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Itahashi, Syuichi

Irie, Hitoshi

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Surface and aloft NO₂ pollution over the greater Tokyo area observed by ground-based and MAX-DOAS measurements bridged by kilometer-scale regional air quality modeling

Syuichi Itahashi^{1*}  and Hitoshi Irie²

Abstract

To advance our understanding of surface and aloft nitrogen dioxide (NO₂) pollution, this study extensively evaluated NO₂ concentrations simulated by the regional air quality modeling system with a 1.3 km horizontal grid resolution using the Atmospheric Environmental Regional Observation System ground-based observation network and aloft measurements by multi-axis differential optical absorption spectroscopy (MAX-DOAS) over the greater Tokyo area. Observations are usually limited to the surface level, and gaps remain in our understanding of the behavior of air pollutants above the near-surface layer, particularly within the planetary boundary layer (PBL). Therefore, MAX-DOAS measurement was used, which observes scattered sunlight in the ultraviolet/visible range at several elevation angles between the horizon and zenith to determine the aloft NO₂ pollution averaged over 0–1 km. In total, four MAX-DOAS measurement systems at Chiba University (35.63°N, 140.10°E) systematically covered the north, east, west, and south directions to capture the aloft NO₂ pollution over the greater Tokyo area. The target period was Chiba-Campaign 2015 conducted during 9–23 November 2015. The evaluations showed that the air quality modeling system can generally capture the observed behavior of both surface and aloft NO₂ pollution in terms of spatial and temporal coverage. The diurnal variation, which typically showed an increase from evening to early morning without daylight and a decrease during the daytime, was also captured by the model. During Chiba-Campaign 2015, two cases of episodic higher NO₂ concentration were identified: one during the nighttime and another during the daytime as different diurnal patterns. These were related to a stagnant wind field, with the latter also connected to a lower PBL height in cloudy conditions. Comparison of the modeled daily-averaged surface and aloft NO₂ concentrations showed that aloft NO₂ concentration exhibited a strong linear correlation with surface NO₂ concentration, with the aloft (0–1 km) value scaled to 0.4–0.5-fold the surface value, irrespective of whether the day was clean or polluted. This scaling value was lower during the nighttime and higher during the daytime. Based on this synergetic analysis of surface and aloft observation bridged by a kilometer-scale fine-resolution modeling simulation, this study contributes to fostering understanding of aloft NO₂ pollution.

*Correspondence: isyuichi@criepi.denken.or.jp

¹ Sustainable System Research Laboratory (SSRL), Central Research Institute of Electric Power Industry (CRIEPI), 1646 Abiko, Abiko, Chiba 270-1194, Japan

Full list of author information is available at the end of the article



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Keywords: Nitrogen dioxide, AEROS, MAX-DOAS, Air quality model, High-resolution modeling, Planetary boundary layer, Chiba-Campaign 2015

1 Introduction

Nitrogen oxides (NO_x), which collectively refers to nitric oxide (NO) and nitrogen dioxide (NO_2), play a crucial role in tropospheric chemistry and have important implications for air quality and climate (Seinfeld and Pandis 2016). NO_x is mainly emitted into the atmosphere as NO from anthropogenic sources (e.g., fossil fuel combustion) and natural sources (e.g., microbiological processes in soil and lightning) (Lin 2012; Kurokawa et al. 2013). Since NO_2 can affect human respiratory organs at high concentrations, an air quality standard (AQS) was established for NO_2 in 1973 and revised in 1978 in Japan. To satisfy this AQS, the daily average of hourly values of NO_2 concentrations must be 0.04–0.06 ppm or lower. In Japan, the AQS for NO_2 is mostly satisfied except at a few sites located close to roadsides (Wakamatsu et al. 2013). Given that the placement of ground-based observation is generally designed to protect human health, and most observation sites are located in densely populated areas, observation sites are sparse in rural and remote areas. To obtain a homogeneous spatial distribution of air pollutants, satellite observation can serve as a useful tool and our previous studies have reported space-borne NO_2 pollution over Asia and Japan (Irie et al. 2016; Itahashi et al. 2014, 2019; Choi et al. 2021). Although satellite observations have the potential to contribute to understanding spatial distribution patterns, the observed concentration is the amount vertically integrated over the column from the surface through the entire troposphere or sliced layers within the troposphere. To focus on the near-surface NO_2 behavior, additional observations are desired. This study utilizes multi-axis differential optical absorption spectroscopy (MAX-DOAS). MAX-DOAS is able to measure vertical profiles by observing scattered sunlight in the ultraviolet (UV)/visible range at several elevation angles between the horizon and zenith. By combining ground-based observation with MAX-DOAS, surface and aloft NO_2 pollution can be monitored. However, nighttime aloft NO_2 cannot be detected because the principle of MAX-DOAS relies on sunlight. To bridge spatial and temporal information, further regional air quality modeling needs to be conducted. This study therefore aims to understand the behavior of surface and aloft NO_2 and to fill in gaps in our understanding of the status of NO_2 pollution within the planetary boundary layer (PBL).

2 Methods

Chiba prefecture is located to the east of Tokyo and is part of the greater Tokyo area (Tokyo, Kanagawa, Saitama, and Chiba). At Chiba University, located in the central part of Chiba prefecture, an intensive observation campaign (Chiba-Campaign 2015) was conducted from 9 to 22 November 2015. This campaign is the target of this study, through which we aim to understand the behavior of surface and aloft NO_2 and to fill in gaps in our understanding of the status of NO_2 pollution within the PBL. Section 2.1 presents the ground-based observation. Section 2.2 describes the MAX-DOAS measurement. Section 2.3 introduces the regional air quality modeling.

2.1 Ground-based observation

Ground-based observation of air pollutants that are regulated by AQS (carbon monoxide (CO), sulfur dioxide (SO_2), NO_2 , photochemical oxidants (O_x), suspended particulate matter (SPM), and particulate matter with an aerodynamic diameter less than $2.5 \mu\text{m}$ ($\text{PM}_{2.5}$)) have been routinely measured by the Atmospheric Environmental Regional Observation System (AEROS) (<http://soramame.taiki.go.jp>). AEROS has two goals: to monitor ambient air quality using ambient air pollution monitoring stations (APMSs) and to measure air pollution particularly due to transportation such as automobiles using roadside air pollution monitoring stations (RAPMSs). The locations of AEROS over Chiba prefecture are shown in Fig. 1a. During the analyzed period of Chiba-Campaign 2015, surface NO_2 concentration was monitored at a total of 100 APMS sites and 27 RAPMS sites. The chemiluminescent method using ozone (JIS B 7953) is recommended for measuring NO_2 , and most sites used this method, although two APMS sites and one RAPMS site used the Saltzman reagent. Chemiluminescent methods using molybdenum converters could suffer from interference from NO_z (defined as all reactive oxygenated nitrogen except NO_x); however, the effect is probably small (Kondo et al. 2008). The observation data from AEROS are validated by data quality checking, and then are distributed as open data. Among these AEROS sites, sites located within 24 km (the maximum horizontal distance measured by MAX-DOAS, see Sect. 2.2) of Chiba University were used for the analysis.

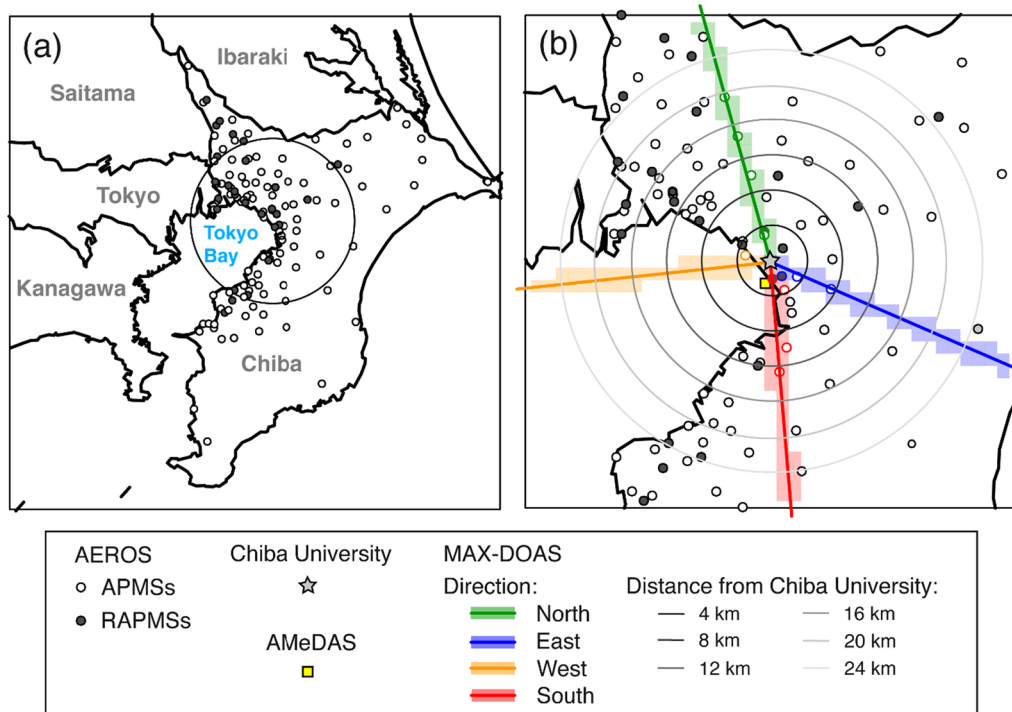


Fig. 1 Locations of ground-based observations by AEROS and 4AZ-MAX-DOAS. **a** Map of Kanto region, Japan and locations of ground-based observation by AEROS over Chiba prefecture. **b** Enlarged map around Chiba University, which is the center of 4AZ-MAX-DOAS. Circles are drawn at 4 km radius intervals from Chiba University up to 24 km (maximum horizontal distance observed by MAX-DOAS). AEROS observation sites are colored white (APMSs) and dark gray (RAPMSs)

