First Concurrent Observations of NO₂ and CO₂ From Power Plant Plumes by Airborne Remote Sensing

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Abstract Combined NO₂ and CO₂ observations have the potential to constrain the identification of the locations and strength of urban CO₂ emissions, in particular, point sources such as power plants. We report the first results of airborne spectroscopic NO₂ and CO₂ observations over an urban area in Japan in February 2018. Inversed emission rates of two stacks of the coal-fired power plant for CO₂ showed relatively good agreement with those estimated by a bottom-up inventory—the Regional Emission inventory in ASia (REAS) v3.1—within −7% to 40% because the plume shapes were well identified due to constraint by NO₂ measurements. The estimated NO₂ emission rates showed discrepancies more than 80% with those estimated by the REAS v3.1, mainly due to the uncertainties in activity data and emission factors, or in the greatly varying NO/NOₓ ratios in fresh plumes, which warrant further investigations when estimating NO₂ emissions from satellite NO₂ observations on km-scales.

Plain Language Summary Burning of fossil fuels at high temperatures constitutes a major anthropogenic source of nitrogen oxides (NOₓ) and carbon dioxide (CO₂). While CO₂ stays in the atmosphere for hundreds of years, thereby being a well-mixed gas, NOₓ has a much shorter lifetime of only a few hours. This substantial difference in lifetime between NO₂ and CO₂ means that concurrent NO₂ and CO₂ observations obtained by the same platform can be used to identify the locations and strength of CO₂ emissions from point sources such as power plants. In February 2018, for the first time, we obtained concurrent airborne spectroscopic NO₂ and CO₂ observations over an urban area, to demonstrate the traceability of NO₂ to CO₂. The plumes of co-emitted NO₂ and CO₂ were derived from measured spectra. The plumes of NO₂ and CO₂ co-emitted from the stacks of power plants were well identified owing to constraint by NO₂. Uncertainties of inversed emission rates were statistically derived. For CO₂, the results were within 40% in agreement with a bottom-up emission inventory known as REAS v3.1. For NO₂, however, a disagreement of 80% was identified, likely due to the uncertainties of the inventory data or in the NO₂ partitioning in fresh plumes.

1. Introduction Carbon dioxide (CO₂) is a major greenhouse gas (GHG) in the atmosphere and an important driver of climate change. According to the World Meteorological Organization (WMO) Greenhouse Gas Bulletin, the globally averaged mole fraction of CO₂ reached 407.8 ± 0.1 ppm in 2018, representing ∼50% increase compared to the pre-industrial level (i.e., 278 ppm) and accounting for 2.05 W m⁻² of the atmospheric radiative forcing. While in-situ measurement networks as well as ground-based and airborne remote sensing of CO₂, such as the WMO’s Global Atmosphere Watch or the Total Carbon Column Observing Network (TCCON; Wunch et al., 2011), support accurate estimation of the globally averaged CO₂ concentration and its long-term temporal variation (Artuso et al., 2009; Fang et al., 2014), satellite observations provide denser spatial information on the CO₂ columns over land and ocean on a global scale. Satellite observations of column-averaged dry-air mole fractions of CO₂ (XCO₂) have been successfully obtained by the spectrometer SCanning Imaging Absorption SpectroMeter for Atmospheric CHartographY (SCIAMACHY; Bovensmann et al., 1999) onboard the ENVISAT satellite, and the Greenhouse Gases Observing Satellite (GOSAT; Kuze et al., 2009), GOSAT-2 (Nakajima et al., 2013). Orbiting Carbon Observatory-2 (OCO-2; Crisp et al., 2004)...
and OCO-3 (Eldering et al., 2019) satellite missions. These observations demonstrate that space-based remote sensing of greenhouse gases can be used to detect CO₂ emissions from anthropogenic sources at city scales (Cambaliza et al., 2015; Nassar et al., 2017). Due to the long lifetime and existence of various sources and sinks, however, local CO₂ enhancements from individual anthropogenic point sources are relatively small compared to the background level and its natural variability, rendering satellite-based quantification of the CO₂ emission flux challenging.

