Influences of aerosols and thin cirrus clouds on GOSAT XCO₂ and XCH₄ using Total Carbon Column Observing Network, sky radiometer, and lidar data

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ABSTRACT
The column-averaged, dry air mole fractions of CO₂ and CH₄ (XCO₂ and XCH₄, respectively) were retrieved from short-wavelength infrared (SWIR) spectra observed by the Greenhouse gases Observing SATellite (GOSAT). Continuous measurements of SWIR spectra have been made via GOSAT since 2009, but there has been insufficient investigation of the effects of cirrus clouds and aerosols on the observations. In this work, we investigated the influences of aerosols and cirrus clouds on the differences between GOSAT observations and Total Carbon Column Observing Network (TCCON) data for XCO₂ and XCH₄ (ΔXCO₂ and ΔXCH₄) at three sites: Tsukuba and Saga in Japan, and Lauder in New Zealand. We used aerosol optical thickness (AOT), Angstrom exponents (AEs), and single scattering albedo (SSA), all obtained from sky radiometer observations, as well as vertical profiles of aerosols and thin cirrus clouds from lidar observations. Matchups were performed within ±0.1° latitude/longitude rectangular areas of each TCCON site, and within 30 min of the GOSAT overpass time. The results show a negative slope between ΔXCO₂ and AOT at 500 nm determined from sky radiometer data at Tsukuba and Saga. The GOSAT XCO₂ values tended to be lower in the presence of cirrus clouds and dense boundary-layer aerosols. Moreover, a significant negative ΔXCO₂ was observed at times of large AOTs that resulted from dust-like events. At Lauder, ΔXCO₂ was negatively correlated with the AOT at 500 nm, although the AOT at this site was generally small. The mean ± standard deviation for ΔXCO₂ and ΔXCH₄ at Lauder are −0.80 ± 1.83 (ppm) and −5.27 ± 10.79 (ppb) with correlation coefficients r between GOSAT and TCCON of 0.94 and 0.83, respectively. Both ΔXCO₂ and ΔXCH₄ were significantly and negatively correlated with the AOT during Sep-Oct-Nov. In addition, stratospheric aerosols caused large negative biases of ΔXCO₂ and ΔXCH₄ at Lauder despite the small stratospheric aerosol optical depth at that site.

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Supplemental data for this article can be accessed here.

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1. Introduction

Greenhouse gases are a group of gases including carbon dioxide, methane, ozone, nitrous oxide, water vapour, and fluorinated gases that cause climate change and global warming (IPCC 2014; Myhre et al. 2013; Schmidt et al. 2010). Carbon dioxide (CO₂) and methane (CH₄) are the most prevalent anthropogenic greenhouse gases that are currently major drivers of climate change. Concentrations of these gases have increased rapidly during the Anthropocene. The global annual mean of surface CO₂ concentrations breached the threshold of 400 ppm in 2015 and exceeded 410 ppm in 2019, while the global annual mean of surface CH₄ concentrations in 2019 (1877 ppb) was 260% of pre-industrial level (WMO 2020). The current rate of increase in global CO₂ concentrations is unprecedented in the history of World Meteorological Organization (WMO) records.

Precise measurements of CO₂ and CH₄ concentrations are important for estimating emission intensity, flux changes, and the impact of CO₂ and CH₄ on climate change (Gillett and Matthews 2010; Jonas et al. 2019; Weiss and Prinn 2011; Zhao et al. 2009). Ground-based in situ observations fully meet the precision requirements for these purposes. Although the number of accurate, ground-based observations has been steadily increasing, the spatial distribution of the data is still sparse. In contrast, satellite-based observations have been very effective in providing global distributions and temporal variations of greenhouse gas concentrations, but they are less accurate than ground-based measurements and need to be validated against ground-truth measurements (Morino et al. 2011; Schneising et al. 2012; Wunch et al. 2017).