2.2 MAX-DOAS observation

Aloft observations using MAX-DOAS systems (Irie et al. 2008, 2011, 2015, 2019, 2021) have been conducted at Chiba University (35.63°N, 140.10°E, 21 m a.s.l.), Chiba, Japan. The MAX-DOAS methodology is based on the differential optical absorption spectroscopy (DOAS) method, which quantitatively detects narrow-band absorption by trace gases by applying the Beer-Lambert law (Platt and Stutz 2008). Following on from the pioneering studies of Hönninger and Platt (2002) and Hönninger et al. (2004), various instruments and algorithms have been developed worldwide for MAX-DOAS. The MAX-DOAS system at Chiba University has participated in the Cabauw Intercomparison Campaign of Nitrogen Dioxide measuring Instruments (CINDI) (Roscoe et al. 2010) and CINDI-2 (Kreher et al. 2020). The MAX-DOAS system in this study uses the Maya2000Pro spectrometer (Ocean Optics, Inc.) with a 25 μm slit placed in a temperature-controlled enclosure to record high-resolution spectra (full width at half maximum: 0.3–0.4 nm; oversampling: 3–4) in the wavelength range 310 to 515 nm. Measurement was performed at five off-axis elevation angles (e.g., 2°, 3°, 4°, 6°, and 8°) and at a single reference elevation angle,

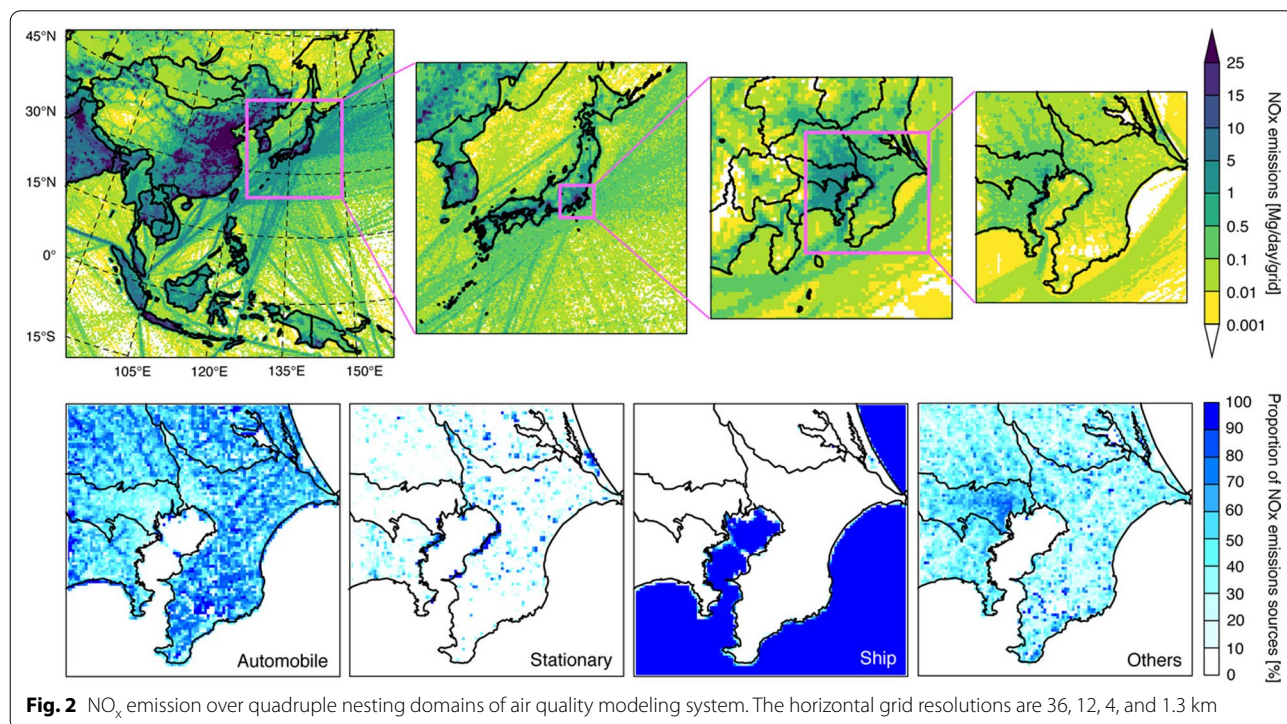
with 70° used instead of 90° to avoid saturation of the spectral intensity due to the effect of direct sunlight. The elevation angle setting was considered fully in calculating the differential air mass factors for the vertical profile retrieval (e.g., Irie et al. 2011, 2015). Thus, the choice of either 70° or 90° was not critical. Although the original observation by MAX-DOAS targets a single direction, four different azimuth-viewing MAX-DOAS (4AZ-MAX-DOAS) instruments are used at Chiba University to cover the cardinal compass points simultaneously: north (347°E), east (118°E), west (265°E), and south (175°E). The location of Chiba University and the directions covered by the 4AZ-MAX-DOAS are shown in Fig. 1b. To derive near-surface concentrations, we used our retrieval algorithm called JM2 (Japanese MAX-DOAS profile retrieval algorithm, version 2) (Irie et al. 2008, 2011, 2015). JM2 uses the recorded high-resolution UV–visible spectra from 310 to 515 nm to perform spectral analysis by the DOAS method (Platt and Stutz 2008) by spectral fitting using the nonlinear least-squares method and subsequent vertical profile retrievals using the optimal estimation method, which allows us to retrieve lower-tropospheric vertical profile information for eight quantities including NO_2 . The fitting

windows (460–490 nm) and cross section data were identical to those described by Irie et al. (2011, 2015). The minimum detectable level of the NO_2 differential slant column density estimated from the fitting residual was about 10^{14} molecules cm^{-2} . In the retrieval, off-axis elevation angles are limited to below 10° to minimize the systematic error in the oxygen collision complex fitting results (Irie et al. 2015). This limitation improves the observation of the PBL owing the loss of sensitivity to extinction at high altitudes, where clouds rather than aerosols predominate. Clouds in the PBL were excluded by choosing data with a relative humidity over water (RHw) of less than 90% for the 0–1 km layer. The RHw was estimated from the MAX-DOAS H_2O data retrieved using a fitting window of 495–515 nm and the National Centers for Environmental Prediction (NCEP) pressure and temperature reanalysis data (2.5° grid and 6-hourly). Thus, our MAX-DOAS system was optimized to retrieve information on aerosols and trace gases in the PBL rather than in the whole tropospheric column. For the vertical profile retrieval, full consideration is given to the elevation angle setting when computing differential air mass factors (Irie et al. 2011, 2015). There are usually about 1–2 degrees of freedom of signals for trace gas vertical profiles retrieved in this way. With the retrieved vertical profiles, the present study analyzed the layer at 0–1 km, which corresponds to the lowermost layer with the highest sensitivity

owing to the longest light path in profiles retrieved by JM2. In the horizontal direction, the viewed horizontal distance in each direction is up to 24 km. In Fig. 1b, horizontal scales up to 24 km from Chiba University are shown at 4 km intervals as a visual guide. The total uncertainties, including random and systematic errors, in a single measurement were estimated as 15% for NO_2 (Irie et al. 2011), and the estimated systematic error was based on additional retrievals of JM2 aerosol retrieval uncertainties of as large as 30% (Irie et al. 2008, 2011). Previous studies also highlighted the use of MAX-DOAS measurements. In Europe, MAX-DOAS and regional air quality model simulations have been compared over Paris (Shaiganfar et al. 2015) and the Netherlands (Vlemmix et al. 2015), and four sites over Europe with six model ensemble experiments (Bleischmidt et al. 2020). The MAX-DOAS network in Russia and Asia (MADRAS) elucidated the long-term trends in NO_2 pollution status (Kanaya et al. 2014; Choi et al. 2021), and these trends have been compared with model results (Kanaya et al. 2014). In this study, to improve the fine-scale horizontal coverage by 4AZ-MAX-DOAS measurements (Irie et al. 2021), kilometer-scale regional air quality modeling was conducted.

2.3 Regional air quality modeling

The regional air quality modeling was based on Community Multiscale Air Quality (CMAQ) version 5.2.1



released by the US Environmental Protection Agency (US EPA 2018). The simulation used quadruple nesting covering all of Asia, Japan, the extended Kanto region, and Chiba prefecture with horizontal grid resolutions of 36, 12, 4, and 1.3 km (Fig. 2). For comparison with observations, the domain with the finest resolution of 1.3 km was used. In the vertical direction, 44 nonuniform layers from the surface to 50 hPa were set to fully represent the stratosphere-to-troposphere transport (Mathur et al. 2017; Itahashi et al. 2020). The height of the lowermost layer, which corresponded to modeling surface level, was approximately 20 m. The meteorological input data were simulated by the Weather Research and Forecasting (WRF) model version 3.6.1 (Skamarock et al. 2008) to drive the CMAQ. The configuration of the WRF simulation was basically the same as our previous study (Itahashi 2018). For both longwave and shortwave radiation, the rapid radiative transfer model for global climate models (RRTMG) radiation scheme was used (Iacono et al. 2008), and the Morrison double-moment scheme (Morrison et al. 2009) and Grell convective parameterization (Grell and Devenyi 2002) were used for microphysics and cumulus parameterization. The cumulus scheme was not applied for domains of 4 and 1.3 km horizontal resolution. The Mellor-Yamada-Janjic scheme was used as the PBL option (Janjić 1994). The outermost domain of 36 km horizontal grid resolution was independently simulated with nudging for wind, temperature, and water vapor fields to NCEP/National Center for Atmospheric Research (NCAR) final analysis data for all vertical layers, where the analysis data has 1° spatial and 6 h temporal resolution (NCEP/NCAR 2021). For inner domains of 12, 4, and 1.3 km, the nudging was applied toward the Mesoscale Analysis (MANAL) by the Japan Meteorological Agency (JMA), which has a grid spacing of 5 km spatial and 3 h temporal variation.