Nitrogen oxides (NOₓ), known as short-lived air pollutants, are emitted during burning of fossil fuels at high temperatures, and thus have common origin with anthropogenic CO₂. Since 1995, satellite observations of the tropospheric nitrogen dioxide (NO₂) have been successfully obtained by the Global Ozone Monitoring Experiment (GOME; Burrows et al., 1999), SCIAMACHY, Ozone Monitoring Instrument (OMI; Levelt et al., 2006), GOME-2 (Callies et al., 2000), and TROPOspheric Monitoring Instrument (TROPOMI; Vreeken et al., 2012). These satellite observations provided a better understanding of spatiotemporal variations and long-term NO₂ trends (e.g., Hilboll et al., 2013; Richter et al., 2005; van der A et al., 2006, 2008). Furthermore, recent satellite NO₂ observations with high spatial resolution of several kilometers are useful for estimating the CO₂ emissions and identifying their sources (e.g., Goldberg et al., 2019; Reuter et al., 2019). Toward the Global Stocktake of the Paris Agreement—a 5-yearly review of the signatory countries’ actions for climate change—it is important not only to observe CO₂ with a high spatiotemporal resolution but also to identify unknown CO₂ point sources by utilizing the NO₂ traceability. In this context, concurrent NO₂ and CO₂ observations from an identical platform have great potential, and relevant satellite missions are currently planned, including GOSAT-GW by Japan and CO2M by Europe (https://www.esa.int/Applications/Observing_the_Earth/Copernicus/Copernicus_High_Priority_Candidates). The launch of the GOSAT-GW satellite is expected in 2024: A specific task of this satellite will be to concurrently monitor CO₂ and NO₂ using a suite of grating spectrometers covering short wave infrared (SWIR) and visible bands. The European CO2M—Copernicus Anthropogenic CO₂ Monitoring—mission also focuses on concurrent anthropogenic CO₂ and NO₂ observations. Both missions provide higher spatial resolution with approximate nominal value of 2 × 2 km². Although aircraft observations are useful for demonstrating the imaging capability of such satellite missions, previous studies focused only on either NO₂ or CO₂, for example, MAMAP (Krings et al., 2011), Geo-TASO (Leitch et al., 2014), AirMAP (Schönhardt et al., 2015), SWING (Merlaud et al., 2018), and APEX (Popp et al., 2012). Therefore, for the first time in this study, we evaluate the efficiency of concurrent NO₂ and CO₂ observations from the same platform using an airplane.

2. Airborne Measurements

In February 2018, we obtained airborne NO₂ and CO₂ observations over the city of Nagoya, one of the most populated urban areas in Japan, where large quantities of anthropogenic CO₂ and NO₂ are emitted by the power plant, industry, and transportation sectors. Specifically, a coal-fired power plant (PP) located near Nagoya is reported to have the highest CO₂ emissions in Japan as a single source. The observations were obtained mostly under clear-sky conditions; in a few rare instances, thin cloud shadows were present. The flight geometry and track on February 16, 2018 can be found in (Figure S1).

The SWIR spectrometer was custom made for dedicated airborne experiments; the UV-VIS spectrometer was conducted with a commercial spectrometer. All spectrometers were manufactured by Headwall photonics Inc. In addition, a video camera was installed to record atmospheric features such as clouds in the nadir direction of the spectrometer. For the UV-VIS spectrometer, a bias correction along the across-track direction was conducted (Text S1; Figure S2). To obtain high signal-to-noise ratio, 16 pixels were binned for the CO₂ spectrometers. For the detailed spectrometers’ specifications onboard the airplane, see Table S1.

The targeted PP emitted approximately 24 MT yr⁻¹ CO₂ in 2012, which were the highest emission rates among coal-fired PPs in Japan. The targeted PP’s total rated capacity is 4100 MW: three plants (plant group A) with 700 MW and two plants (plant group B) with 1000 MW rated capacity. There are two stacks located at 34.8330°N, 136.9596°E (Southern stack) and 34.8363°N, 136.9593°E (Northern stack), which hereafter are referred to as A and B, respectively. Flue gas emitted from stack A is attributed to plant group A and that emitted from stack B is attributed to plant group B.
In the calibration and pre-processing modules, pixel binning is first performed for raw observed data after excluding abnormal output pixels. The abnormal output pixels are determined based on radiometric calibration data by using a blackbody and offset data obtained under dark environment. Wavelength and radiometric calibration were performed in the laboratory before the aircraft campaign. The solar zenith angle (SZA) was geometrically calculated for each spectrum assuming the spherical surface, and the viewing zenith angle (VZA) was inferred from the aircraft position and attitude during the survey, which were obtained by global navigation satellite system and inertial measurement unit (GNSS/IMU) mounted on the optical plate. Observed wind speed and direction on the surface at the time of the survey—about 2 km away from the PP, on the opposite side across a river—were 2–3 m s\(^{-1}\) and west-northwest, respectively.