The Greenhouse gases Observing SATellite (GOSAT) is the first satellite dedicated mainly to monitoring the atmospheric concentrations of greenhouse gases such as CO₂ and CH₄ from space. Column-averaged dry-air mole fractions of CO₂ and CH₄ (denoted XCO₂ and XCH₄, respectively) were retrieved from the Short-Wavelength InfraRed (SWIR) spectra observed by the Thermal And Near-infrared Sensor for carbon Observation – Fourier Transform Spectrometer (TANSO-FTS) on board the GOSAT. Use of these data by the scientific community requires that their uncertainties must be clarified through validation (Inoue et al. 2013, 2014; Parker et al. 2011).

The Total Carbon Column Observing Network (TCCON) data have been used extensively for GOSAT product validation (Inoue et al. 2014; Morino et al. 2011; Yoshida et al. 2013). High-quality XCO₂ and XCH₄ data obtained independently by ground-based instruments associated with the TCCON have uncertainties (2σ) of 0.8 ppm for XCO₂ and 7 ppb for XCH₄ (Wunch et al. 2010). The ground-based sky radiometers observation network (SKYNET) measures mainly aerosol optical properties such as aerosol optical thickness (AOT), Angstrom exponent (AE), and single scattering albedo (SSA), which are dedicated not only to aerosol-cloud-radiation interaction studies but also to validations of satellite data (Takamura et al. 2009).

Previous studies have shown that the vertical distributions of aerosol and cirrus, especially those of high-altitude aerosols and thin cirrus clouds, have large effects on GOSAT SWIR XCO₂ data. Uchino et al. (2012a) investigated the influence of vertical aerosol profiles and thin cirrus clouds observed by lidar and sky radiometer on GOSAT SWIR XCO₂ data (Version 01.xx) which were largely underestimated as shown by Morino et al. (2011). Uniformly distributed aerosols below an altitude of 2 km and no cirrus clouds assumed in the GOSAT SWIR retrieval algorithm of this version may be one of the main causes. The
difference between the GOSAT SWIR XCO₂ data and the Tsukuba TCCON data was reduced to two-thirds when considering the observed aerosol/cirrus vertical profiles in the GOSAT SWIR retrieval. The effects of aerosols as well as cirrus clouds on the accuracy of retrieved CO₂ and CH₄ data have been considered in several previous studies. O’Brien and Rayner (2002) evaluated the error due to undetected atmospheric scattering by optically thin cirrus and aerosol. In their study, the error in the estimated CO₂ column caused by the optical thicknesses of aerosol and cirrus as large as 0.2 and 0.1, respectively, is approximately 0.5%. Dufour and Breon (2003) also concluded that atmospheric scattering is a major source of error in monitoring CO₂ concentration from space. They quantified an error of 4 ppm for a cirrus cloud with an assumed optical thickness of 0.02 and an error of 1.5 ppm for a boundary-layer aerosol at an average height of 1 km with an optical thickness of 0.05. Additionally, Huang et al. (2020) concluded that the presence of aerosols causes an underestimation of CH₄ in retrievals and the biases increase with increasing surface albedo and aerosol optical depth.

Many retrieval algorithms have been developed and revised to account for scattering by aerosols in GOSAT data (Butz et al. 2009, 2010; Yoshida et al. 2011, 2013). GOSAT measures the absorption of sunlight reflected from surfaces and scattered from the atmosphere, whereas TCCON measures the absorption of direct sunlight by trace gases. Hence, GOSAT observations are more affected by scattering due to aerosols and cirrus clouds. To incorporate treatment of aerosol profile, properties of aerosol particles such as amount, size, and height were parameterized and then retrieved simultaneously with XCO₂ (Butz et al. 2009; Guerlet et al. 2013). Based on simple assumptions of a uniform distribution of aerosols below an altitude of 2 km and no cirrus clouds in version 01.xx algorithm (Yoshida et al. 2011), the aerosol vertical profiles and a solar irradiance database were updated in an improved algorithm, version 02.xx (Yoshida et al. 2012, 2013). Although significant improvements have been achieved, these algorithms tend to overestimate aerosol amounts, resulting in an underestimation of GOSAT XCO₂ (Oshchepkov et al. 2013). If atmospheric light-scattering by aerosols and clouds is not fully considered, errors in GOSAT retrievals will remain. We thus compared the XCO₂ and XCH₄ data from GOSAT and TCCON and then carried out a regression analysis of their differences such as ΔXCO₂ (XCO₂ GOSAT – XCO₂ TCCON) and ΔXCH₄ (XCH₄ GOSAT – XCH₄ TCCON), respectively, over sites at Tsukuba and Saga in Japan, and Lauder in New Zealand. We examined the relationships between these differences and AOT, AE, and SSA from sky radiometer data to determine whether GOSAT XCO₂ and XCH₄ data were significantly influenced by aerosol optical properties. Thanks to concurrent observations by GOSAT and TCCON as well as sky radiometer and lidar data at Tsukuba, Saga, and Lauder over a long period of time, we were able to investigate the influence of aerosols and cirrus clouds on GOSAT XCO₂ and XCH₄ retrievals using aerosol/cloud vertical profiles from lidar. The purpose of this study was to examine the effects of aerosol optical properties as well as the impact of thin cirrus clouds on the ΔXCO₂ and ΔXCH₄.