The emission dataset was created using anthropogenic emissions from Hemispheric Transport of Air Pollution (HTAP) version 2.2 (Janssens-Maenhout et al. 2015) over Asia and the PM_{2.5} emission inventory for fiscal year 2015 (defined as April 2015 to March 2016) developed by the Ministry of Environment (MOE 2019) over Japan, biogenic emissions from the Model of Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther et al. 2012), emissions from biomass burning in the Global Fire Emissions Database (GFED) version 4.1 (van der Werf et al. 2017), emissions from 16 volcanoes in Japan from observational data obtained by the Japan Meteorological Agency (JMA 2021), and emissions from 33 degassing volcanoes in Russia, India, Indonesia, Philippines, Papua New Guinea, and Northern Mariana Islands constrained by satellite measurement (Carn et al. 2017). Because anthropogenic emissions from China have shown dramatic changes in recent

years (Irie et al. 2016; Itahashi et al. 2019), Chinese emissions from each sector were adjusted to the estimated values for 2015 (Zheng et al. 2018). The domestic emission inventory by the Ministry of Environment was created over Japanese MESH3 boundaries, which corresponded to approximate 1 km. NO_x (including the ratio of NO and NO₂), SO₂, ammonia (NH₃), CO, non-methane volatile organic compounds (NMVOC; 46 species), and primary PM_{2.5} (18 elements) were considered as hourly data for each month. For some sectors (e.g., automobiles), weekdays and weekends were also taken. Ship emissions were provided by the Sasakawa Peace Foundation (SPF), which were estimated based on the Automatic Identification System (AIS) for each day of the week and hour. The NO_x emissions used in this study are shown in Fig. 2. For the finest resolution of 1.3 km, the proportions of the main NO_x emission sources of automobiles, stationary sources (sum of industry and power plants), ships, and others (all emissions minus automobiles, stationary, and ships) are also shown in Fig. 2. Gas chemistry in the CMAQ was used for the SAPRC07 mechanism (Hutzell et al. 2011) and aerosol chemistry in the CMAQ was used for the aerosol mechanism with non-volatile primary organic aerosol (Simon and Bhawe 2012). The lateral boundary condition for the outermost domain was taken from the global model of the Model for Ozone and Related Chemical Tracers (MOZART) version 4 (Emmons et al. 2010), and the nested domains used the outer domains simulation results for the lateral boundary condition. The default CMAQ profile was used for the initial condition. The WRF and CMAQ simulations were conducted from 2 November 2015 in order to set a 1 week spin-up time for removing the effect of the initial conditions.

For assessing model performance, three statistical metrics were used: correlation coefficient (R), normalized mean bias (NMB), and normalized mean error (NME). These are defined as follows:

$$R = \frac{\sum_1^N (O_i - \bar{O})(M_i - \bar{M})}{\sqrt{\sum_1^N (O_i - \bar{O})^2} \sqrt{\sum_1^N (M_i - \bar{M})^2}} \quad (1)$$

$$\text{NMB} = \frac{\sum_1^N (M_i - O_i)}{\sum_1^N O_i} \quad (2)$$

$$\text{NME} = \frac{\sum_1^N |M_i - O_i|}{\sum_1^N O_i} \quad (3)$$

where N is the total number of paired observations (O) and models (M), and these averages are denoted as \bar{O} and \bar{M} , respectively.

3 Results and discussion

Section 3.1 presents the temporal variation of NO_2 concentration at the surface level using ground-based observation by AEROS, and Sect. 3.2 presents the temporal variation of aloft NO_2 concentration based on MAX-DOAS. The model evaluations are also presented. Section 3.3 examines the diurnal variation of NO_2 concentration at the surface and aloft. Section 3.4 considers the reason for episodic high concentrations of NO_2 based on the meteorological field. Finally, Sect. 3.5 provides the correspondence between surface and aloft NO_2 pollution as analyzed by the well-evaluated modeling simulation.

3.1 Temporal variation of surface NO_2 concentration

The overall modeling performance during Chiba-Campaign 2015 was evaluated using ground-based AEROS observation. A comparison of the surface observation by AEROS located within 24 km (maximum horizontal distance measured by MAX-DOAS, see Fig. 1) of Chiba University and the model at the surface level is shown as a scatterplot in Fig. 3. In this comparison, the corresponding grid for observational sites over the finest modeling domain of 1.3 km was used. During Chiba-Campaign 2015, surface NO_2 concentration around Chiba University ranged mostly within 30 ppbv (dense circles in Fig. 3), and sometimes exhibited high concentrations of up to 70 ppbv. The histogram in Fig. 3 showed that the model undercounted the observed feature between 20 and 30 ppbv and overcounted it over 30 ppbv. Over 20,000 pairs of observed and modeled hourly NO_2 concentrations, R of 0.50 showed a moderate linear correlation, NMB was +9.8%, and NME was 55.9%. It has been reported that R ranged from 0.51 to 0.92 and

NME ranged from 25 to 85% over China based on model results using three different emission datasets and their evaluation against the various surface observation categories (Liu et al. 2018). The Model Inter-Comparison Study for Asia (MICS-Asia) Phase III reported that R was 0.02–0.70 and NMB ranged between –44% and +36% (Kong et al. 2020). The model intercomparison study over Japan, J-STREAM, reported that R was 0.38–0.54 and NMB ranged between –19.4% and +14.7% over the Kanto region (Yamaji et al. 2020). The results of this study demonstrate that the model could generally capture the surface NO_2 concentration around Chiba University through Chiba-Campaign 2015.

For the purpose of detailed comparison using ground-based AEROS observations, the modeling performance at ground-based AEROS observations located along the path of 4AZ-MAX-DOAS measurements (see Fig. 1) was further evaluated. The temporal variations during Chiba-Campaign 2015 at these AEROS sites are shown in Fig. 4. Hereinafter, timeseries data are shown as local time (LT), which is defined as Universal Time Coordinate (UTC) + 9 h. In the north, east, west, and south directions, a total of 5, 3, 1, and 4 sites were located along the measurement paths of 4AZ-MAX-DOAS, respectively. These AEROS sites are named in order of closeness from Chiba University in each direction (see Fig. 4). High NO_2 concentration is defined as greater than 30 ppbv in this discussion, and is denoted by a yellow highlight at each site in Fig. 4. In the north direction (green in Fig. 4), R was around 0.5, NMB was within $\pm 25\%$, and NME was within 50%. At the N2 site, classified as an AEROS RAPMS site, the model performance was lower than that at other APMS sites in the north direction. Because the N1 and

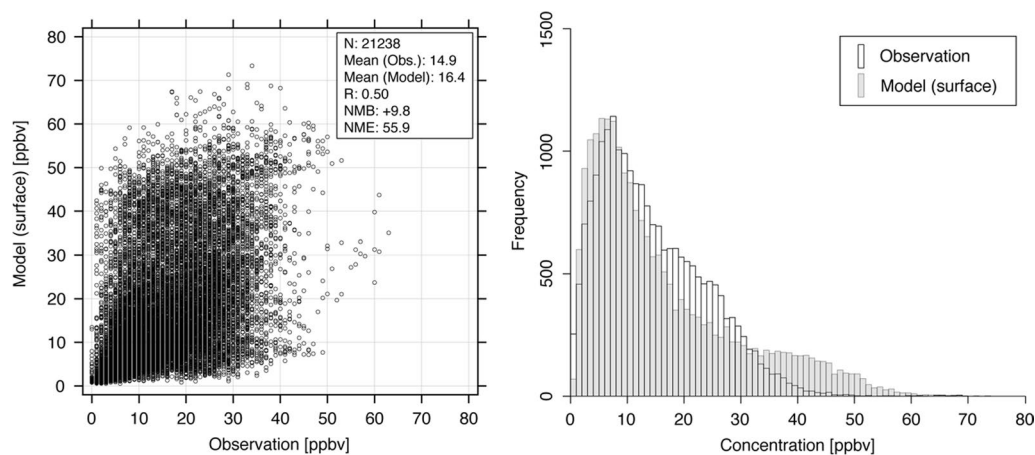
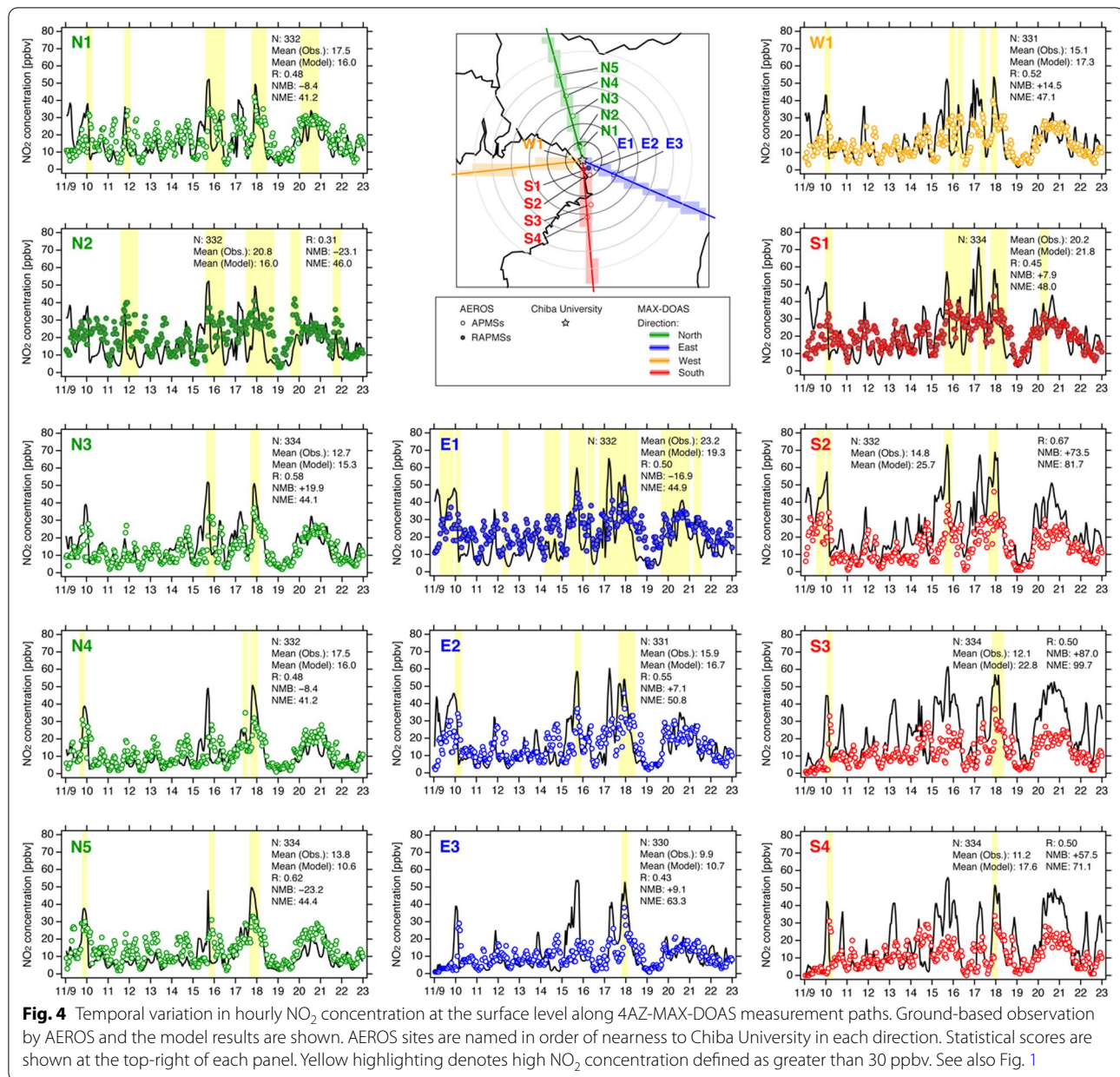


Fig. 3 (Left) Scatter-plot of hourly surface level NO_2 concentration during Chiba-Campaign 2015. (Right) Histogram for observations (white) and the model (gray). Ground-based observation by AEROS and the model are shown. AEROS sites located within 24 km of Chiba University (see, Fig. 1) are used



N2 sites are located close together, the modeling results were taken from the same grid point in this comparison. Compared with the modeling performance found at N1, all statistical scores were worse, and model underestimation was identified at N2. This result indicates that even when the finer horizontal resolution of 1.3 km was used for air quality modeling, it was difficult to capture local NO_2 pollution mainly caused by automobile sources. In the east direction (blue in Fig. 4), the comparison at the E2 and E3 sites, which are classified as AEROS APMS sites, was better than at the E1 site, which was classified as an AEROS RAPMS site, in terms of NMB and NME.