3. Retrieval of VCD\(_{\text{NO}_2}\) and XCO\(_2\)

3.1. NO\(_2\)

In order to obtain the tropospheric vertical column density of NO\(_2\) (VCD\(_{\text{NO}_2}\)), we employed the differential optical absorption spectroscopy (DOAS; Platt, 1994), which derives a slant column density (SCD) of NO\(_2\) existing on the light path of interest with respect to a reference path, using the absorption cross-sections of trace gases within the UV-VIS region. A spatiotemporally averaged spectrum taken over an area with clean atmosphere is used as a reference in airborne DOAS retrievals (e.g., Meier et al., 2017). Thus, the retrieved SCD is the differential slant column density (dSCD), that is, the difference of path-integrated number densities between the target area and background; therefore, we expect cancellation of signals from stratospheric NO\(_2\) if the variation of stratospheric NO\(_2\) between the target area and background is negligible. Furthermore, the spectral resolutions vary depending on the across-track direction due to aberration of the optical system. In this study, therefore, the reference spectra for each viewing direction were generated as averages over 24 spectra observed during 11:01:30–11:01:42 (JST) over the ocean.

The SCD in the troposphere (SCD\(_{\text{NO}_2}\)) in the airborne DOAS is derived from the following equation:

\[
\text{SCD}_{\text{NO}_2} = \text{dSCD} + \text{SCD}_0 + \text{SCD}_{\text{strat}},
\]

where SCD\(_0\) is the offset of the tropospheric SCD existing in the background area and SCD\(_{\text{strat}}\) is a variation of the stratospheric SCD from the background to target area. In our analysis, we assumed that SCD\(_{\text{strat}}\) is negligible because our target area was such that the stratospheric SCD and the similar SZA could be regarded as constants during the whole observation period.

We assumed the random error as the standard deviation of the retrieved NO\(_2\) dSCD observed in the time and region where the reference spectra were taken. The NO\(_2\) dSCD standard deviation was 0.835 × 10\(^{16}\) molec. cm\(^{-2}\), which was consistent with the root-mean-square error of the DOAS results over the background region (([0.535–1.01] × 10\(^{15}\) molec.cm\(^{-2}\)). The SCD\(_0\) was determined by the product of the tropospheric background VCD (i.e., VCD\(_b\)) and the tropospheric air mass factor (AMF\(_{\text{trop}}\)). In this study, we assumed VCD\(_b\) = 0.228 × 10\(^{16}\) molec.cm\(^{-2}\), with 100% uncertainty as a typical value of the background according to the monthly mean of tropospheric VCDs observed by TROPOMI in February 2019 over the ocean near Nagoya city. The bias of the airborne observations was evaluated by comparison with the TROPOMI observations (Text S3; Figure S5). From Equation 1 and our negligible SCD\(_{\text{strat}}\) assumption, the uncertainty of SCD\(_{\text{NO}_2}\) (\(\varepsilon_{\text{SCD}}\)) was regarded as the root-sum-square of the uncertainties of dSCD (\(\varepsilon_{\text{dSCD}}\)) and SCD\(_0\) (\(\varepsilon_{\text{SCD}_0}\)). Given \(\varepsilon_{\text{dSCD}} = 0.835 \times 10^{16}\) molec.cm\(^{-2}\) and \(\varepsilon_{\text{SCD}_0} = 0.228 \times 10^{16}\) molec.cm\(^{-2}\), \(\varepsilon_{\text{SCD}}\) was estimated to be 0.961 × 10\(^{16}\) molec.cm\(^{-2}\).

The SCD\(_{\text{NO}_2}\) was converted to VCD\(_{\text{NO}_2}\) by considering the AMF\(_{\text{trop}}\) inferred from the radiative transfer model. Practically, during the retrieval procedure, the vertically resolved AMFs were utilized and taken from the pre-calculated look-up table (LUT; Table S1). In this study, we employed the SCIATRAN model developed in the University of Bremen (Rozanov et al., 2005) to build the AMF LUT. The surface reflectance used in the AMF calculation was assumed to be 0.04 at 440 nm (Figure S3). Because there is no information with regard to aerosol properties around this area during the survey, we derived the aerosol extinction profile assuming the urban atmospheric scenario following Fujinawa et al. (2019).

