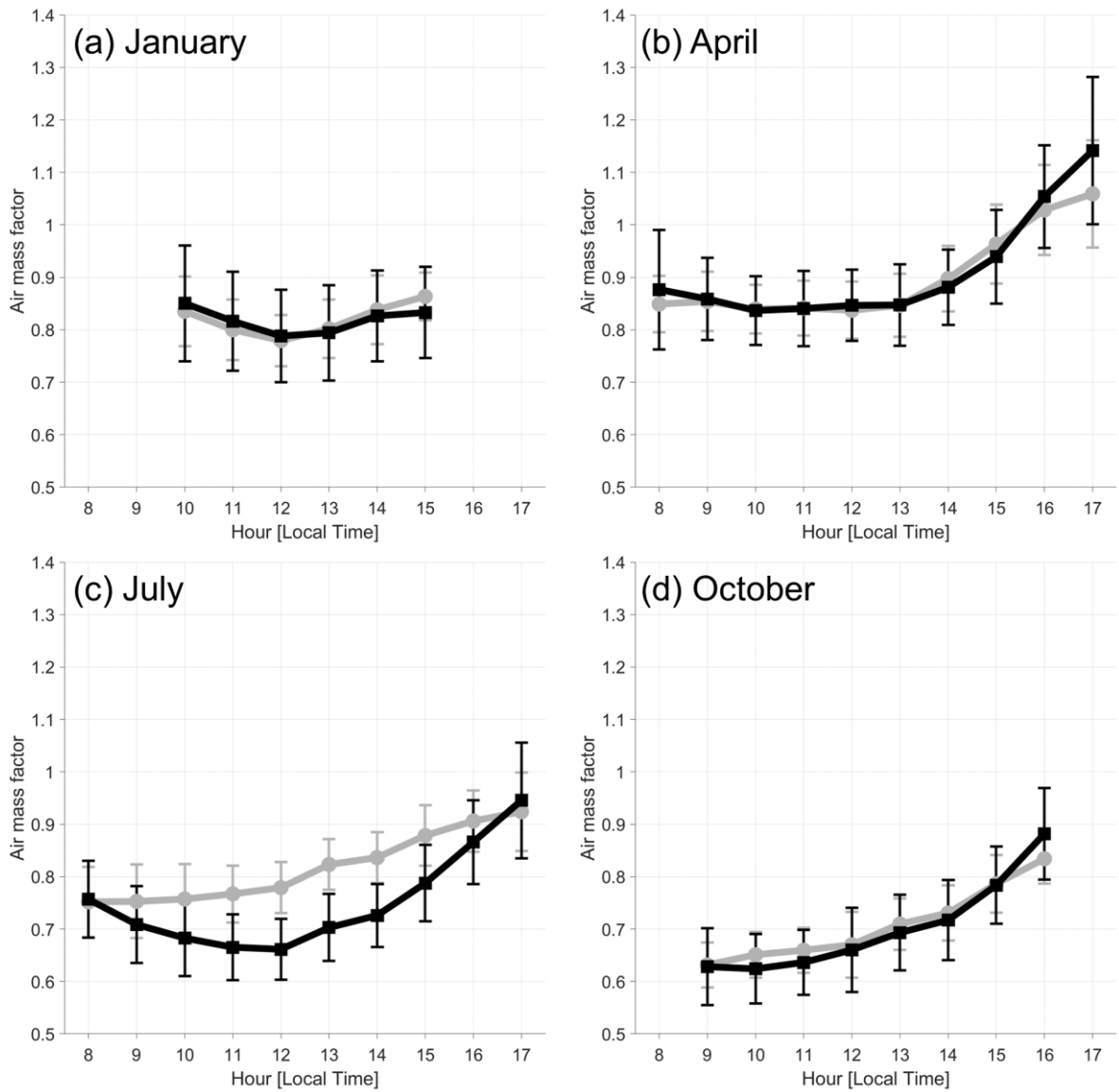
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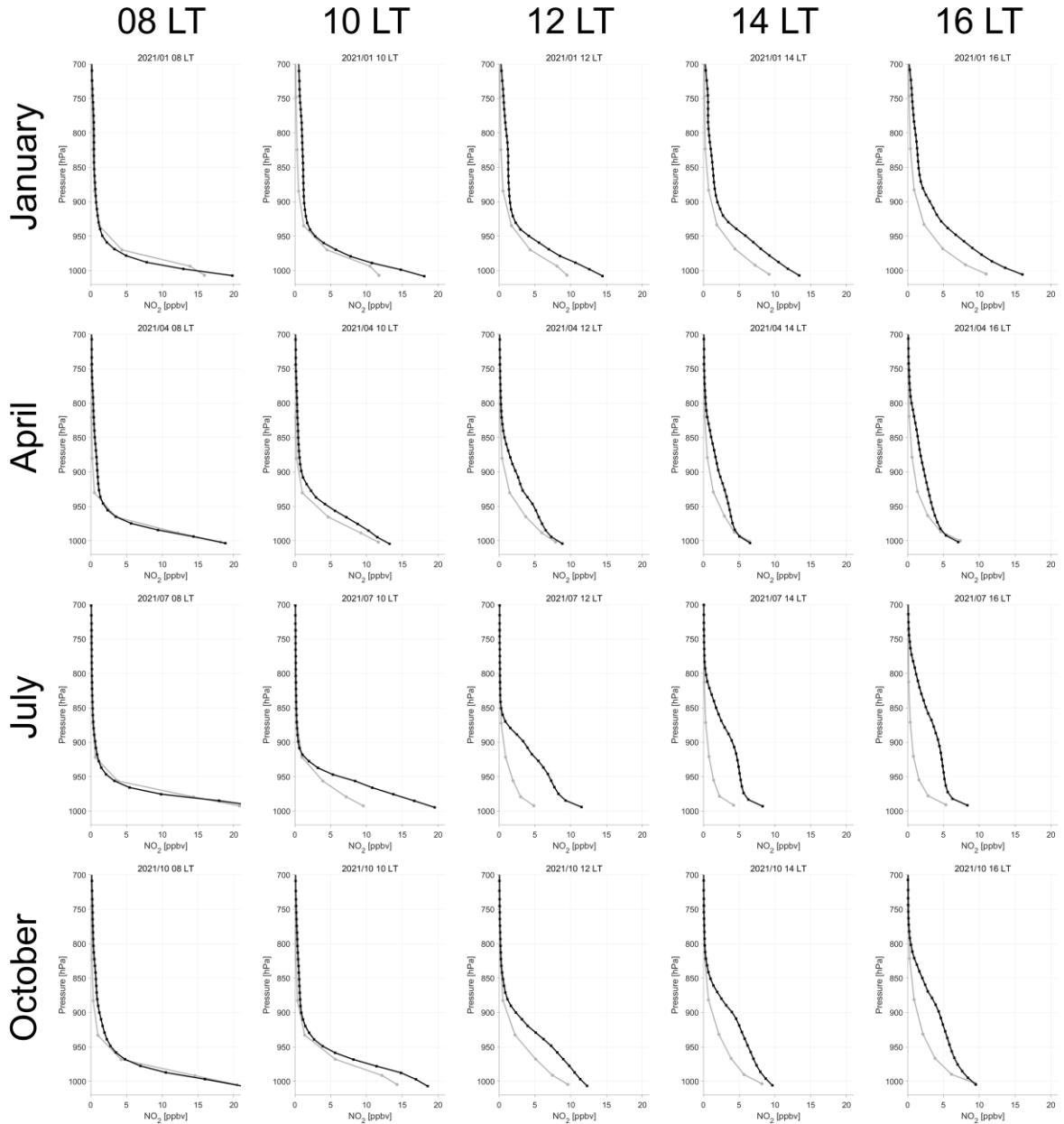
2 **Figure 3.** Spatial distributions of air mass factor (AMF) differences (CTRL – TM5) in
 3 January, April, July, and October 2021. The pixels with wind speed faster than 3m/s are
 4 excluded.



1

2 **Figure 4.** Diurnal patterns of the air mass factor during weekdays in (a) January, (b)
 3 April, (c) July, and (d) October 2021 over the SMA region. Gray lines indicate the TM5
 4 run, while black lines mean the CTRL run. The pixels with wind speed faster than 3m/s
 5 are excluded.

