

# Spatiotemporal Variations of NO<sub>2</sub> over Fukuoka Japan, Observed by Multiple MAX-DOAS and 3-D Coherent Doppler Lidar

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## Abstract

To clarify three-dimensional (3-D) spatiotemporal variations and horizontal–vertical transport processes in nitrogen dioxide (NO<sub>2</sub>) over urban areas, combined NO<sub>2</sub> profile observations by multiple Multi AXis Differential Optical Absorption Spectroscopy (MAX-DOAS) and direct wind observations by 3-D coherent Doppler lidar were made over an urban area in Japan. MAX-DOAS measurements were conducted at Yakuin and Fukuoka University in Fukuoka urban area with high temporal resolution of four minutes.

Enhanced NO<sub>2</sub> concentrations were often observed over the city center. We conducted a case study on 29 November 2018 under clear sky conditions and NO<sub>2</sub> profiles were well retrieved. Using MAX-DOAS at two locations, high NO<sub>2</sub> concentrations were observed near the surface of the city center in the morning. Higher NO<sub>2</sub> concentrations appearing gradually at higher altitudes over the city center and disappearing in the afternoon are explained using direct evidence of a 3-D wind field: an air mass with a high NO<sub>2</sub> concentration was transported upward over the city center. Then, the air mass was advected landward by a sea breeze. Multiple MAX-DOAS combined with 3-D Doppler lidar constitutes a powerful tool for elucidating horizontal–vertical transport processes. It can contribute to the improvement of data retrieval by satellites for urban areas.

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## 1. Introduction

Nitrogen oxides (NO<sub>x</sub> = NO + NO<sub>2</sub>) strongly affect air quality. They are expected to play a key role in climate change in the troposphere (e.g., Solomon et al. 1999). Elucidating tropospheric NO<sub>2</sub> variation in urban areas to enhance our knowledge of atmospheric chemistry is important because spatial variations in NO<sub>2</sub> concentrations are large there as a result of the spatial complexity of emission sources and short photochemical lifetimes of hours to days (e.g., Lamsal et al. 2010; Miyazaki et al. 2012; Takashima et al. 2015; Yamaguchi et al. 2016).

Tropospheric NO<sub>2</sub> measurements have long been conducted by satellite (e.g., Boersma et al. 2004; Richter et al. 2005; Boersma et al. 2011; Hilboll et al. 2013). In recent years, satellite observations have been developed to have finer spatial resolution, e.g., 5.5 × 3.5 km<sup>2</sup>, for the Sentinel-5 Precursor/TROPospheric Ozone Monitoring Instrument (TROPOMI) satellite (Veefkind et al. 2012); it can be a powerful tool for elucidating the atmospheric environment in urban area. To validate the satellite tropospheric NO<sub>2</sub>, ground-based Multi AXis Differential Optical Absorption Spectroscopy (MAX-DOAS) has been used (e.g., Brinksma et al. 2008; Irie et al. 2008b; Valks et al. 2011; Ma et al. 2013; Kanaya et al. 2014; Pinardi et al. 2020). This passive remote sensing

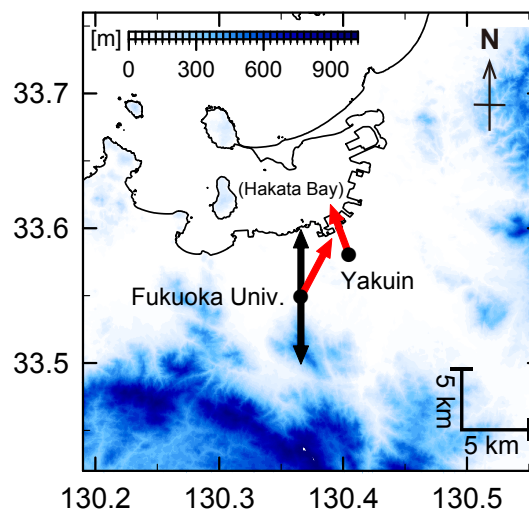


Fig. 1. Locations of observatories (longitude–latitude section). Color shows altitude; the solid line represents the coastline. Black arrows indicate the azimuth directions for RHI scanning of 3-D CDL. Red arrows represent lines of sight of MAX-DOAS.

technique is designed for atmospheric aerosol and gas profile measurements using scattered visible and ultraviolet solar radiation at several elevation angles (e.g., Höenninger et al. 2004; Wagner et al. 2004; Wittrock et al. 2004; Sinreich et al. 2005; Frieß et al. 2006; Irie et al. 2011). Because MAX-DOAS has high sensitivity to NO<sub>2</sub> near the ground, it can measure the NO<sub>2</sub> profile continuously during the daytime.