The content of this paper is structured as follows: in Section 2, we introduce GOSAT observation data and the additional datasets that we investigated, and we provide an overview of the sites that we studied. Section 3 presents an overview of the analytical methods. In Section 4, we focus on the main results from the detail of aerosol optical properties and the vertical profiles of aerosols and clouds at the study sites. We also discuss the influence of these aerosol optical properties on XCO₂ and XCH₄ retrievals as
well as the reasons for aerosol episodes of high AOT values at the study sites. Finally, Section 5 discusses expectations for future improvements in XCO₂ and XCH₄ retrievals from GOSAT series data.

2. Observational data and study sites

2.1. Observational data

2.1.1. GOSAT data (version V02.90)
GOSAT was launched in January 2009 in a Sun-synchronous orbit with a three-day repeat at an inclination of 98°, an altitude of 666 km, and an equator crossing at 12:48 PM, local time. The Thermal And Near-infrared Sensor for carbon Observation (TANSO) instrument onboard GOSAT includes a Fourier Transform Spectrometer (FTS) and a Cloud and Aerosol Imager (CAI). The FTS measures gas absorption spectra of solar light reflected on the surface and scattered by the atmosphere in SWIR (wavelengths of 0.76, 1.6, and 2.0 µm) and emission from the surface and atmosphere in the thermal infrared (TIR) (wavelengths of 5.5–14.3 µm), whereas the CAI measure radiances interacting with clouds and aerosols (Kuze et al. 2009). After cloud screening by CAI observations, GOSAT TANSO-FTS SWIR XCO₂ and XCH₄ data are retrieved from the SWIR spectra observed by the GOSAT TANSO-FTS (Yoshida et al. 2011, 2013). The data were obtained from the GOSAT Project webpage (http://www.gosat.nies.go.jp/en/). GOSAT data are divided into ocean data (soundings over the ocean), land data (soundings over land), and mixed data (soundings over a mixture of land and ocean). The data were acquired in either high (H) or medium (M) gain modes. The latest version of the GOSAT data (V02.90) covers a timeframe of more than 10 years. Only the H-gain land data recorded from 23 April 2009 to 31 December 2019 were used in this study over the three TCCON sites.

2.1.2. TCCON data
TCCON is a worldwide network of ground-based, high-resolution FTS recording direct solar spectra in the near-infrared spectral region, the primary purpose of which is to provide a reliable, long-term time series of column-averaged dry air mole fractions of various atmospheric constituents for carbon cycle studies and the evaluation of satellite observations. These constituents including XCO₂ and XCH₄, are retrieved from near-infrared solar absorption spectra using a nonlinear least-squares fitting algorithm GFIT (Wunch et al. 2010, 2011). The TCCON data are available at the TCCON data archive (https://tccondata.org/). Version GGG2014 of the TCCON dataset was used as the reference dataset in this study (Morino, Matsuzaki and Horikawa 2018; Pollard, Robinson and Shiona 2019; Sherlock et al. 2014a, 2014b; Shiomi et al. 2014; Wunch et al. 2011).