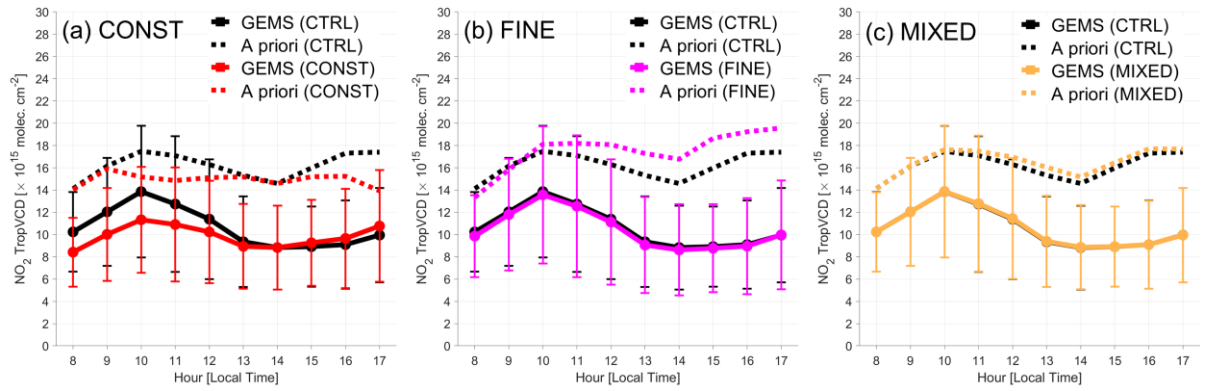
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2 **Figure 5.** Vertical profiles of *a priori* NO₂ mixing ratios at 08, 10, 12, 14, and 16 LT
 3 from the TM5 (gray) and CTRL (black) runs in January, April, July, and October 2021
 4 over the SMA region.

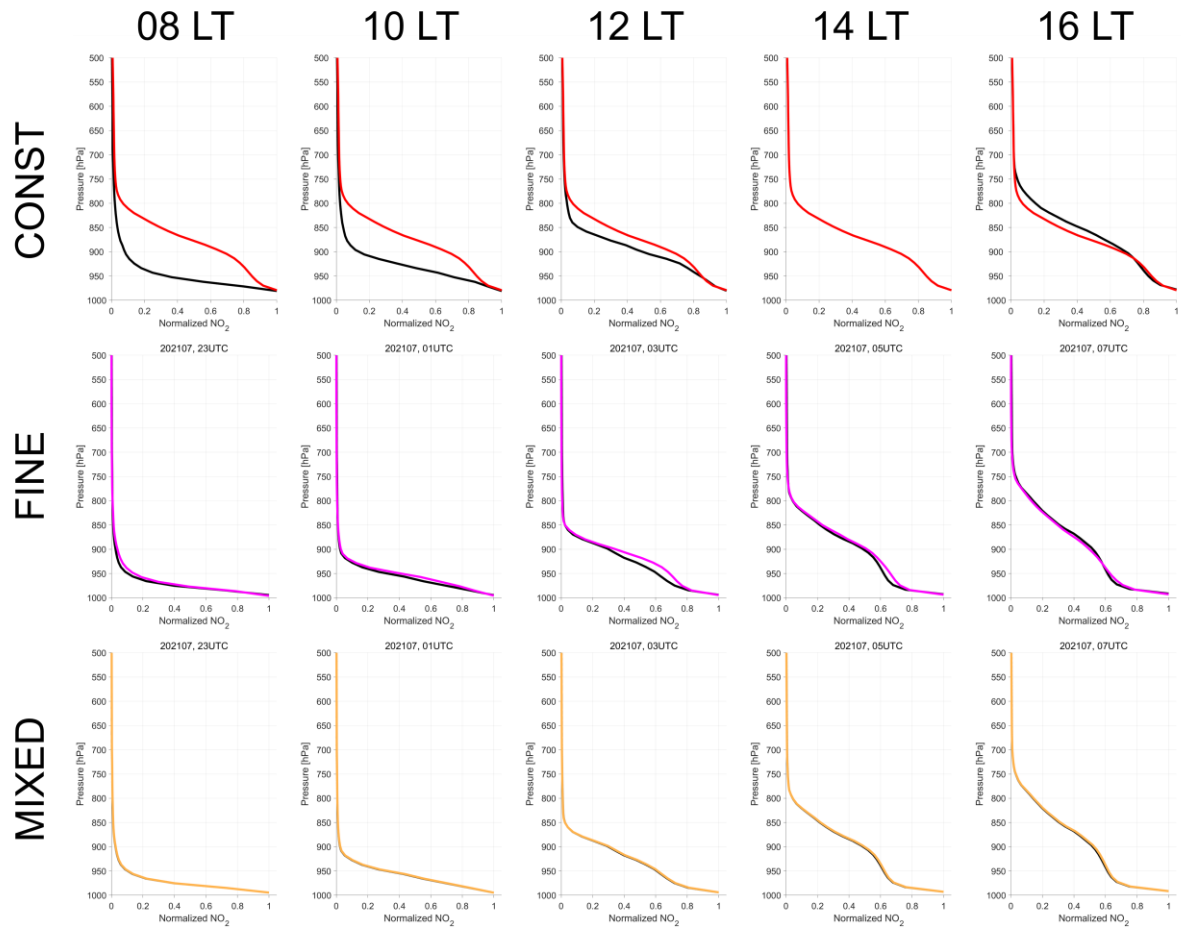
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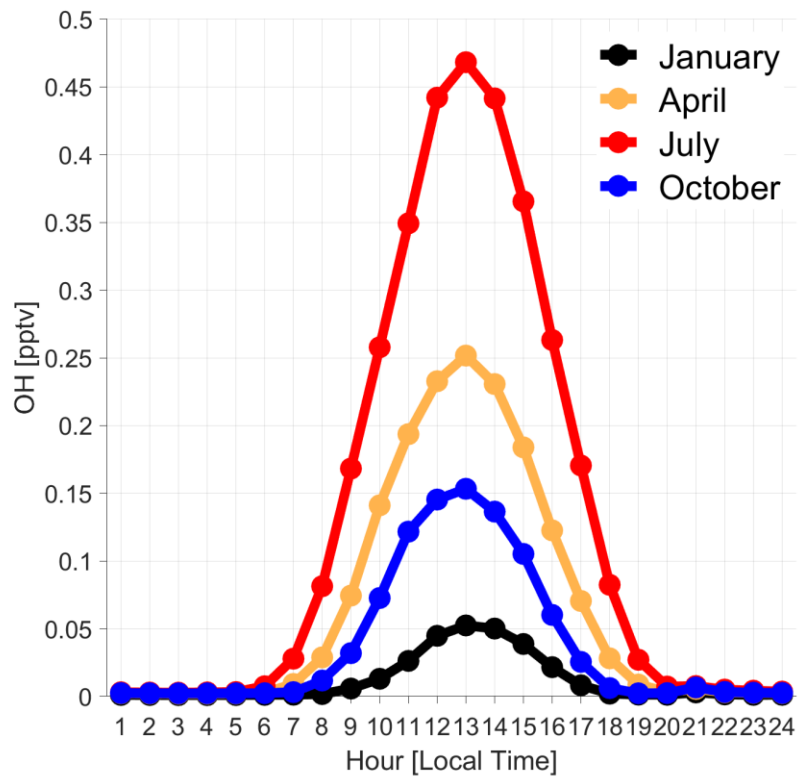
2 **Figure 6.** Diurnal patterns of retrieved (solid) and *a priori* (dashed) NO₂ TropVCDs in
 3 July 2021 over SMA region from the CTRL run (black) and (a) CONST run (red), (b)
 4 FINE run (pink), and (c) MIXED run (yellow). The pixels with wind speed faster than
 5 3m/s are excluded. Note that diurnal changes of *a priori* NO₂ TropVCDs in the CONST
 6 run occur during calculating domain-averaged values – the location and number of
 7 pixels excluded during the collocation with satellite data vary over time during the day.