Total VCD\(_{\text{NO}_2}\) error (\(\varepsilon_{\text{VCD}}\)) was statistically derived assuming all errors independent as follows:
\[ \varepsilon_{\text{VCD}} = \left( \frac{\varepsilon_{\text{SCD}}}{\text{AMF}_{\text{top}}} \right)^2 + \left( \frac{\varepsilon_{\text{SCDh}}}{\text{AMF}_{\text{top}}} \right)^2 + \left( \frac{\varepsilon_{\text{NO}_2}\text{AMF}_{\text{top}}}{\text{AMF}_{2\text{top}}^2} \right)^2 . \] 

To estimate a typical \( \varepsilon_{\text{VCD}} \) under polluted conditions, \( \varepsilon_{\text{NO}_2} \), \( \text{AMF}_{\text{top}} \), and an uncertainty of \( \text{AMF}_{2\text{top}} \) (\( \varepsilon_{\text{AMF}_{2\text{top}}} \)) were assumed to be \( 5.0 \times 10^{16} \) mole\( \cdot \)cm\(^{-2} \), 2, and 0.60, respectively. The relative error of \( \varepsilon_{\text{AMF}_{2\text{top}}} \) (i.e., 30%) was according to the “Heavily polluted” condition in Boersma et al. (2004). Consequently, \( \varepsilon_{\text{VCD}} \) was estimated to be \( 0.891 \times 10^{16} \) mole\( \cdot \)cm\(^{-2} \).

3.2. CO

The optimal estimation method (OEM; Rodgers, 2000) is generally used for CO\(_2\) retrieval from SWIR spectra. However, the OEM tends to be time-consuming due to online radiative transfer calculation. In addition, the method needs precise \( \text{a priori} \) data, which makes the accurate retrieval difficult in case of no \( \text{a priori} \) information such as in this study. Therefore, we employed a simple and fast CO\(_2\) retrieval procedure to obtain the XCO\(_2\) enhancement emitted by the PP, although the procedure is insufficient to accurately retrieve the absolute XCO\(_2\) compared to earlier studies (e.g., Boesch et al., 2011; Butt et al., 2009; O’Dell et al., 2012; Reuter et al., 2017; Schneising et al., 2008; Yoshida et al., 2013). In this study, we wish to obtain the plume enhancement of the XCO\(_2\) relative to the background, and for our purposes the simple method is sufficient. The algorithm is composed of two parts: one part is for calibrating the aircraft observation spectrum, and the other part is for generating the synthetic spectrum, which is obtained by inputting the solar irradiance spectrum into a forward model based on the Lambert-Beer law that does not consider scattering, as follows:

\[
I_{\text{syn}}(\nu) \equiv F_0(\nu) \frac{\rho}{\pi} \exp \left\{ -X\text{CO}_2 \left\{ \frac{1}{\cos \theta_s} \int_{z_0}^{z_{\text{max}}} \frac{1}{\cos \theta_t} \int_{z_{\text{top}}}^{z} n_{\text{dry}} \sigma(\nu) \, dz \right\} \right\} .
\]  

where \( \nu \) is wave number (cm\(^{-1} \)); \( I_{\text{syn}}(\nu) \) is the synthetic radian spectrum and \( F_0(\nu) \) is the solar irradiance spectrum; a continuum model of Thuillier et al. (2003) multiplied by transmittance of Toon (2015); \( \rho \) is surface albedo; \( n_{\text{dry}} \) is number density of the background atmosphere; \( \theta_s \) and \( \theta_t \) are SZA and VZA, respectively; \( z_0 \) is elevation of ground surface; \( z_{\text{max}} \) is altitude at the top of the atmosphere and \( z_{\text{top}} \) is observation altitude; \( \sigma(\nu) \) is CO\(_2\) absorption cross-section from the ABSCO v4.2 data base (Thompson et al., 2012). The background atmosphere in the forward model was divided into 120 layers. \( n_{\text{dry}} \) was calculated for each layer from the hydrostatic equilibrium. \( \sigma(\nu) \) was also selected for each layer from the LUT of ABSCO v4.2 according to the temperature and pressure of each layer. The vertical profiles of temperature (T) and pressure (P) of the background atmospheric model were based on grid-point values (GPV) calculated by the Global Spectral Model (GSM) of the Japan Meteorological Agency (JMA) on the day of the survey. However, since the GPV is incremented by 0.5-degree step both latitudinally and longitudinally, each physical quantity was interpolated along the geolocation of a representative point. To generate a raw synthetic spectrum, solar irradiance spectrum (i.e., \( F_0(\nu) \)) was resampled in the weak CO\(_2\) band (6,140–6,300 cm\(^{-1} \)), where ABSCO v4.2 covers with 0.01 cm\(^{-1} \) interval.