To clarify three-dimensional (3-D) spatiotemporal variations and horizontal–vertical transport processes affecting NO<sub>2</sub> over urban areas, MAX-DOAS measurements were taken from October 2018 with high temporal resolution of four minutes at two observatories in Fukuoka city, an urban area in Japan: Yakuin (33.58°N, 130.40°E) and Fukuoka University (33.55°N, 130.36°E). The distance between Yakuin and Fukuoka University is approximately 4.5 km, which is roughly equivalent to the spatial resolution of TROPOMI (Fig. 1). Although many past studies have been conducted with particular emphasis on transport processes of trace gases and pollutants, most studies have been based on surface observations. Observation methods for vertical profiles of trace gas, such as MAX-DOAS, have been developed in recent decades.

Fukuoka city, located on the Fukuoka plain south of Hakata Bay, has a population of approximately 1.6 million people concentrated in the city center. In contrast to other large cities in Japan, no large city is near Fukuoka city, making it an ideal location for elucidating the spatiotemporal variation of NO<sub>2</sub>, particularly for the transport and dilution processes of pollutants emitted from the city itself.

In earlier studies, a two-axis type MAX-DOAS instrument and car-mounted type MAX-DOAS instrument were used in Fukuoka city to clarify the inhomogeneity of NO<sub>2</sub> (Takashima et al. 2015; Yamaguchi et al. 2016). Results of those studies show

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that spatiotemporal variations of NO<sub>2</sub> are consistent with a sea-breeze circulation system by surface wind observations. Nevertheless, they merely deduced transport processes through surface wind observations, without direct observational evidence. For the present study, we conducted NO<sub>2</sub> profile observations at two locations together with direct 3-D wind observations, providing direct observational evidence for horizontal and vertical transport processes of an air mass with high NO<sub>2</sub> concentration emitted from Fukuoka city.

We have been conducting observations using a 3-D coherent Doppler lidar (3-D CDL) installed at Fukuoka University (33.55°N, 130.37°E; 55 m above sea level) since 30 November 2016 (Takashima et al. 2019). From those observations, we have analyzed data for continuous 3-D wind and signal-to-noise ratio (SNR) (approximately proportional to aerosol concentration). Data from 3-D wind observations can clarify horizontal–vertical transport processes of air masses enriched with NO<sub>2</sub> from near surface emission sources on small spatial scales.

The NO<sub>2</sub> profiles retrieved from MAX-DOAS observations often show enhanced NO<sub>2</sub> concentrations above the city center throughout the year. The simultaneous observations made using MAX-DOAS at the two locations together with 3-D CDL are limited. In addition, limiting the coincident periods to those with clear sky conditions and availability of lidar measurements with at least a certain SNR leaves very few days. Consequently, for this investigation, to clarify spatiotemporal variations in NO<sub>2</sub>, we conducted a case study of 29 November 2018 as the best example, when the NO<sub>2</sub> profiles at two locations were well retrieved at different times throughout the day. We also compared the tropospheric NO<sub>2</sub> vertical column densities (NO<sub>2</sub>VCD) observed using TROPOMI with retrieved results from measurements taken with multiple MAX-DOAS instruments.

## 2. Observations

### 2.1 NO<sub>2</sub> profile observations made using multiple MAX-DOAS instruments

For this study, NO<sub>2</sub> profile observations were made with MAX-DOAS instruments at Yakuin (33.58°N, 130.40°E) and at Fukuoka University (33.55°N, 130.36°E), located respectively about 1 km southwest of the city center (Tenjin) and about 5 km southwest of the city center (Fig. 1). Yakuin is located inside the urban area. The line of sight of the telescope is seaward (Hakata Bay). Fukuoka University is located far from the city center, but the telescope line of sight is toward the city center. It is noteworthy that the MAX-DOAS instrument at Fukuoka University obtains NO<sub>2</sub> profiles over the city center.