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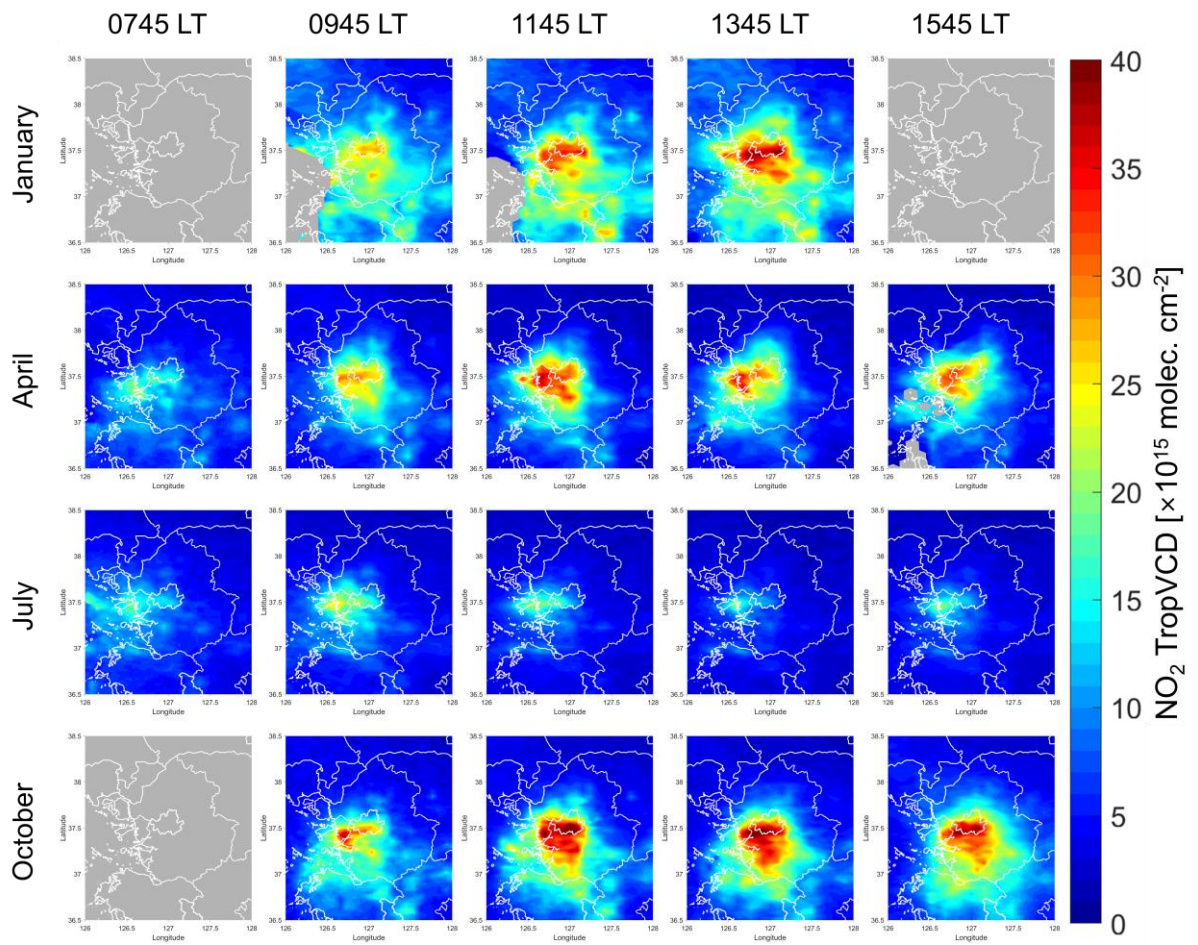
2 **Figure 7.** Vertical profiles of *a priori* NO₂ mixing ratios at 08, 10, 12, 14, and 16 LT
 3 from the CTRL (black), CONST (red), FINE (pink), and MIXED run (yellow) in
 4 January, April, July, and October 2021 over the SMA region.



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2 **Figure 8.** Diurnal patterns of boundary layer mean OH concentrations over the SMA
 3 region in January (black), April (yellow), July (red), and October (blue) 2021 from the