2.1.3. Sky radiometer data
The SKYNET is a ground-based atmospheric radiation observation network that obtains optical and microphysical properties of aerosols and clouds. The standard instrument of each SKYNET site is the sun-sky radiometer. We used sky radiometer data from the SKYNET Research Product Release Service at the Center for Environmental Remote Sensing, Chiba University (http://atmos3.cr.chiba-u.jp/skynet/index.html). Sky radiometers (models POM-01 and POM-02, PREDE Co., Ltd., Tokyo) were used to measure solar and sky irradiance, including
the influence of aerosols and clouds. SKYNET serves as a reference for satellite validation as well as climate modelling simulations and data assimilation (Takamura and Nakajima 2009). Values of AOT, SSA, and AE were retrieved from these irradiances using the inversion package SKYRAD.pack (Nakajima et al. 2015). This study used AOT, SSA at a wavelength of 500 nm, and AE, which is derived from a multispectral log linear fit to $AOD \sim \lambda^{-AE}$ at seven wavelengths of 340 nm, 380 nm, 400 nm, 500 nm, 675 nm, 870 nm, and 1020 nm.

### 2.1.4. Lidar data and sky view images

Lidar acquires the vertical profiles of aerosols and thin cirrus clouds. In this study, we used lidar data to investigate the relationship between GOSAT data and vertical profiles of aerosols and thin cirrus clouds. The lidar data used in this analysis were polarized, Mie-scattering lidar data obtained with a neodymium-doped, yttrium-aluminium-garnet (Nd:YAG) laser with two operating wavelengths of 532 and 1064 nm. This instrument can be measured with a vertical resolution of 150 m. Lidar can detect both tropospheric and stratospheric aerosols. While tropospheric aerosols can be detected during daytime and nighttime, stratospheric aerosols can only be detected in the nighttime if there are no thick clouds (Sakai et al. 2016; Uchino et al. 2017). The backscattering ratio $R$ is defined by Equation (1):

$$R = \frac{\langle BR + BA \rangle}{BR}$$

(1)

where $BR$ and $BA$ are the Rayleigh and particle backscattering coefficients, respectively. The stratospheric aerosol optical depth (SAOD) is defined by Equation (2)

$$SAOD = s \times IBC$$

(2)

where $s$ is the aerosol extinction-to-backscatter ratio (lidar ratio). The value of $s$ over Lauder was assumed to be 46 steradians (Sakai et al. 2016). The integrated backscattering coefficient (IBC) of stratospheric aerosols is obtained by integrating up $BA$ from the first tropopause to 33 km, as written in Equation (3):

$$IBC = \int_{Tppause}^{33\text{km}} BA \, dz = \int_{Tppause}^{33\text{km}} (R - 1)BR \, dz$$

(3)

The parallel and perpendicular components of the backscattered signals $P$ and $S$ are used to calculate the total depolarization ratio (Dep) according to Equation (4):

$$Dep = \frac{S}{(P + S)} \times 100(\%)$$

(4)

From the measured vertical profiles of $R$ and $Dep$, it is possible to derive the distribution of atmospheric particles such as aerosols or clouds, including cirrus clouds (Uchino et al. 2012b).

Ground-based sky cameras set up in Tsukuba, Saga, and Lauder captured images during GOSAT soundings for cloud-cover assessment (Letu et al. 2014).
2.2. Ground-Based observation sites used in this study

Ground-based FTS, sky radiometer, and lidar instruments that were installed at prioritized GOSAT validation sites such as Tsukuba and Saga in Japan and Lauder in New Zealand (Figure 1) provided data that were used in this study.

The Tsukuba TCCON FTS was set up at the National Institute for Environmental Studies (NIES) (36.051° N, 140.122° E, 31 m a.s.l.), about 60 km from the northeastern edge of Tokyo, Japan. A POM-02 sky radiometer and a lidar were installed at the Meteorological Research Institute (MRI) (36.056°N, 140.125°E, 25 m a.s.l.), a distance of 500 m from the Tsukuba TCCON site. There are no sources of major anthropogenic emissions in Tsukuba. Depending on meteorological conditions, air masses from the Tokyo metropolitan area sometimes pass over Tsukuba because of its proximity to Tokyo. In summer, the dominant Pacific air mass brings clean air to Tsukuba (Naja and Akimoto 2004).