The model also underestimated NO_2 concentration at RAPMS sites, and this result in the east direction was similar to that found in the north direction. In the west direction (orange in Fig. 4), only a single APMS site close to Chiba University was available because this direction covers the Tokyo Bay area. The model performance at the W1 site was comparable to that at the N3 and E3 sites. In the south direction (red in Fig. 4), at the S1 site, which is classified as an AEROS RAPMS site, located near Chiba University, the model performance was mostly comparable to that found in the other directions. In contrast, the model generally tended to overestimate the surface NO_2

pollution at the other sites S2, S3, and S4 in the south direction. At the S2, S3, and S4 sites, the model overestimation was suggested by NMB exceeding +50%. As indicated by the temporal variation at S2, S3, and S4, the model overestimation was seen during 15–16, 17–18, and 20–21 November 2015, when NO_2 concentrations were generally high. Because of this tendency of the model for overestimation in the south direction, model underestimation at RAPMS sites as found in the north and east directions was not detected. Based on these detailed comparisons of the temporal variation at AEROS sites located along the path of 4AZ-AMAX-DOAS, it was found that the model had some difficulties with underestimation when capturing local air pollution caused by automobile sources at RAPMSs sites in the north and east direction and overestimation in the south direction. Overall, the model generally captured the temporal variation of surface NO_2 pollution around Chiba University along the path of 4AZ-MAX-DOAS.

From the temporal variation shown in Fig. 4, surface NO_2 concentration typically showed diurnal variation with minima during the daytime and maxima during the nighttime. During Chiba-Campaign 2015, elevated NO_2 concentrations were detected on 9, 15, 16, 17–18, and

20–21 November 2015, as distinguished by yellow highlights in Fig. 4. For the elevated NO_2 concentrations on 9, 15, and 16 November, a brief peak was found during the nighttime, whereas 17–18 and 20–21 November exhibited continuously higher concentrations over the whole day as the diurnal variations changed such that there was no daytime minima. Diurnal variation and episodic peaks of NO_2 concentration are respectively discussed in Sect. 3.3 and 3.4.

3.2 Temporal variation of aloft NO_2 concentration

The temporal variation of aloft NO_2 concentration obtained by comparing 4AZ-MAX-DOAS and model results averaged over 0–1 km are shown in Fig. 5. The horizontal measurement distance of MAX-DOAS is also plotted. Since the horizontal distance viewed by MAX-DOAS is dependent on aerosol pollution, it varied from 4 to 24 km, with longer distances on clean days, such as during lower NO_2 concentration periods on 11–13 November 2015. The modeled NO_2 concentration within 24 km from Chiba University from model results is shown as the range in Fig. 5, and statistical analysis of the comparison with MAX-DOAS observation was conducted using the data within the observed horizontal

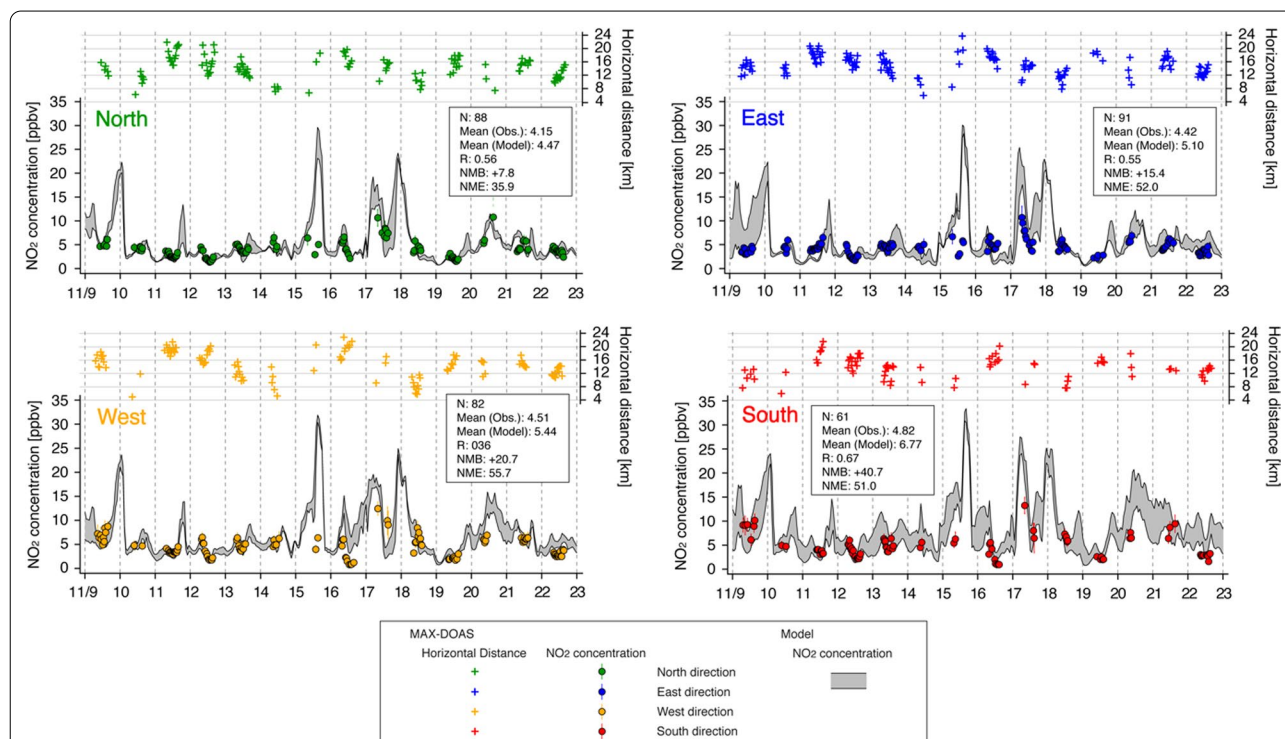


Fig. 5 Temporal variation of NO_2 concentration by MAX-DOAS and model averaged over 0–1 km (each direction). The MAX-DOAS measured horizontal distance is also plotted. The range of model results showed simulated NO_2 concentration within 4–24 km from Chiba University in each direction. Statistical scores are shown in the inset, and 1 h averaged MAX-DOAS observation and corresponded horizontal distance to observation were used for this analysis

distance to match the view by MAX-DOAS. From the range of modeled NO_2 concentration within 4–24 km from Chiba University, the modeled NO_2 pollution range within the horizontal direction of MAX-DOAS was generally small except for pollution events. This result implies that NO_2 pollution around Chiba University was generally dominated by regional-scale broad pollution over the greater Tokyo area. In the north, east, and west directions, the range of modeled NO_2 concentration was small except for high concentration events on 9, 15, 16, 17–18, and 20–21 November 2015. Because MAX-DOAS observation was limited to during the daytime, the higher NO_2 concentrations at night on 9, 15, and 16 November were not measured. However, the cases of episodic high concentration on 17–18 and 20–21 November, which were characterized by continuous high concentration, corresponded well between MAX-DOAS and aloft model results. The modeling performance for the north, east, and west directions was R in the range 0.36–0.56, NMB around +20%, and NME around 50%. These results are comparable with the surface results. In the south direction, the range of modeled NO_2 concentration was also large during episodic NO_2 pollution events, but also showed larger variation throughout Chiba-Campaign 2015 compared with the other directions. As found in the surface comparison, the model also tended to overestimate aloft NO_2 concentration in the south direction. Although the value of NMB was larger in the south direction than in the other directions, NME was comparable and R was better compared with the other directions. In summary, it was validated that the model captured aloft NO_2 concentration through comparison with 4AZ-MAX-DOAS observations.

3.3 Diurnal variation of surface and aloft NO_2 concentrations

Sections 3.1 and 3.2 evaluated the temporal variations of surface and aloft NO_2 concentrations and confirmed that the modeling system generally captured surface and aloft NO_2 pollution. The diurnal variations are further analyzed here in Sect. 3.3. The surface and aloft diurnal variations in NO_2 concentration averaged during Chiba-Campaign 2015 for the direction observed by 4AZ-MAX-DOAS are presented in Fig. 6. From the comparison at the surface level (left side of Fig. 6), the model could capture the observed diurnal variation, which consisted of a morning peak, subsequent decline during the daytime, and then an evening peak. In detail, the observed values showed morning maxima during 8 to 9 AM whereas the model showed morning maxima at the sunrise time of 6 AM. In contrast, for the evening peak seen after the sunset time of 5 PM, the timing of maxima was well reproduced by the model, particularly in the north and east

directions. As found from the analysis of temporal variations shown in Fig. 3, the model tended to overestimate in the south direction, and this modeling overestimation was identified during the nighttime from the analysis of diurnal variation at the surface level. Apart from this difficulty, the model captured the diurnal variation of NO_2 concentration at the surface level. The evening peak was a good match between surface observation and the model, whereas the morning peak showed a slight difference in timing. This suggests that investigation into the chemical mechanisms related to NO_x and sunlight is needed in future research. In addition, because the nighttime NO_2 concentration is also related to NO and O_3 reproduction, this reaction and related meteorological variation must be examined to obtain the morning maxima at sunrise.