 4 CTRL run.

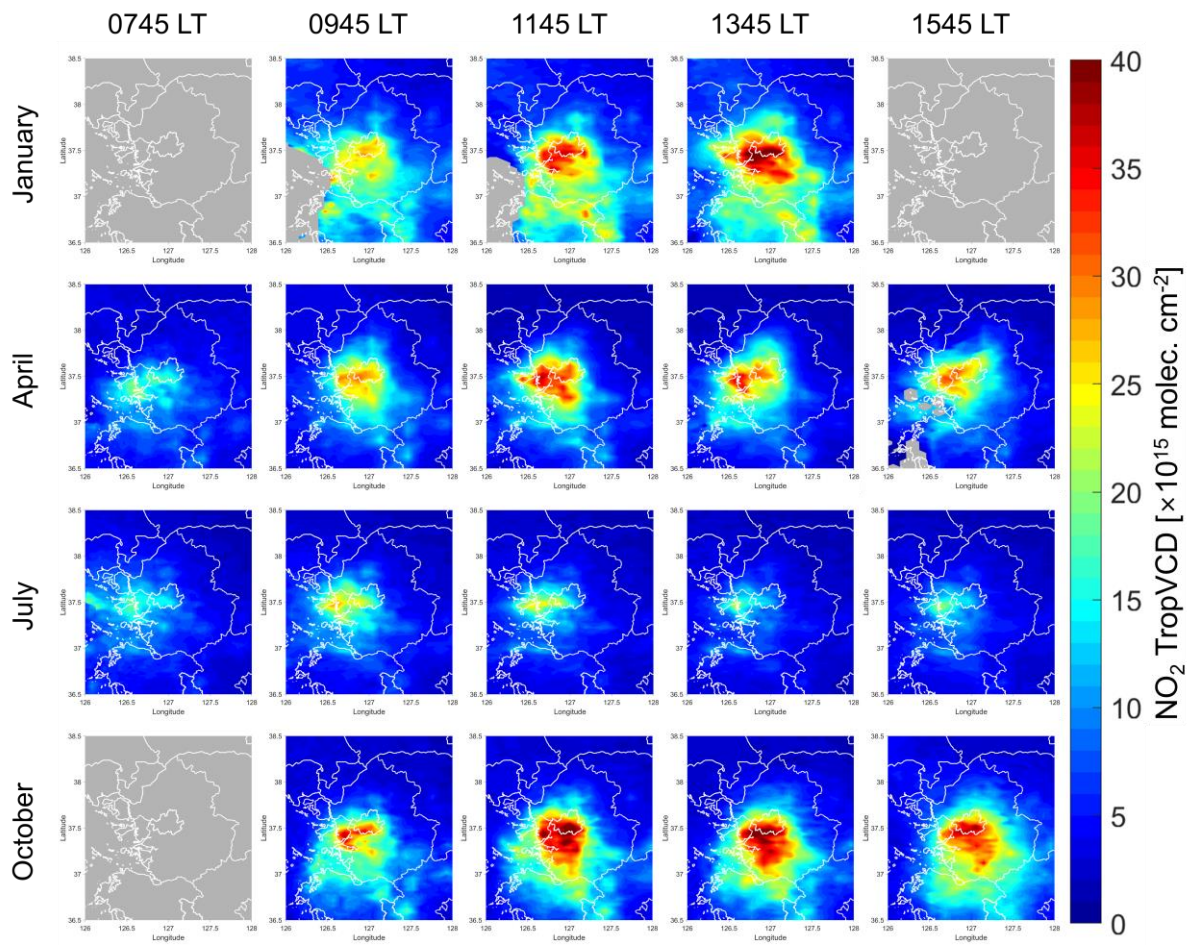
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2 **Figure 9.** Spatial distributions of retrieved NO₂ TropVCDs in January, April, July, and
 3 October 2021 taking the a priori data for the AMF from the TM5 run. The scenes with
 4 wind speed faster than 3m/s are excluded to minimize the impact of rapid transport.