The Saga TCCON, which is operated by the Japan Aerospace Exploration Agency and Saga University, was set up at Saga University (33.241° N, 130.288° E, 8 m a.s.l.) on the northwest side of the island of Kyushu, Japan, where a Mie lidar and a sky radiometer, both operated by the NIES and Saga University, were also installed. Air masses over Saga are transported mainly from the Asian continent, except during the summer, when the air mass comes primarily from the Pacific Ocean (Uchino et al. 2014).

The Lauder TCCON, which is operated by the National Institute of Water and Atmospheric Research (NIWA), is located at 45.038° S, 169.682° E, 370 m a.s.l. on South Island, New Zealand, in a valley with mountains on three sides. The lidar is operated by the MRI, NIES, and NIWA; the sky radiometer is operated by the NIES and NIWA. The Lauder area is sparsely populated. Because it is shielded from the prevailing winds by the Southern Alps, the area has a high number of clear-sky days and a clean air mass (Liley and Forgan 2009; Pollard et al. 2013).

Figure 1. Ground-Based observation sites and their satellite images (Google Maps images (map data: Google, Maxar technologies)).
3. Method for comparing GOSAT data with TCCON, sky radiometer, and lidar data

With regard to the data release, data comparisons were made primarily between GOSAT TANSO-FTS SWIR XCO₂ and XCH₄ data (Ver. 02.90) from April 2009 to December 2019 in land (H and M gain) and ocean areas within ±2.0° and ±5.0° latitude/longitude boxes centred at the TCCON sites and the mean of the TCCON data within ±30 min of the GOSAT overpass time. These results have been summarized on the GOSAT Data Archive Service (GDAS) website at https://data2.gosat.nies.go.jp/doc/documents/ValidationResult_FTSSWIRL2_V02.90_GU_en.pdf

In this study, we evaluated the influence of aerosols and cirrus clouds on GOSAT XCO₂ and XCH₄ data using a combination of TCCON data, sky radiometer data, lidar data, and sky-view images. To exclude the spatial difference of aerosols and cirrus clouds, we first compared in a stricter criterion GOSAT XCO₂ and XCH₄ data, which we acquired within ±0.1° latitude/longitude boxes centred at the Tsukuba, Saga, and Lauder sites, with the mean of the TCCON data acquired within ±30 min of the GOSAT overpass time. Thereafter, we calculated ΔXCO₂ and ΔXCH₄. In contrast to the reflected-sunlight observations of GOSAT, the direct sunlight observations in the same spectral region of TCCON are minimally influenced by clouds and aerosols. We hypothesized that the ΔXCO₂ and ΔXCH₄ were caused partly by clouds and aerosols. Next, we used sky radiometer data to perform an analysis of the influences of AOT, SSA at a wavelength of 500 nm, and AE in the wavelength range of 340–1020 nm on ΔXCO₂ and ΔXCH₄. AOT and SSA data at 500 nm and AE from the sky radiometer were also collected at each site and matched in the same way as the GOSAT XCO₂ and XCH₄ data to the TCCON data. Finally, lidar data and sky-view images at the three sites were selected at the nearest GOSAT overpass time to check the vertical profiles of clouds, aerosols, and sky conditions. We determined whether the effects of clouds/aerosols on GOSAT XCO₂ and XCH₄ data were statistically significant based on the changes of ΔXCO₂ and ΔXCH₄ in the presence of clouds/aerosols.

4. Results and discussion

The matched-up data that could be compared were limited to the time intervals from May 2013 to November 2018 at Tsukuba, from February 2013 to May 2019 at Saga, and from March 2012 to June 2019 at Lauder because of the severe criteria for collocation and the selection of land data for analysis. Table 1 lists the number of matched datasets between GOSAT, TCCON, and sky radiometer data. The matched-up data of GOSAT and TCCON for XCO₂ and XCH₄ as well as AOT, SSA, and AE from sky radiometer at each site