Subsequently, synthetic spectrum with inclusion of instrumental characteristics was considered. The synthetic spectrum was first obtained by conversion from wavenumber to wavelength domain by pixel sampling, along with wavelength axis determination by wavelength calibration. The spectrum was convolved with the instrument line shape (ILS), which was assumed to be a general normalized Gaussian function. In the nonlinear fitting process, in addition to XCO\(_2\), instrumental characteristics such as wavelength offset \( (\lambda_0) \), ILS width, spectral amplitude and its slope, and offset were considered as fitting parameters, because these parameters were not completely known in advance, owing to the deviation from an ideal alignment inside the spectrometer. This alignment deviation was due to temperature instability during flight, as well as radiance inhomogeneity inside the scene due to sharp change of ground surface reflectivity. The wavelength region was limited to 1,588.1–1,628.0 nm, which ABSCO v4.2 covers, and the sampling interval was
0.172 nm, resulting in 232 equations that were solved during the nonlinear least squares fitting to optimize six parameters.

4. Results and Discussions

We selected 11:55:00 to 12:14:00 (JST) as the time period of the flight over the PP, to compare the columnar amounts of NO₂ and CO₂ and their plume shapes emitted from two stacks of the PP. Figure 1 shows the mesh-plot of the VCD_{NO₂} (left panel) and XCO₂ (right panel) retrieved during the period, the locations of the two stacks of the PP (white diamonds), and wind directions (white and black arrows) derived from the numerical weather-prediction model (i.e., MesoScale Modeled [MSM] GPV) and manual adjustment based on the plume extent of the retrieved VCD_{NO₂} using similar approach to Reuter et al. (2019), Nassar et al. (2017), and Krings et al. (2011). Both retrieved columns were gridded with 0.001° × 0.001° mesh (corresponding to approximately 100 m). The white dashed rectangles in each panel represent cross-sectional areas (800 × 2,000 m) perpendicular to the adjusted wind direction, which were centered at a point 400 m away from the middle point between the two stacks. For concurrent plumes of NO₂ and CO₂, the similar plume shapes are evident in both panels, albeit with large spatial variability in the case of VCD_{NO₂}. A scatter plot of the observed VCD_{NO₂} and XCO₂ (Figure S8) between 34.825°–34.840° latitude and 136.955°–136.970° longitude, depicts good correlation (i.e., R = 0.67) between the distribution of VCD_{NO₂} and XCO₂ enhancements.

We approximated the emission rate of the PP using bottom-up inventory data, at point sources of PPs from the the Regional Emission inventory in ASia (REAS) v3.1. This database provides not only gridded distribution but also the individual emissions of CO₂ and NOₓ. Given that the emission inventory data of CO₂ and NOₓ are 23.0 MtCO₂ yr⁻¹ and 11.7 ktNOₓ yr⁻¹, with uncertainties of 13% and 32%, respectively (Kurokawa & Ohara, 2020), and that the emissions ratios from stack A and B are 0.51 and 0.49, respectively, the emission
rates of CO₂ and NO₃ from stack A and B were estimated to be 373 ± 48, 355 ± 46, 0.19 ± 0.06 × 10⁻³, and 0.18 ± 0.06 × 10⁻³ in kg s⁻¹, respectively.

To inverse emission rates of CO₂ and NO₃ emitted from the two stacks using observed columns, we modified the two inverse methods: the cross-sectional flux (CSF) and the integrated mass enhancement (IME) method. The emission rate Q are defined as follows:

\[
Q = \begin{cases} 
U_{\text{eff}} \int_{x=0}^{x=x} \Delta \Omega(x, y) dy & \text{for CSF} \\
(U_{\text{eff}}/l) \sum_{j=1}^{N} \Delta \Omega A_j & \text{for IME}
\end{cases}
\]