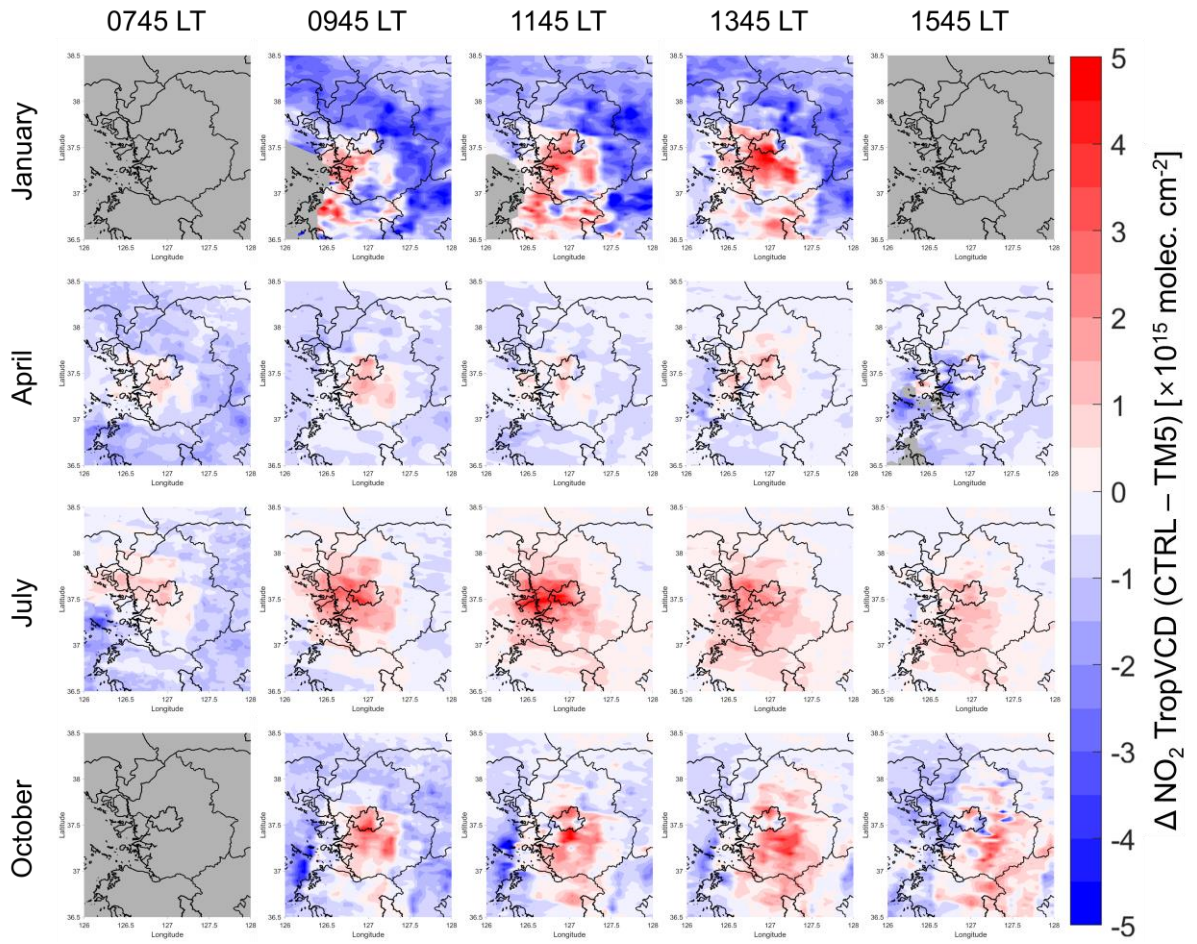
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2 **Figure 10.** Same as Figure 9, except that *a priori* values for the AMF calculation are
 3 taken from the CTRL run.