| Site   | N  | Season | Data period         |
|--------|----|--------|---------------------|
|        | All Seasons | DJF | MAM | JJA | SON | GOSAT (0.1°×0.1°) | TCCON | sky radiometer |
| Tsukuba| 119 | 47    | 39  | 5   | 28  | 09/2009 – 12/2019  | 08/2011 – 12/2018  | 01/2009 – 12/2018 |
| Saga   | 46  | 5     | 25  | 3   | 13  | 03/2011 – 10/2019  | 07/2011 – 05/2019  | 03/2011 – 12/2019  |
| Lauder | 161 | 20    | 69  | 46  | 26  | 05/2009 – 09/2019  | 06/2004 – 06/2019  | 10/2011 – 12/2019  |
were incorporated into 3-month calendar seasons Dec–Jan–Feb (DJF); Mar–Apr–May (MAM); Jun–Jul–Aug (JJA); Sep–Oct–Nov (SON). Seasonal values are then computed by averaging data of ΔXCO₂, ΔXCH₄ and AOT, SSA, AE over each season.

There were 10 match-ups of lidar data at Tsukuba, 26 at Saga, and 78 at Lauder. The maximum values of ΔXCO₂ and ΔXCH₄ were selected in the case of multiple match-ups with lidar data. In addition, we have sky-view images, which provide existing visible clouds for almost all the lidar profiles at these sites. Unlike previous studies that only focus on one or two profiles at one site, this study presents three sites from the Northern Hemisphere to the Southern Hemisphere with many vertical profiles of aerosols which are useful to compare aerosol distribution and its effect between studied sites. The lidar profiles and sky-view images for all cases of aerosols/cirrus clouds and clear-sky conditions during this study are included in the Supplementary materials. All lidar profiles at Tsukuba and Saga were appeared by aerosols or cirrus (Table S1, Table S2). Based on the lidar data and sky view images at Lauder, we were able to identify clear-sky days, which were characterized by a cloud-free atmosphere with background concentrations of aerosols. This is ‘clear-sky’ condition used in the present study. There were 36 clear-sky days at this site. We define matched data collected on such days as clear-sky data. We got 64 clear-sky data from a total of 161 matched datasets at Lauder (Table S3).

### 4.1. Tsukuba site

We performed a regression analysis with either ΔXCO₂ or ΔXCH₄ as the dependent variable and either AOT, SSA, or AE as the independent variable for all seasons as well as for each season. We tested the null hypothesis that there is no relationship between the dependent and independent variables using t-tests. We reject the null hypothesis if the type I error rate (P-value) is less than 0.05.

When data for all seasons were included in the analysis, there was a significant linear relationship and a negative correlation between ΔXCO₂ and AOT at 500 nm. The slope of the regression line was −3.58 ppm, and the Pearson correlation coefficient (r) was −0.23 (Figure 2(a)). The slope was statistically significant (t-test, $P = 0.01$). In Figure 2(b), there was also a significant linear relationship between the ΔXCO₂ for all seasons and the SSA at 500 nm. The slope of −6.25 ppm and $r$ of −0.21 were both significant at $P = 0.02$. In Figure 2(c), the $P$-value exceeded 0.05; hence, there was no significant relationship between ΔXCO₂ for all seasons and the AE at Tsukuba (Figure 2(c), $P = 0.77$). For separate seasons, there was a significant relationship between ΔXCO₂ and AOT in MAM with the slope of the regression line being −3.87 ppm at $P = 0.02$.

Similar analyses of the relationships of ΔXCH₄ versus AOT, SSA, and AE for all seasons and for separate seasons at Tsukuba revealed no significant relationships ($P > 0.05$).

In Figure 3, the seasonal means of ΔXCO₂ were negative during MAM and JJA, and they were positive during SON and DJF, although in all cases they differed from zero by less than one standard deviation. Seasonal means of AOT at 500 nm at Tsukuba (Figure 4) were relatively large during MAM and JJA, the seasons during which the seasonal means of ΔXCO₂ were negative (Figure 3). The reason during MAM and JJA might be that there were many days with high AOT level (Fukagawa et al. 2006) when atmospheric scattering makes the shortening of the atmospheric path. The consequence is underestimate of GOSAT XCO₂ while values of the TCCON data don’t change by the air pollution and dust
since TCCON data is retrieved from direct solar spectra. The implication is that GOSAT XCO₂ values tended to be less than TCCON XCO₂ values when the AOT was large in MAM and JJA. The small values of the seasonal mean AOT at 500 nm in SON and DJF (Figure 4) corresponded to the positive seasonal means of ΔXCO₂ (Figure 3). The implication is that GOSAT XCO₂ values tended to be larger than TCCON XCO₂ values when AOT was small during these seasons.