In the comparison of aloft NO_2 concentration, all MAX-DOAS observations are plotted as light colors, and 1 h averaged data are plotted as dark colors (right side of Fig. 6). The hourly averages at 7 AM and 4 PM were not well counted (1–3 times through Chiba-Campaign-2015) and hence the 1 h averaged plot is not shown for these times. All 4AZ-MAX-DOAS showed a slight decrease in concentration during the daytime around noon, and the model reproduced these observations for all directions. As with the surface concentration in the south direction, the aloft NO_2 concentration in the south direction was also overestimated. However, the temporal variation consisting of a drop around noon was captured. Based on this comparison, it was concluded that the diurnal variations were well reproduced by the model at both the surface and aloft. To our knowledge, there has been limited research on the diurnal variation in surface and aloft NO_2 concentrations. The findings from CINDI revealed that the diurnal variation during summertime had a U-shaped curve with morning and evening maxima and daytime minima (Irie et al. 2011). At Yokosuka, which is about 30 km south of the Tokyo metropolitan area, MAX-DOAS and surface monitoring were compared (Kanaya et al. 2014). The daytime decrease was also recorded for both the MAX-DOAS and surface observations in these studies, and this was consistent with the findings in the present study. The present study extended comprehensive analyses for verifying the air quality modeling performance at both the surface and aloft using surface observation and MAX-DOAS. Considering the better modeling performance in the north and east directions than in the south direction, one possible reason for the difficulty in modeling may be issues in the emission estimates. The coastline of the Tokyo Bay area has many stationary sources (industry and power plants) (Fig. 2), which have relatively high stack heights, and the impact of these may be observed in the south

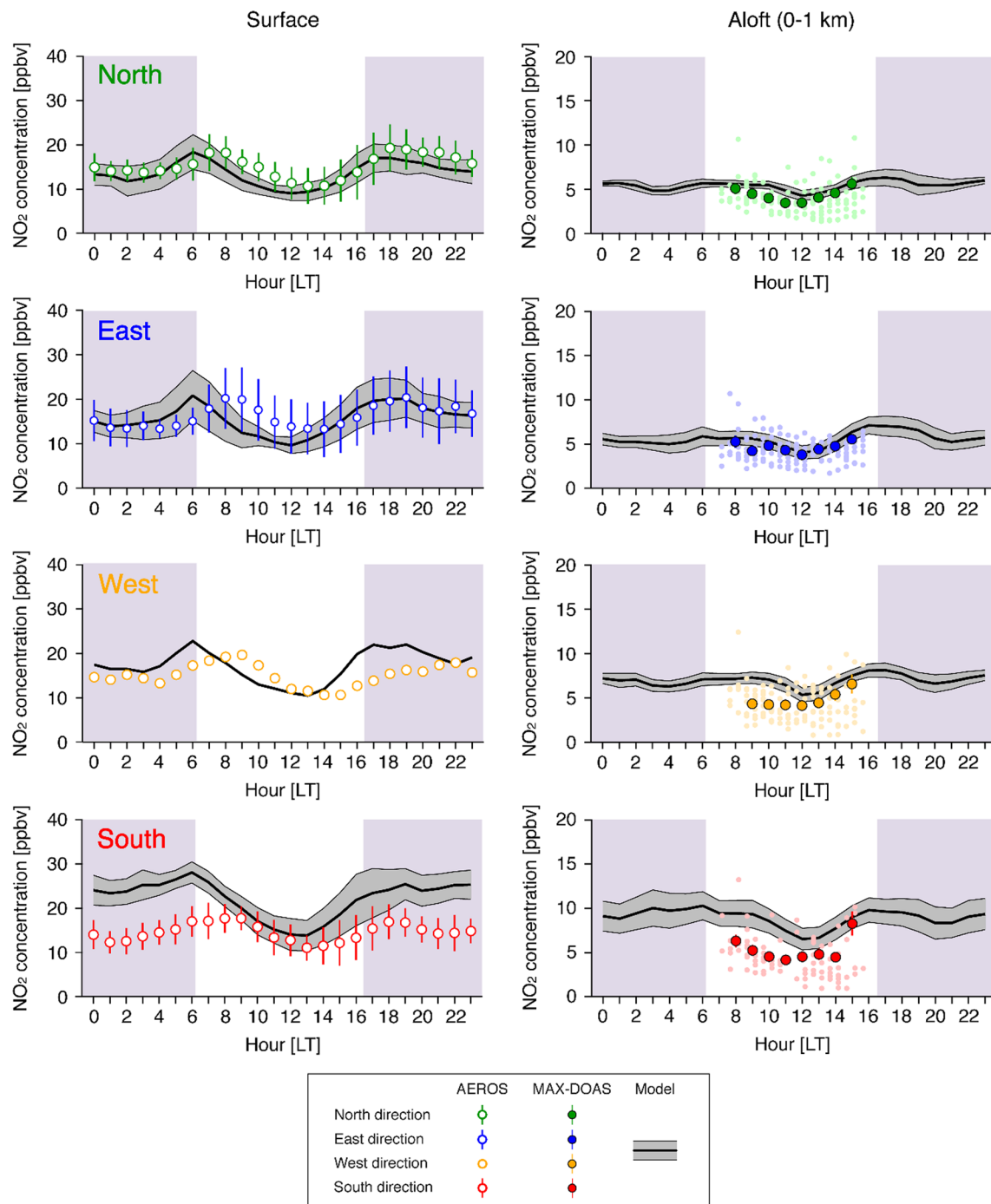


Fig. 6 Diurnal variation of NO_2 concentration at (left) surface and (right) aloft. Surface data are taken from AEROS sites located along 4AZ-MAX-DOAS directions and model results at surface level. Aloft data is taken from MAX-DOAS with light colors for all data and thick colors for 1 h averaged data, and model results averaged over 0–1 km. Dark blue shading indicates nighttime

direction from Chiba University. Because NO_2 concentration was overestimated both at the surface and aloft, the emission amount itself from stationary sources might be overestimated. Moreover, chemical and

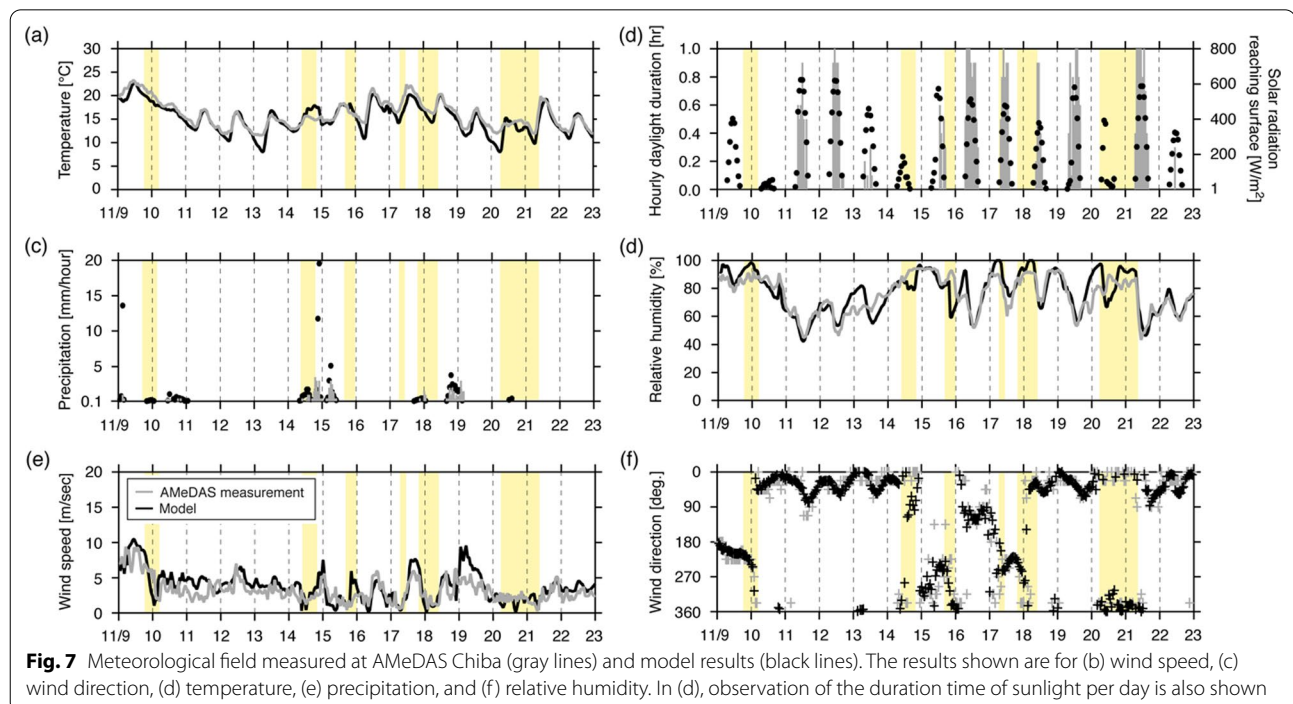
physical processes related to NO_x emissions from point sources could be related. Other issues may stem from the ship emissions, which dominated NO_x emissions over the ocean zone. These points should be explored

in the future to improve the modeling performance around the greater Tokyo area.