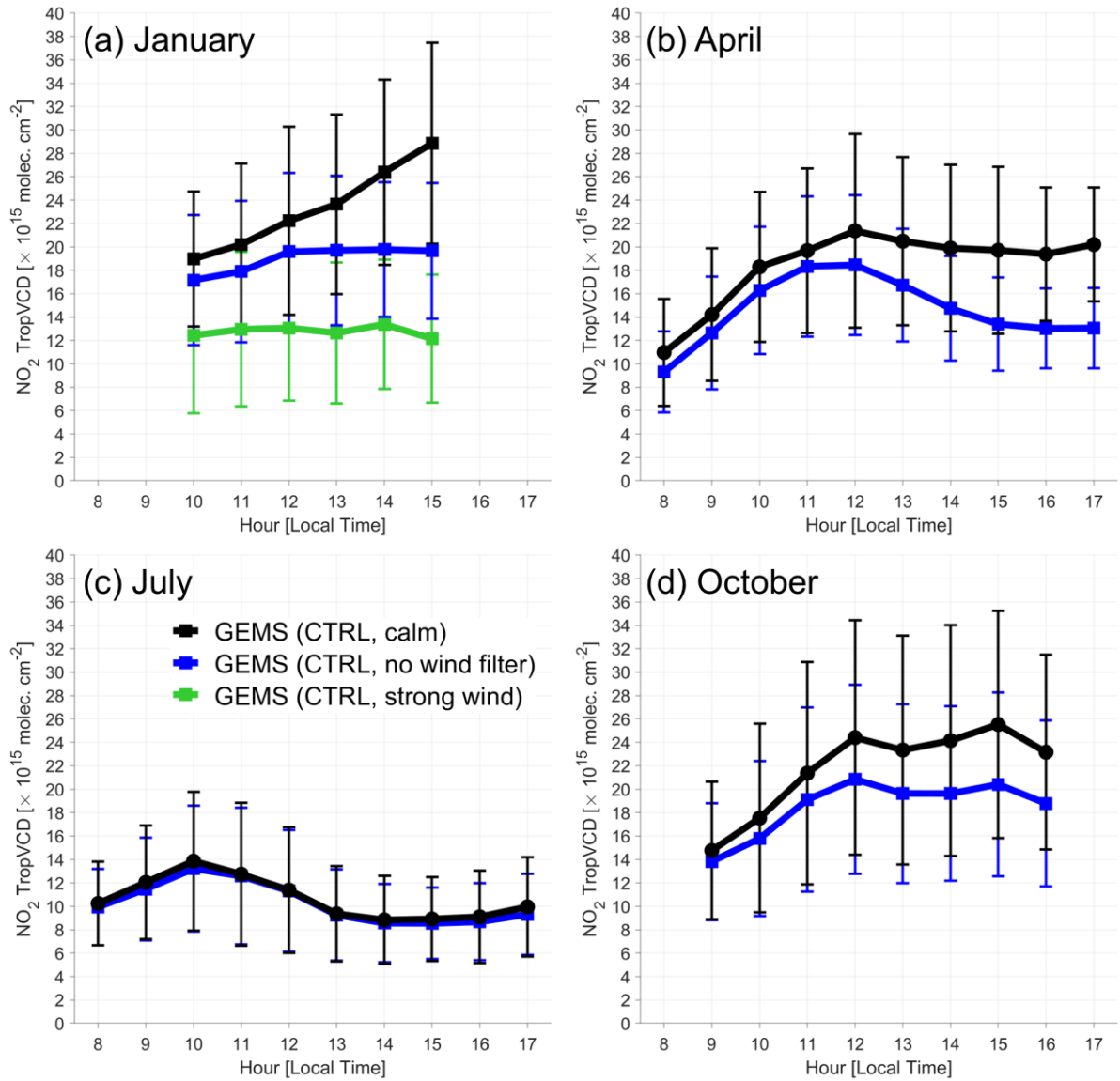
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2 **Figure 11.** Similar to Figure 9, but for the differences of NO₂ TropVCD between CTRL
 3 and TM5 run (CTRL – TM5).

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2 **Figure 12.** Diurnal patterns of retrieved NO₂ TropVCDs from the CTRL run in (a)
 3 January, (b) April, (c) July, and (d) October 2021 over the SMA region. Black lines
 4 indicate the NO₂ TropVCD values with wind-filtered data; only the scenes with wind
 5 speed lower than 3m/s are utilized. Blue lines are the averaged values without any wind
 6 filters. The green line is for case of strong-wind run with the NO₂ TropVCD being
 7 selected and averaged for wind speeds faster than 5m/s in January.