The MAX-DOAS instrument consists of two main parts: an outdoor telescope unit (Fig. S1) and an indoor spectrometer unit. The instrument is designed to be compact and to be operated with low power (up to 60 W). The telescope rotates to observe scattered sunlight at elevation angles of 2°, 3°, 4°, 5°, 10°, 20°, 30°, and 90° (elevation angles are changed every 30 s). The NO<sub>2</sub> profile is obtained with temporal resolution of 4 min. The telescope field of view is < 0.6°. A seven-core (100 μm radii) fiber optic bundle cable connects the outdoor telescope unit to the indoor spectrometer unit. The indoor spectrometer unit consists of the spectrometer (USB 4000; Ocean Optics Inc.) and a computer for spectral data recording and telescope elevation angle control. The spectrometer is housed in a heat-insulating plastic box to keep the spectrometer temperature at 288 ± 0.1 K using a temperature controller (E5CC; Omron Corp.) with a Peltier cooler. The spectral resolution of full-width at half-maximum (FWHM) is approximately 0.6 nm.

The recorded spectra for 460–490 nm are analyzed using the DOAS technique (Platt 1994) to retrieve the differential slant column densities (DSCD) of the oxygen collision complex (O<sub>2</sub>–O<sub>2</sub> or O<sub>4</sub>) and NO<sub>2</sub>, as reported by Irie et al. (2011). Here, DSCD is defined as the difference between the column concentration integrated along the sunlight path measured at low elevation angles and at a midday reference elevation angle (90°). Absorption by NO<sub>2</sub>, O<sub>4</sub>, O<sub>3</sub>, H<sub>2</sub>O, and the Ring effect were considered. We

used the NO<sub>2</sub> absorption cross-section data at 294 K reported by Vandaele et al. (1996), O<sub>4</sub> at 296 K by Thalman et al. (2013), O<sub>3</sub> at 243 K by Bogumil et al. (2003), and H<sub>2</sub>O at 296 K by Rothman et al. (2010).

The NO<sub>2</sub>DSCD, O<sub>4</sub>DSCD, and their respective errors are converted to the NO<sub>2</sub> profile and column using the retrieval algorithm, Mexican MAX-DOAS Fit (MMF; Friedrich et al. 2019). As reported by Friedrich et al. (2019), the aerosol extinction coefficient profile is retrieved using the known O<sub>4</sub> profile. Then, the NO<sub>2</sub> profile is retrieved. A priori information for aerosol and NO<sub>2</sub> profiles was set to decrease exponentially from the surface. A priori aerosol data for total optical depth, single scattering albedo, asymmetry parameter, and angstrom exponent were fixed respectively as 0.18, 0.95, 0.65, and 1.00. Wavelengths of 474 nm and 477 nm were used, respectively, for NO<sub>2</sub> and aerosol retrieval. Vertical resolution near the surface is more of the order of a couple of hundred meters. It reaches roughly a kilometer or even coarser around 1.5–2.5 km (Tirpitz et al. 2021). The total error of the NO<sub>2</sub>VCD retrieved by MMF was estimated as approximately 20% (Friedrich et al. 2019). The effective distance of MAX-DOAS for 29 November 2018 was approx. 12–20 km.

### 2.2 Direct 3-D wind observation by 3-D CDL

Direct 3-D wind and aerosol (SNR) observations were conducted using a 3-D CDL (Mitsubishi Electric Corp.) installed at Fukuoka University (33.55°N, 130.37°E; 55 m above sea level) (Fig. 1). The 3-D CDL performs plan position indicator (PPI) scans continuously at 0°, 2°, and 10° and range height indicator (RHI) scans at 0° (northward) and 112° (clockwise from north). Then PPI scans with elevation angles of 0°, 2°, and 10° (five scans: 0°, 0°, 2°, 10°, 0°), and RHI scans with azimuth angles of 0° (north) and 112° (clockwise from north) (eight scans: 0°, 0°, 0°, 0°, 112°, 112°, 112°, 112°) were conducted in sequence. The scanning speed was 2° s<sup>-1</sup>; a PPI (RHI) scan takes about 3 (1.5) min. For this study, we used PPI scans of the horizontal section (elevation angle = 0°) and RHI scans of the vertical north–south section (azimuth angle = 0°). The 3-D CDL has 200 range bins. The range resolution was set to 60 m. Thereby, the measurable distance was up to 12 km. Because of the shadows of buildings and mountains, we are unable to receive meaningful signals to the northwest/west/southwest/south in the PPI scans and RHI scans (Fig. 3). Because SNR decreases sharply along with increased distance from the lidar, we used radial wind velocity data with a threshold of SNR ≥ 8 dB to analyze the radial velocity variations in the boundary layer within an approx. 5 km horizontal area covering the Fukuoka urban area. Details of the setup and technical performance of 3-D CDL are described by Takashima et al. (2019).