3.4 Episodic peaks in NO₂ concentration

The diurnal variation presented in Sect. 3.3 was the averaged characteristics during Chiba-Campaign 2015. As shown in the temporal variation presented in Figs. 4 and 5, some episodic peaks in NO₂ concentration were detected on 9, 15, 16, 17–18, and 20–21 November 2015. Here in Sect. 3.4, the reason for the increased NO₂ concentration is considered based on the meteorological field. The meteorological parameters measured by the Automated Meteorological Data Acquisition System (AMeDAS) of the Chiba special area meteorological observatory, which is located near Chiba University (see Fig. 1), and its modeling performance are shown in Fig. 7. In this figure, high concentrations of NO₂ of more than 20 ppbv are also indicated by yellow highlights based on the averaged concentration of APMS sites located along the path of 4AZ-MAX-DOAS for clarity (see also Fig. 4). The meteorological field was well reproduced by the modeling system throughout the Chiba-Campaign 2015. The air temperature (Fig. 7a), ranged from 10 to 20 °C with clear diurnal variation of daytime maxima. During Chiba-Campaign 2015, there were drops in temperature on 13 and 20 November 2015. The observed hourly

daylight duration and modeled solar radiation reaching the surface level are plotted in Fig. 7b. These indicate the weather conditions, and the results show that 10, 13–14, and 20 November 2015 had cloudy conditions. Rainy conditions were clarified by analyses of precipitation as shown in Fig. 7c and relative humidity as shown in Fig. 7d. During Chiba-Campaign 2015, precipitation events occurred on 9, 10, 14, 15, and 19 November. In these precipitation events, relative humidity also exhibited high values of close to 100%. Wind components are presented as wind speed in Fig. 7e and wind direction in Fig. 7f. The wind speed was generally around 5 m/s and sometimes reached 10 m/s, and it was found that the high NO₂ concentration (yellow highlight) correlated well with weak wind speed. Throughout Chiba-Campaign 2015, the prevailing wind was in the north (0°) to east (90°) direction. However, wind changes to the west (270°) or northwest direction were found which corresponded to intense emission sources around the Tokyo Bay area. Therefore, the stagnant conditions in the transport of air mass from the dense emission sources around the Tokyo Bay area are thought to be the reason for increased NO₂ concentration in terms of meteorological conditions. The highest NO₂ concentration was seen on 17–18 November and the longest event with continuously high NO₂ concentration was found during 20–21 November with different diurnal patterns; these are further discussed in Sect. 3.5.



3.5 Correspondence between surface and aloft NO₂ pollution

Section 3.1 and 3.2 validated the modeling at the surface and aloft and Sect. 3.3 evaluated the diurnal variation at the surface and aloft by combining surface AEROS and aloft MAX-DOAS measurements. Section 3.4 examined the meteorological field causing episodic high NO₂ concentrations, and stagnant conditions with a westerly wind direction can be considered as a reason for the high concentrations. Finally, here in Sect. 3.5, the relationship between surface and aloft NO₂ pollution was analyzed based on well-evaluated modeling simulation results. The hourly NO₂ concentration averaged during Chiba-Campaign 2015 is plotted at each hour for surface and aloft in Fig. 8. Throughout the day, high NO₂ concentration was found over the Tokyo Bay area both at the surface and aloft. As seen for the diurnal variation reported in Fig. 6, the range of diurnal variation was larger at the surface than aloft. At the surface, high concentrations of greater than 20 ppbv (shown as orange to red in Fig. 8) were distributed over eastern Tokyo and Kanagawa prefecture and western Chiba prefecture, but these areas of high concentration were limited to only along the coastline of Tokyo Bay area from 10–14 LT. Compared with the features of the spatial distribution pattern of the high concentration area found at the surface, a moderately high concentration (shown as green in Fig. 8) was continuously seen aloft through the day. In order to find the relationship between the surface and aloft NO₂ concentrations, linear regression was performed using all grid point data over this finest modeling domain for surface and aloft NO₂ concentrations. The results are listed in Table 1 with domain averaged concentration (mean \pm standard deviation), slope, intercept, and R. The results show clear linearity with R exceeding 0.918 through the day. The value of slope in the linear regression was around 0.4 during the nighttime and reached around 0.55 during the daytime. This result indicates that the aloft concentration was linearly scaled to 0.4 times the surface concentration during the nighttime and also linearly scaled to 0.55 times the surface level around noon (10 LT to 14 LT). The higher fraction during the daytime was due to the well-mixed air pollution within the PBL. This linear relationship between surface and aloft NO₂ concentrations through the day was a key finding of this study; and this relation varied with a lower scaling value of around 0.4 during the nighttime and a higher value reaching 0.55 during the daytime. The same analyses listed in Table 1 but limited to only over land area were conducted to see the relation over the land area where the NO_x emissions were more intense compared with those over the ocean area (Fig. 2). The results are listed in Table 2. The relationships between the surface and aloft NO₂ concentration were

mostly similar to that for the whole modeling domain with slightly a smaller slope, slightly higher intercept, and a higher correlation suggested by R. The diurnal variation of the scaled value around 0.4 (minimum value of 0.348) during the nighttime and a higher value reaching 0.55 during the daytime were also similar. During the daytime, the observed correspondence could also be calculated directly from the ground-based observation and MAX-DOAS (Fig. 6). The scaled values calculated at each hour were 0.227–0.425, 0.211–0.384, 0.220–0.618, 0.263–0.627 for the north, east, west, and south directions, respectively. The variation of the scaled values were larger than the model estimated values (Tables 1 and 2). The lower scale factors were mostly found early in the morning (7–9 LT), when the observed concentration reached a peak, whereas the model missed this observed peak, as discussed in Sect. 3.3. Considering this problem with the model, we concluded that the modeled scale factor between the surface and aloft NO₂ generally corresponded to the observed results except early morning.

The previous discussion focused on the diurnal variation. Because episodic high concentrations were found during Chiba-Campaign 2015, the daily average surface and aloft NO₂ concentrations over the modeling domain were further analyzed in order to investigate the temporal change in the linear relationship between surface and aloft. The daily average was based on the average over 0–23 LT. The spatial mappings of each day are plotted in Fig. 9, and the period average during Chiba-Campaign 2015 are also plotted at the bottom-right corner of Fig. 9. NO₂ concentration showed day-to-day variation and the spatial distribution pattern between surface and aloft were in generally good agreement. For example, in the case of high NO₂ concentration on 9 November 2015 as suggested by temporal variation (Figs. 4 and 5), the higher NO₂ concentration stretched into northern Chiba and up to Ibaraki and Saitama prefectures at the surface level, and this feature was also seen aloft. On 9 November 2015, a southerly wind close to 10 m/s prevailed (Fig. 7). In another case of high NO₂ concentration during 20–21 November 2015, higher NO₂ concentrations were found over a broad area of Kanagawa prefecture, eastern Tokyo, and western Chiba prefecture on these days at the surface, and the aloft concentration also showed a similar spatial distribution. To confirm these characterizations of the surface and aloft NO₂ pollution, linear regression was performed for each day and period average in the same way as for the diurnal variation listed in Table 1. The result for daily and period averages are listed in Table 3. The daily averaged concentrations varied from 6 to 15 ppbv during Chiba-Campaign 2015. In spite of this variation in concentration, comparison of surface and aloft found an R exceeding 0.924, which suggested

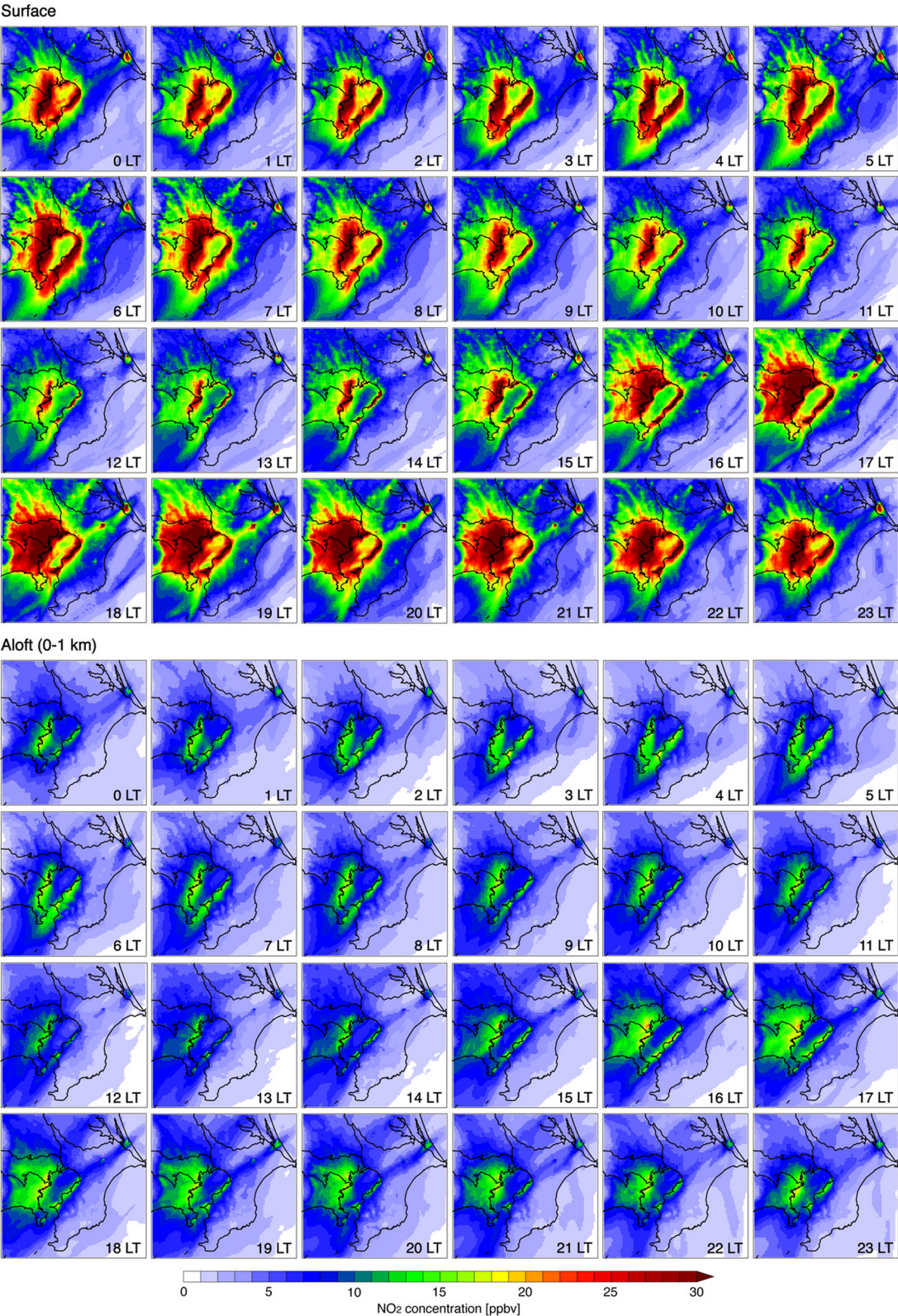


Fig. 8 Spatial distribution of modeled hourly NO₂ concentration at surface and aloft (averaged over 0–1 km). Results are averages over the Chiba-Campaign 2015

Table 1 Hourly correspondence between modeled surface and aloft (averaged over 0–1 km) NO₂ concentrations