respectively, where \(U_{\text{eff}}\) the effective wind speed (m s⁻¹), \(l\) the plume length (m), \(\Delta \Omega(x, y)\) the column mass enhancement (kg m⁻²) with \(x, y\) coordinate (\(x\) axis parallel to the wind direction), \(j\) the pixel number, and \(A\) the pixel area (m²). These two methods have a simple physical basis for relating \(U_{\text{eff}}\) and emission rate, and \(U_{\text{eff}}\) can be empirically approximated using 10-m wind speed \((U_{10})\), that is, \(U_{\text{eff}} = 1.47 \times U_{10}\) for CSF and \(U_{\text{eff}} = 0.9 \times \log(U_{10}) + 0.6\) for IME, respectively. Detailed descriptions of these methods are available in Varon et al. (2018) and references therein, who applied these methods to methane because the standard methods by the Gaussian plume inversion or source pixel mass balance were inadequate, owing to the small scale of the instantaneous plume, wind variability, and turbulent diffusion. We also concluded that the standard methods were not applicable in our case because, despite the high spatial resolution, the narrow swatch in our study does not allow effective statistical averaging with a large scale and thus better Gaussian representation. Thus, the two methods were considered more appropriate for NO₂ and CO₂ in our case. The \(U_{10}\) value of 1.7 m s⁻¹, which is extracted from the MSM-GPV data, is hereafter used. Moreover, the IME method was simplified through the plume masking step; we manually determined the horizontal plume extent as a fan shape introducing fan’s angle (\(\theta\)) and the radius (i.e., \(l\)), although earlier studies separated the actual plume enhancement and noise using filtering.

We tried to identify plumes of XCO₂ and VCDNO₂ emitted from two individual stacks assuming Gaussian plume shape in the CSF method. For plume shapes to be sufficiently identified, two peaks of the enhancement from the two stacks should be individually detectable in the cross-section defined in Figure 1. Specifically, we performed optimal fits to the observed plume shapes of VCDNO₃ and XCO₂ for quantifying the emission from two different stacks, constraining a peak position and a full width at the half maximum (FWHM) of the Gaussian function. For CO₂, we also constrained the peak position and the FWHM of CO₂ using the NO₂ fitting results as initial fitting values. The emission rates were the cross-sectional area of the plumes (i.e., the integration of the fitted Gaussian function) multiplied by \(U_{\text{eff}}\). We considered this relationship appropriate in our case, though the uncertainty of this approximation may affect the total uncertainties. In the case of the NO₂ estimation, the calculated NO₂ emission was scaled by a factor of three to account for the NO/NO₂ partitioning (Text S4). Figure 2 shows the results of the cross-sectional Gaussian plume fitting for VCDNO₂ and XCO₂ using observations from the rectangle area shown in each panel of Figure 1. Individual plumes emitted from the two different stacks were well distinguished. The legend in Figure 2 shows the inverted and pre-calculated emission rates using the REAS v3.1 of CO₂ and NO₂ in units of kg s⁻¹ in red and blue text, respectively. In case of CO₂, the quantified emission rates were in relatively good agreement with the REAS v3.1 within −7% to 40%. For NO₂, however, we found a large discrepancy of more than 80% between the inverted emission rates and the REAS v3.1.

We also inferred the emission rates using the IME method. As mentioned above, we manually determined the plume mask—summed up as components of the plume, using the angle (\(\theta\)) and the plume size (\(l\))—which represents the plume extent and area. The emission rates estimated using \(\theta = 20°\) and \(l = 1,000\) m were 0.031, 0.034, 346, and 395 in units of kg s⁻¹, for the stack A and B of NO₂ and CO₂, respectively, showing relative differences of 15%–127% and −34% to 18% with the CSF, respectively.

For both inversion methods, the total error of the inverted NO₂ emission rates consists of the estimation error of the VCDNO₂ enhancements (0.891 × 10¹⁶ molec.cm⁻², or 36%, assuming typical VCDNO₂ as 2.5 × 10¹⁶ molec.cm⁻³) and the uncertainties of the NO₂ partitioning and the wind field. In this study, we assumed 100% partitioning uncertainty and 30% wind uncertainty. Moreover, we assumed that the first term mainly depends on the VCDNO₂ error, indicating that the fitting errors of the Gaussian plume fitting for the CSF
and the determination errors of plume shapes for the IME are both negligible. As a result, the calculated total error for NO\textsubscript{x} was 110%. In the case of CO\textsubscript{2}, however, the total error was smaller because the partitioning error does not need to be considered; therefore, the calculated total CO\textsubscript{2} error was 30%, consisting of the XCO\textsubscript{2} error (2%, assuming a typical XCO\textsubscript{2} value of 410 ppm) and the uncertainty of the wind field (30%).