## 3. Results and discussion

Figure 2 shows the NO<sub>2</sub> volume mixing ratio observed using MAX-DOAS at (a) Yakuin and (b) Fukuoka University on 29 November 2018 as a function of time (x-axis) and height above the ground (y-axis). At Yakuin, maximum NO<sub>2</sub> was observed near the surface at around 7:00–10:00. A minimum of NO<sub>2</sub> concentration was observed above the surface during 13:30–15:00. At Fukuoka University, a maximum was observed near the surface at 7:00–12:30. Higher contents of NO<sub>2</sub> appeared gradually at higher altitudes during 10:30–12:00. A minimum was observed above the surface at 14:00–16:30 (about 30 min later than at Yakuin). From these NO<sub>2</sub> variations, the 3-D wind field can be described by 3-D CDL in three phases: (i) 7:00–10:30, (ii) 10:30–13:30, and (iii) 13:30–17:00.

Figure 3 presents horizontal sections of radial wind velocity (m s<sup>-1</sup>) with elevation angle of 0° and north–south height sections of radial wind velocity (m s<sup>-1</sup>) with the azimuth direction of 0°, with representative snapshots corresponding to phases (i), (ii), and (iii). Near the surface, PPI scans revealed the following: (i) southeasterly winds of about 1–2 m s<sup>-1</sup> (Fig. 3a), (ii) weak winds with a fishnet-like structure (Fujiwara et al. 2010) (Fig. 3b), and (iii) northerly winds of about 3–5 m s<sup>-1</sup> (Fig. 3c). In the north–south

section, RHI scans revealed the following: (i) southerly winds in the thin layer up to about 0.2 km (Fig. 3d); (ii) disappearance of southerly winds in the thin layer and weak winds, at least until about 1.0 km (Fig. 3e); and (iii) northerly winds of about 3–5 m s<sup>-1</sup>

up to about 0.6 km (Fig. 3f).

The spatiotemporal variation of NO<sub>2</sub> is explainable by the results of 3-D wind fields: in phase (i), the air mass with high NO<sub>2</sub> concentrations emitted in the morning (around 7:00–10:30) was trapped in the thin layer near the surface. It was unable to mix vertically with the overlying clean air mass. In addition, high NO<sub>2</sub> concentrations in the air mass observed under southerly wind conditions are expected to originate mainly from Fukuoka city itself. In phase (ii), the thin layer near the surface has disappeared. The air mass with high NO<sub>2</sub> concentrations at the city center was transported upward from near the surface under weak horizontal wind conditions. It is noteworthy that the vertical transport/dilution speed estimated from Fig. 2b was approx. 0.5 m s<sup>-1</sup> (10:30–11:40), which is comparable to the spatial average of the vertical velocity observed using 3-D CDL in Fig. 3e (< 1 m s<sup>-1</sup>). In phase (iii), an air mass with high NO<sub>2</sub> concentration over the city center and an air mass with low NO<sub>2</sub> concentration over Hakata Bay advected southward (landward) by sea breezes in the afternoon. Yakuin is located seaside: a minimum NO<sub>2</sub> was observed about 30 min earlier than at Fukuoka University. The time difference between the two observatories is consistent with the northerly wind of about 3–5 m s<sup>-1</sup> during phase (iii). The case of 29 November 2018 suggests that a combination of multiple MAX-DOAS and 3-D CDL is a powerful tool for understanding horizontal–vertical transport processes of NO<sub>2</sub> in urban areas.

Radiosonde observations are conducted routinely by the Fukuoka District Meteorological Observatory at Roppon-matsu (33.58°N, 130.38°E). At about 8:30 am, strong temperature inversion was observed below about 0.2 km (Fig. S2). The results are consistent with the high NO<sub>2</sub> concentrations near the surface during phase (i) because of the lack of mixing with air masses located at higher altitudes.