The mean AE for all the season was 1.35 at Tsukuba (Figure 4) which indicates small particles in the atmosphere prevailed as pollutants, and this site was also affected by mostly urban aerosols from Tokyo nearby (Naja and Akimoto 2004).

Figure 2. Relationship of ΔXCO₂ with AOT, SSA at 500 nm, and AE at Tsukuba for all seasons (a, b, c) and separate seasons (a', b', c').
The time series of ΔXCO₂, AOT, and SSA at Tsukuba (Figure 5) reveals a large negative ΔXCO₂ on 1 May 2016 and a maximum AOT of 0.54 on 28 March 2018. We selected these days to examine the negative correlations between ΔXCO₂ and both AOT and SSA at this site.
4.1.1. 1 May 2016

We chose to investigate the $\Delta$XCO$_2$ of $-3.95$ ppm at Tsukuba on 1 May 2016 and the corresponding values of AOT (0.49) and SSA (0.96). Because $\Delta$XCO$_2$ was negatively correlated with both AOT and SSA (Section 4.1), the large AOT and SSA values on this day would have been expected to lead to a negative $\Delta$XCO$_2$ and the $\Delta$XCO$_2$ on this day ($-3.95$ ppm) was unexpectedly very negative. The implication is that either the large AOT or large SSA could have led to the negative $\Delta$XCO$_2$ on 1 May 2016. Moreover, lidar observations on 1 May 2016 captured a dense tropospheric aerosol layer at an altitude $<3.0$ km ($R = 2.5–3.0$, Dep = 2.9–7.0) during the day at the time of the GOSAT overpass (Figure 6(a)), and there was haze in the sky-view image on that day (Figure 6(e)). Figure 6(b) shows the background level of stratospheric aerosols over Tsukuba at night on 1 May 2016. The high top of the boundary layer and the aerosols included therein might have resulted in a very negative $\Delta$XCO$_2$ on that day, as suggested by Sakai et al. (2013). A similar example has been reported by Ohyama et al. (2015, Figure 10(a)) which $\Delta$XCO$_2$ reached $-4.82$ ppm at Saga with a similar aerosol vertical profile uniformly distributed below a height of 3 km, although the boundary layer aerosols (AOD = 1.28) in Ohyama et al. (2015) were denser than at Tsukuba. A low-altitude, dense aerosol layer with a large AOT could therefore have led to a large negative bias of the GOSAT XCO$_2$.

4.1.2. 28 March 2018

This day was chosen to investigate the effects of the maximum AOT of 0.54 and SSA of 0.91 at Tsukuba on the corresponding $\Delta$XCO$_2$ of $-2.18$ ppm. The large AOT in this case would have been expected to lead to a very negative $\Delta$XCO$_2$. However, the actual $\Delta$XCO$_2$ of $-2.18$ ppm on this day was not very negative. Lidar observations on 28 March 2018 captured a dense aerosol boundary layer ($R = 1.0–4.9$, Dep = 5.0–18.0) at an altitude $<1.5$ km during the day at the time of the GOSAT overpass (Figure 6(c)). There was a haze in the sky-view image on that day (Figure 6(f)). This aerosol boundary layer was likely composed of dust particles because the large Dep was an indication of non-spherical particles. Figure 6(d) shows the
Figure 6. Vertical profiles of the backscattering ratio (R) and total depolarization ratio (Dep) at 532 nm and the sky-view images at Tsukuba recorded by the Meteorological Research Institute. Blue lines in the panels (a)–(d) are the height of the first tropopause. LT = local time.
background level of stratospheric aerosols over Tsukuba at night on 28 March 2018. The boundary layer including dense aerosols appeared at lower altitude on 28 March 2018 than on 1 May 2016. It may be the reason for ∆XCO₂ of −2.18 ppm with AOT of 0.54 on 28 March 2018, while ∆XCO₂ on 1 May 2016 was −3.95 ppm with AOT of 0.49. Hence, the optical path modification due to atmospheric light scattering as well as the impact of atmospheric light scattering on XCO₂ retrieval on 28 March 2018 was less than on 1 May 2016. Oshchepkov et al. (2011) argued that near-ground aerosols with large AOT values might not modify the optical path significantly, in contrast elevated aerosols or thin clouds with small AOT values might modify the optical path significantly. The combination of the large AOT and the low-altitude, dust-like event might explain the negative bias of the GOSAT XCO₂.