LT	Surface	Aloft (0–1 km)	Slope	Intercept	R
0	8.73 ± 7.00	4.37 ± 2.88	0.398	0.897	0.966
1	8.33 ± 6.68	4.15 ± 2.82	0.409	0.753	0.968
2	8.19 ± 6.72	4.03 ± 2.91	0.418	0.604	0.966
3	8.31 ± 6.91	4.00 ± 3.03	0.421	0.503	0.959
4	8.72 ± 7.04	4.08 ± 3.09	0.415	0.459	0.945
5	9.65 ± 7.45	4.28 ± 3.14	0.391	0.511	0.926
6	10.99 ± 8.10	4.65 ± 3.26	0.370	0.577	0.918
7	10.57 ± 7.44	4.67 ± 3.11	0.391	0.536	0.933
8	9.50 ± 6.59	4.64 ± 3.08	0.447	0.394	0.957
9	8.63 ± 6.13	4.63 ± 3.10	0.493	0.381	0.972
10	7.84 ± 5.80	4.55 ± 3.17	0.531	0.382	0.975
11	7.15 ± 5.42	4.39 ± 3.12	0.558	0.397	0.969
12	6.72 ± 4.95	4.23 ± 2.92	0.567	0.417	0.962
13	6.76 ± 4.89	4.26 ± 2.86	0.561	0.462	0.958
14	7.12 ± 5.19	4.44 ± 2.98	0.548	0.536	0.954
15	8.12 ± 6.04	4.91 ± 3.32	0.523	0.654	0.951
16	10.04 ± 7.83	5.52 ± 3.87	0.469	0.809	0.950
17	11.24 ± 8.96	5.64 ± 3.99	0.424	0.881	0.952
18	11.44 ± 8.85	5.58 ± 3.81	0.408	0.913	0.947
19	11.61 ± 8.66	5.49 ± 3.53	0.383	1.038	0.940
20	11.47 ± 6.34	5.32 ± 3.28	0.371	1.063	0.942
21	10.79 ± 7.93	5.03 ± 3.17	0.380	0.933	0.950
22	9.82 ± 7.69	4.71 ± 3.11	0.388	0.902	0.959
23	9.23 ± 7.60	4.53 ± 3.10	0.393	0.903	0.964

Total grid numbers in finest modeling domain are 11,025

Table 2 Hourly correspondence between modeled surface and aloft (averaged over 0–1 km) NO₂ concentrations over land area

LT	Surface	Aloft (0–1 km)	Slope	Intercept	R
0	10.54 ± 6.73	5.10 ± 2.68	0.384	1.047	0.964
1	9.71 ± 6.41	4.66 ± 2.60	0.393	0.847	0.966
2	9.29 ± 6.47	4.36 ± 2.67	0.399	0.654	0.966
3	9.36 ± 6.72	4.22 ± 2.75	0.393	0.547	0.957
4	9.89 ± 6.92	4.26 ± 2.78	0.380	0.497	0.945
5	11.20 ± 7.47	4.49 ± 2.89	0.360	0.451	0.932
6	13.16 ± 8.22	4.98 ± 3.07	0.348	0.406	0.931
7	12.64 ± 7.39	5.10 ± 2.90	0.370	0.419	0.945
8	11.06 ± 6.35	5.15 ± 2.81	0.426	0.435	0.963
9	9.81 ± 5.87	5.22 ± 2.85	0.475	0.563	0.978
10	8.82 ± 5.60	5.20 ± 2.97	0.520	0.613	0.980
11	8.03 ± 5.21	5.07 ± 2.93	0.547	0.676	0.974
12	7.61 ± 4.74	4.91 ± 2.67	0.542	0.785	0.963
13	7.89 ± 4.78	5.00 ± 2.61	0.520	0.892	0.955
14	8.56 ± 5.15	5.29 ± 2.73	0.503	0.979	0.951
15	10.08 ± 6.12	5.94 ± 3.09	0.478	1.115	0.949
16	12.95 ± 8.02	6.78 ± 3.67	0.436	1.134	0.953
17	14.78 ± 9.13	6.95 ± 3.85	0.406	0.948	0.963
18	14.83 ± 8.90	6.74 ± 3.65	0.393	0.903	0.957
19	14.74 ± 8.63	6.55 ± 3.34	0.368	1.115	0.952
20	14.26 ± 8.43	6.24 ± 3.15	0.357	1.143	0.958
21	13.13 ± 8.01	5.83 ± 3.09	0.371	0.960	0.960
22	12.01 ± 7.73	5.48 ± 3.03	0.379	0.927	0.965
23	11.23 ± 7.52	5.24 ± 2.98	0.383	0.936	0.968

The total number of grid cells in the finest modeling domain of land area is 6234

clearly linear correspondence between surface and aloft NO₂ concentrations through the Chiba-Campaign 2015. Although the intercept of the linear regression ranged between 0.0 and 0.75, it was found that the slope ranged within 0.4–0.5 except on 16 November 2015 when it was 0.37. This result indicates that the aloft NO₂ concentration is linearly scaled to 0.4–0.5 times the surface NO₂ concentration on both clean and polluted days. Similar analyses to those shown in Tables 1 and 2, but only for the land area were conducted for the daily averaged concentrations. The result are listed in Table 4. As shown in Table 2, the results only focusing on the land area were similar with a slightly lower slope, whereas the slope was 0.406 on 16 November 2015 over the land area. We concluded that the daily-averaged aloft NO₂ concentration was also linearly scaled to 0.4–0.5 times the surface NO₂ concentration during clean and polluted days over the land area in Chiba-Campaign 2015.

Finally, the vertical profile of NO₂ concentration from the model simulation is shown in Fig. 10. This curtain plot shows the vertical structure for NO₂ concentration

during the Chiba-Campaign 2015 with modeled PBL height at the grid point of Chiba University. The PBL height showed variation from the near surface up to 1.0 km with a general diurnal profile featuring daytime maxima. As discussed for temporal variations in the surface and aloft NO₂ concentrations, high NO₂ concentration events were found on 9, 15, 16, 17–18, and 20–21 November 2015. The vertical profiles show that high NO₂ concentrations (red in Fig. 10) were mostly limited within 0.2 km, and also within the PBL. In the vertical direction, NO₂ concentrations were lower than 5 ppbv (light blue in Fig. 10) above 0.5 km, even though PBL height sometimes reached up to 1.0 km. Note the highest NO₂ concentration on 17–18 November 2015 and the longest continuous NO₂ concentration during 20–21 November 2015. During these events, the PBL height was close to the near-surface level. Taking into consideration analyses of the meteorological field, as shown in Fig. 7, the stagnant air mass with weak wind speed and cloudy conditions with a lower PBL close to the surface were possible factors causing the increased NO₂ concentration. Through

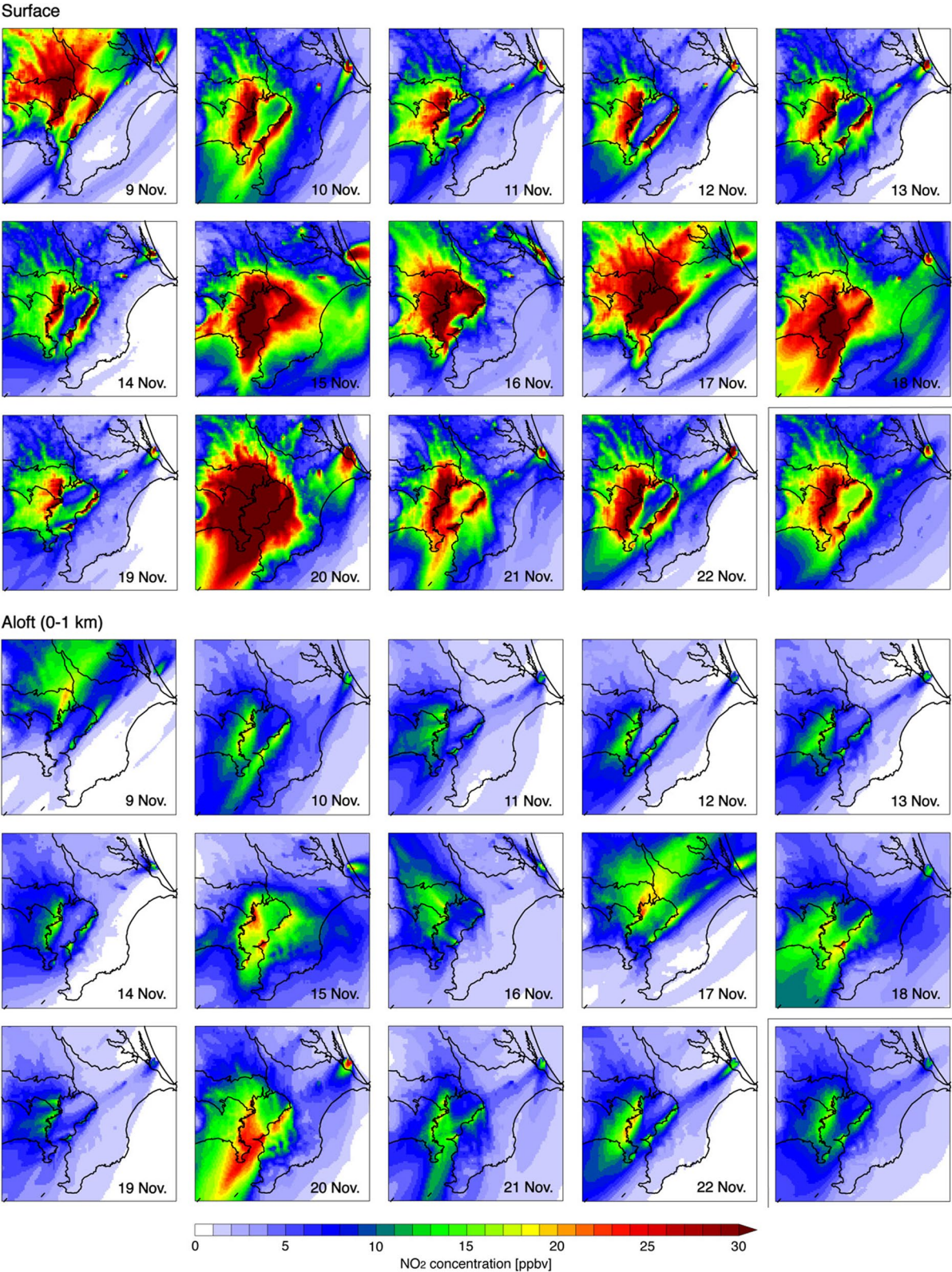


Fig. 9 Spatial distribution of modeled daily NO₂ concentration at surface and aloft (averaged over 0–1 km). The right-bottom panel shows the period average over the Chiba-Campaign 2015