Tropospheric NO<sub>2</sub>VCD was observed by TROPOMI (KNMI, OFFLINE Level-2, Van Geffen et al. 2019) over Fukuoka city at 13:44 (local time) 29 November 2018 (Fig. S3). Comparisons

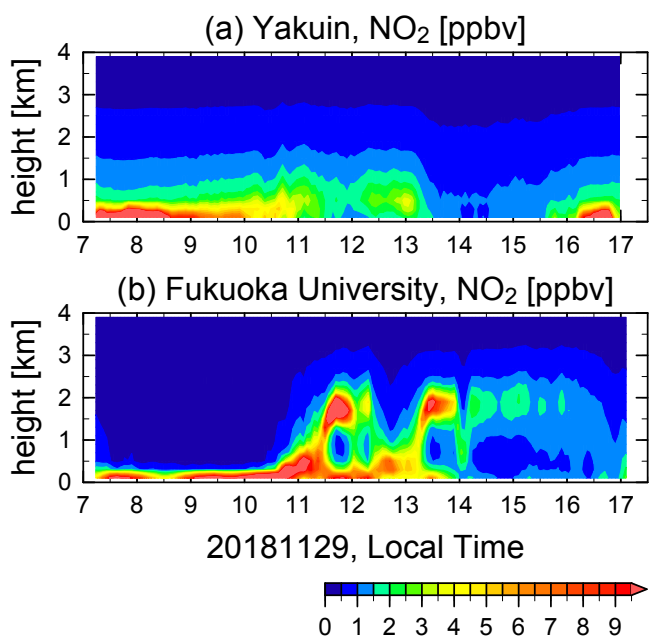


Fig. 2. Time–height section of NO<sub>2</sub> volume mixing ratio (ppbv) observed using MAX-DOAS at (a) Yakuin and (b) Fukuoka University on 29 November 2018.