Table 1 summarizes the estimated emission rates and those related errors of VCD\textsubscript{NO}\textsubscript{2} and XCO\textsubscript{2} estimated from the CSF and IME methods, in comparison to our estimation using the REAS v3.1. For the CO\textsubscript{2} inversion, it is obvious that the emission rates inversed by both CSF and IME methods were in relatively good agreement with the emission rates estimated from REAS v3.1 data. Conversely, for the NO\textsubscript{x} inversion, large discrepancies of 80% were found in both CSF and IME results. Possible reasons could be that the NO/NO\textsubscript{2} partitioning ratio largely varies in fresh plumes, depending on how plumes are processed and transported by the surrounding chemical and weather conditions; lack of information on surface albedo and a priori NO\textsubscript{2} profile with a high spatial resolution may also influence the AMF uncertainty. In addition, unidentified sources can be seen in the northern part of the region, which might contribute to the total uncertainties. Furthermore, the grating spectrometer and the optical system in the UV-visible region might need optimization because the ∼1.5 nm spectral resolution of the UV-VIS spectrometer was slightly coarse compared to the NO\textsubscript{2} observation using the DOAS.

Table 1

| Inversion method | Emission rate (kg s\textsuperscript{-1}) (uncertainty [kg s\textsuperscript{-1}]) |
|------------------|-------------------------------------------------|
|                  | NO\textsubscript{x} | CO\textsubscript{2} | NO\textsubscript{x} | CO\textsubscript{2} |
| Stack A          | Stack B           | Stack A | Stack B |
| CSF              | 0.027 (0.030)     | 0.015 (0.017)    | 523 (157) | 335 (101) |
| IME              | 0.031 (0.034)     | 0.034 (0.037)    | 346 (104) | 395 (119) |
| Estimate from REAS v3.1 | 0.19 (0.06) | 0.18 (0.06) | 373 (48) | 355 (46) |

Abbreviations: CSF, cross-sectional flux; IME, integrated mass enhancement; REAS, Regional Emission inventory in ASia v3.1.
5. Conclusions

We obtained airborne CO₂ and NO₂ observations over a coal fired power plant using grating spectrometers with UV-VIS and SWIR bands. The spectroscopic CO₂ and NO₂ data were analyzed with particular emphasis on the quantification of the columnar enhancements and the emission rates of CO₂ and NO₂ from the coal-fired power plant. Emission rates of CO₂ and NO₂ from the coal-fired FP were calculated using simplified methods similar to the CSF and IME method. The emission rates of CO₂ from two stacks were estimated to be 523 and 335 by the CSF method and 346 and 395 by the IME method, respectively, in units of kg s⁻¹. Both methods were in relatively good agreement with the REAS v3.1 emissions inventory. However, the NO₂ emission rates estimated by both methods showed large discrepancies (>80%) with those of the REAS v3.1. These results indicated that the variability of the NO/NO₂ partitioning ratio and the lack of information on surface albedo, surface properties, and spatially resolved wind field have significant effect on the inversion of the emissions in case of flux inversion using remote sensing of high spatial resolution. It is also possible that the discrepancy is due to inaccurate activity data and emission factors related to power plants in a bottom-up inventory. In case of highly resolved observations within a kilometer scale like in this study, therefore, the NO₂ partitioning, as well as the surface properties and wind field with a high resolution, need to be considered. Further observations of CO₂ and NO₂ with aircraft-, ground-, and satellite-based remote sensing, and the synergetic use of these observations are essential for a better understanding of non-linear chemistry and transport processes of emitted gases around a point source. This study results imply possibilities that observations with a high spatial resolution of km-scale in the future enable us to identify CO₂ plumes emitted from the point source of interest with a stack-level scale even under the existence of the other anthropogenic sources by using the NO₂ plume constraint as well as bottom-up emission inventories.

Conflict of Interest

The authors declare no conflicts of interest relevant to this study.

Data Availability Statement

TCR-2 reanalysis data are available at https://ebcrpa.jamstec.go.jp/tcr2/. The GSM and MSM from JMA are available at https://tes.jpl.nasa.gov/tes/chemical-reanalysis/products/monthly-mean/.

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