Table 3 Daily/period averaged correspondence between modeled surface and aloft (averaged over 0–1 km) NO₂ concentration

Date	Surface	Aloft (0–1 km)	Slope	Intercept	R
9 November 2015	9.49 ± 9.21	4.65 ± 4.69	0.483	0.061	0.949
10 November 2015	9.04 ± 6.87	5.15 ± 3.50	0.486	0.752	0.954
11 November 2015	6.13 ± 6.00	3.46 ± 3.03	0.478	0.530	0.946
12 November 2015	5.97 ± 6.31	3.15 ± 3.16	0.469	0.361	0.933
13 November 2015	6.61 ± 6.58	3.40 ± 3.28	0.464	0.329	0.932
14 November 2015	6.17 ± 6.26	3.28 ± 3.17	0.478	0.329	0.944
15 November 2015	13.36 ± 8.65	6.62 ± 4.23	0.470	0.332	0.961
16 November 2015	9.30 ± 8.51	4.17 ± 3.40	0.371	0.714	0.928
17 November 2015	12.11 ± 10.11	6.19 ± 5.16	0.483	0.347	0.946
18 November 2015	12.97 ± 7.66	6.54 ± 3.96	0.496	0.107	0.959
19 November 2015	5.50 ± 5.44	2.95 ± 2.62	0.448	0.485	0.927
20 November 2015	15.48 ± 12.15	7.34 ± 6.22	0.474	−0.001	0.926
21 November 2015	8.97 ± 7.13	4.48 ± 3.24	0.425	0.674	0.935
22 November 2015	7.81 ± 7.81	4.01 ± 3.90	0.461	0.409	0.924
Period average	9.20 ± 6.74	4.67 ± 3.06	0.436	0.665	0.961

The total number of grid cells in the finest modeling domain is 11025

the validated modeling performance, the vertical NO₂ profile with its important connection to the PBL height was clarified. Because high NO₂ concentration was limited to below 0.5 km, the corresponding scale factors between the surface and aloft (0–1 km) were mainly 0.4–0.5 despite the dynamic variation of the PBL height. In terms of the scale factor between the surface to aloft (0–1 km), the lower scale factors were observed in early morning (7–9 LT). The model showed a slight difference to capture this timing, and furthermore investigation into the chemical mechanisms related to NO_x and sunlight is suggested to improve the modeling diurnal variation.

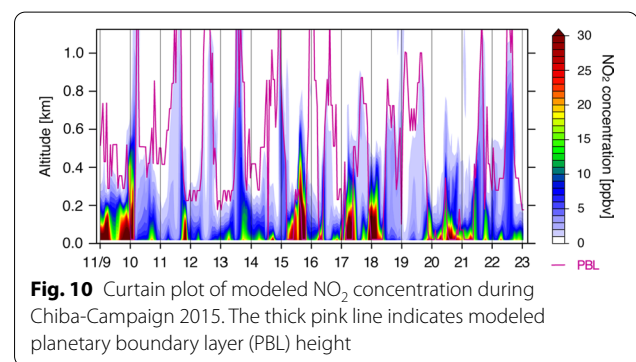
4 Conclusions

Surface and aloft NO₂ concentrations were extensively investigated by comprehensive analyses of the ground-based observation network of AEROS and aloft MAX-DOAS measurement, and bridged by an air quality modeling system with a horizontal grid resolution of

Table 4 Daily/period averaged correspondence between modeled surface and aloft (averaged over 0–1 km) NO₂ concentration over land area

Date	Surface	Aloft (0–1 km)	Slope	Intercept	R
9 November 2015	13.16 ± 9.52	6.77 ± 4.89	0.484	0.391	0.948
10 November 2015	10.50 ± 6.70	5.54 ± 3.17	0.449	0.822	0.950
11 November 2015	7.94 ± 6.61	4.03 ± 3.19	0.463	0.359	0.958
12 November 2015	7.77 ± 6.71	3.66 ± 3.04	0.432	0.305	0.954
13 November 2015	8.69 ± 6.85	3.99 ± 3.17	0.436	0.200	0.944
14 November 2015	8.21 ± 6.43	4.16 ± 3.17	0.458	0.409	0.928
15 November 2015	13.59 ± 8.86	6.44 ± 4.36	0.472	0.020	0.960
16 November 2015	11.54 ± 8.28	5.26 ± 3.63	0.406	0.573	0.926
17 November 2015	15.76 ± 9.13	8.50 ± 4.77	0.493	0.725	0.944
18 November 2015	12.73 ± 7.02	6.56 ± 3.62	0.499	0.210	0.965
19 November 2015	7.20 ± 5.83	3.52 ± 2.68	0.430	0.426	0.936
20 November 2015	17.47 ± 11.01	7.50 ± 4.93	0.425	0.077	0.949
21 November 2015	10.16 ± 7.20	4.54 ± 2.99	0.393	0.541	0.948
22 November 2015	10.21 ± 8.06	4.63 ± 3.68	0.430	0.239	0.942
Period average	11.04 ± 6.66	5.33 ± 2.81	0.409	0.815	0.970

The total number of grid cells in the finest modeling domain of land area is 6234



1.3 km. We presented detailed analyses on surface and aloft NO₂ concentrations targeting the period of the Chiba-Campaign 2015 conducted from 9 to 23 November 2015. The evaluations showed that the present air quality modeling system can generally capture the observed behavior of both surface and aloft NO₂ pollution. Some

difficulties in the current modeling system were also found. One issue is model underestimation when capturing roadside NO₂ pollution with a horizontal grid resolution of 1.3 km, and the other is model overestimation in the south direction from Chiba University. For the latter, this is thought to be related to the emission estimation over the Tokyo Bay area, which has intense sources from industrial and power plants. From the comparison on diurnal variation patterns, a slight difference in the morning peak of NO₂ concentration was also found, and further investigation of daytime chemical mechanisms related to NO_x may help refine our understanding of this. Based on the comparison between modeled daily-averaged surface and aloft NO₂ concentrations, it was found that aloft NO₂ concentration linearly scaled to 0.4–0.5 times the surface NO₂ concentration during both clean and polluted days, with higher value around noon due to the well-mixed air mass within the PBL. This study gave important suggestions for fostering our understanding of the vertical profiles of NO₂ over the greater Tokyo area.

Horizontal coverage based on ground-based measurements is sparse in terms of spatial coverage. Space-based satellite measurement can provide a broad spatial distribution of air pollutants. Although previous sensors have been limited in terms of spatial resolution for the purpose of detecting urban-scale air pollution (Itahashi et al. 2014; Irie et al. 2016), the state-of-the-science sensor of TROPOspheric Monitoring Instrument (TROPOMI) onboard the Sentinel-5 precursor satellite launched on 13 October 2017 can provide finer spatial resolution below 10 km (Griffin et al. 2019). Although the use of these recent satellite observations may be helpful for understanding the spatial distribution patterns, the values measured by the satellite are vertically integrated concentrations. Comprehensive analysis of surface and aloft observation as conducted in this study combined with this kind of satellite observation with finer resolution is required for future studies. From this perspective, because cloudy and lower PBL height conditions, which lead to increased NO₂ pollution, as found in this study, are unfavorable for satellite measurements, accurate modeling is required to bridge the missing information among surface, aloft, and satellite measurements. In addition, a clear linear relationship between surface and aloft NO₂ concentrations was limited to the analysis of Chiba-Campaign 2015 conducted in autumn, and analysis of other seasons is needed for further understanding of NO₂ behavior at the surface and aloft.

Abbreviations

4AZ-MAX-DOAS: Four different azimuth-viewing MAX-DOAS; AEROS: Atmospheric Environmental Regional Observation System; AMeDAS: Automated Meteorological Data Acquisition System; APMs: Ambient air pollution

monitoring stations; AQS: Air Quality Standard; CINDI: Cabauw Intercomparison Campaign of Nitrogen Dioxide Measuring Instruments; CMAQ: Community Multiscale Air Quality; DOAS: Differential optical absorption spectroscopy; GFED: Global Fire Emissions Database; HTAP: Hemispheric Transport of Air Pollution; JM2: Japanese MAX-DOAS profile retrieval algorithm, version 2; JMA: Japan Meteorological Agency; LT: Local time; M: Models; MANAL: Meso-scale Analysis; MAX-DOAS: Multi-axis differential optical absorption spectroscopy; MEGAN: Model of Emissions of Gases and Aerosols from Nature; MOZART: Model for Ozone and Related Chemical Tracers; NCAR: National Center for Atmospheric Research; NCEP: National Centers for Environmental Prediction; NMB: Normalized mean bias; NME: Normalized mean error; NO: Nitric oxide; NO₂: Nitrogen dioxide; NO_x: Nitrogen oxides; O: Observations; O_x: Oxidants; PBL: Planetary boundary layer; PM_{2.5}: Particulate matter with aerodynamic diameter less than to 2.5 µm; R: Correlation coefficient; RAPMS: Roadside air pollution monitoring stations; RHw: Relative humidity over water; RRTMG: Rapid radiative transfer model for global climate models; TROPOMI: TROPOspheric Monitoring Instrument; UTC: Universal Time Coordinate; UV: Ultraviolet; WRF: Weather Research and Forecasting.

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Authors' contributions

SI carried out the numerical modeling simulations, conducted the evaluation with the observation dataset, designed the analyses, and wrote the paper. HI performed the MAX-DOAS observation and its analyses during Chiba-Campaign 2015, proposed this topic, helped write the paper, and designed the study. All authors read and approved the final manuscript.

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Availability of data and materials

The datasets used and analyzed during the current study are available from the corresponding author on request.

Declarations

Competing interests

The authors declare that they have no competing interest.

Author details

¹Sustainable System Research Laboratory (SSRL), Central Research Institute of Electric Power Industry (CRIEPI), 1646 Abiko, Abiko, Chiba 270-1194, Japan. ²Center for Environmental Remote Sensing (CERES), Chiba University, 1-33 Yayoi-cho, Inage-ku, Chiba 263-8522, Japan.

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