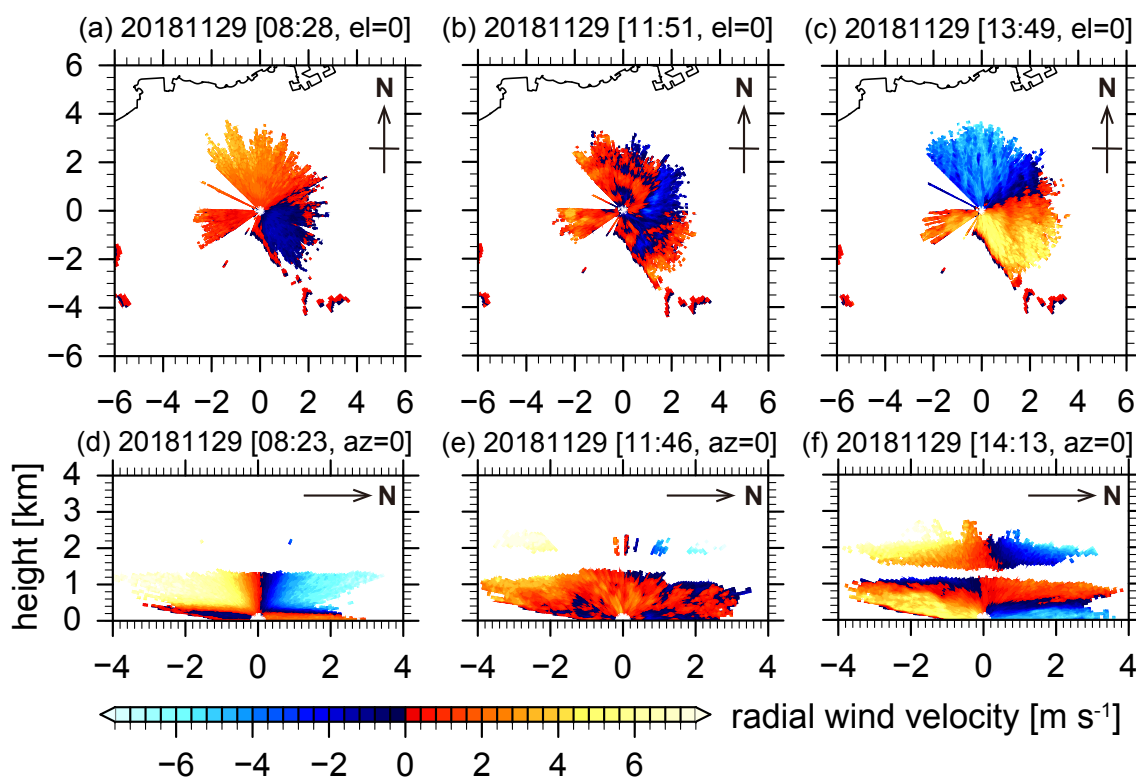


Fig. 3. Horizontal sections of radial wind velocity (m s<sup>-1</sup>) for PPI scan with elevation angle of 0° at 8:28, 11:51, and 13:49 (top panels) and north–south height sections of radial wind velocity (m s<sup>-1</sup>) for RHI with azimuth direction of 0° at 8:23, 11:46, and 14:13 (bottom panels) observed on 29 November 2018 by 3-D CDL in Fukuoka. For the top panels, axes show horizontal distance (km) from 3-D CDL. For bottom panels, the horizontal axis is distance (km) from 3-D CDL. The vertical axis shows height (km). Warm colors represent flow away from 3-D CDL. Cold colors represent flow toward it.



of tropospheric NO<sub>2</sub>VCD by TROPOMI with both MAX-DOAS showed that the horizontal variations of NO<sub>2</sub>VCD by TROPOMI were comparable with the horizontal variations of NO<sub>2</sub>VCD by multiple MAX-DOAS. However, TROPOMI underestimates the tropospheric NO<sub>2</sub>VCD as much as the Ozone Monitoring Instrument (OMI) described in Kanaya et al. (2014).

The NO<sub>2</sub> profile retrieved from MAX-DOAS measurements includes large retrieval error caused by smoothing and measurement noise. It also depends on the a priori profile and the assumed covariance matrix. As demonstrated by results of this study, large differences in NO<sub>2</sub> profiles (e.g., between the phases (i) (ii), and (iii)) were consistent with the vertical wind structure observed by 3-D CDL. These results and continuous MAX-DOAS measurements at multiple locations together with 3-D CDL are expected to contribute to improvement of NO<sub>2</sub> retrieval algorithms over urban areas for satellite observations.

#### 4. Conclusions

To clarify three-dimensional spatiotemporal variations and horizontal–vertical transport processes of NO<sub>2</sub> over the urban area, the combination of NO<sub>2</sub> profile observations by multiple MAX-DOAS and direct wind observations by 3-D CDL was conducted over metropolitan Fukuoka in Japan.

Enhanced NO<sub>2</sub> contents were often observed over the city center. We conducted a case study on 29 November 2018 under clear sky conditions, when the NO<sub>2</sub> profiles were well retrieved. By providing direct observational evidence of a 3-D wind field, we clarified that, at the beginning, the air mass with high NO<sub>2</sub> concentrations was trapped within the thin layer near the surface. Then the air mass with high NO<sub>2</sub> concentrations was transported upward/diluted in the boundary layer by Bénard-type convection (fish net structure). Finally, the air mass with high NO<sub>2</sub> concentration over the city center and the air mass with low NO<sub>2</sub> concentration over the sea were advected by sea breezes in the boundary layer. The combination of multiple MAX-DOAS and 3-D CDL is a powerful tool for elucidating horizontal–vertical transport processes in urban areas.

We also compared the tropospheric NO<sub>2</sub>VCD observed by TROPOMI to results of measurements obtained using the two MAX-DOAS instruments. Horizontal variations of NO<sub>2</sub>VCD by TROPOMI were comparable with the horizontal variations of NO<sub>2</sub>VCD by multiple MAX-DOAS. However, TROPOMI underestimates the tropospheric NO<sub>2</sub>VCD.

This study validated the spatiotemporal variation of NO<sub>2</sub> for one day only, but long-term validation using multiple MAX-DOAS and 3-D CDL is expected to contribute to improving satellite retrieval in urban areas.

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#### Supplements

Supplement 1: Photographs of MAX DOAS instrument used for this study. The telescope can be rotated to point at different

elevation angles (2°, 3°, 4°, 5°, 10°, 20°, 30°, and 90°) to measure scattered sunlight.

Supplement 2: Vertical profiles of temperature (K), potential temperature (Po. Temp) (K) and relative humidity (%) observed by radiosonde in Roppon-matsu (33.58°N, 130.38°E) around 8:30 am, 29 November 2018.

Supplement 3: Longitude–latitude section of tropospheric NO<sub>2</sub>VCD (10<sup>15</sup> molecule cm<sup>-2</sup>) observed by Sentinel-5 Precursor/TROPOMI on 29 November 2018. Colored square symbols represent tropospheric NO<sub>2</sub>VCD.

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