4.2. Saga site

We performed regression analyses in the same way at the Saga and Tsukuba sites and used a type I error rate of <0.05 as a criterion for the statistical significance of the regression slopes.

The regression analysis of ∆XCO₂ versus AOT for all seasons revealed a negative slope of −2.98 ppm (Figure 7(a)). The type I error rate of this slope for a one-sided t-test was 0.027. On the assumption that the slope could not have been positive, we concluded that this negative slope between ∆XCO₂ and AOT was significant. The associated Pearson correlation coefficient was −0.29. No significant slope was found between ∆XCO₂ and SSA (Figure 7(b)) because the P value exceeded 0.05. The regression slope between ∆XCO₂ and AE was −1.55 ppm (Figure 7(c)). The slope was significant (P = 0.024) based on a two-sided t-test and the associated Pearson correlation coefficient was −0.33. The ∆XCO₂ at the Saga site decreased with increases of either AOT or AE.

For separate seasons, regression analyses of ∆XCO₂ versus AOT, SSA, and AE at Saga revealed no significant relationships (P > 0.05).

Regression analyses of ∆XCH₄ versus AOT, SSA, or AE revealed no significant relationships during all seasons as well as during separate seasons at the Saga site. These results are similar to the results of the analogous regression analyses at Tsukuba.

Furthermore, the ∆XCO₂ seasonal means (Figure 8) were negative in MAM and JJA and positive in SON and DJF, although the standard deviation were larger than the magnitudes of the means. The relationship between ∆XCO₂ and AE for all seasons was significant at Saga, and the positive ∆XCO₂ in DJF corresponded to the smallest AE in Figure 9. However, the fact that there were only three data in JJA precluded concluding that the large AE (small particulates) was the cause of the very negative ∆XCO₂ in JJA (Figure 8). While the mean AE for all seasons was 1.03, hence Saga was affected by both small and large particles such as long-range transported anthropogenic pollutants and dust aerosols (Pan et al. 2016).

The significant, negative slope of ∆XCO₂ versus AOT at Saga was consistent with the correspondence between the negative ∆XCO₂ in MAM and the largest seasonal mean value of AOT (Figure 9).

Figure 10 shows the time series of ∆XCO₂, AOT, and AE at Saga. Two atypical cases were selected for a detailed analysis: 14 June 2017, when the ∆XCO₂ was most negative, and 26 March 2018, when the AOT was the maximum.
4.2.1. 14 June 2017

This case was chosen because the most negative $\Delta XCO_2$ (−4.40 ppm) was recorded on this day and the analysis in Section 4.2 indicated that the $\Delta XCO_2$ at Saga was negatively correlated with both AOT and AE. The AOT value of 0.31 on 14 June 2017 was identical to the average AOT during the period of comparison, but the relationship between $\Delta XCO_2$ and AOT suggests that the $\Delta XCO_2$ should be close to zero when the AOT is 0.31 (Figure 7 (a)). Moreover, the relatively large AE of 1.27 on this day should also lead to a $\Delta XCO_2$ close to zero based on the regression line in Figure 7(c). However, the actual $\Delta XCO_2$ on 14 June 2017 was −4.40 ppm. This very negative $\Delta XCO_2$ might have been caused by factors other than aerosols. Indeed, the vertical profiles of R and Dep observed by lidar on this day revealed cirrus clouds at 10.5 km (R = 5, Dep = 25–30) and thin tropospheric aerosols below an altitude of 3 km (R = 2.0–2.4, Dep = 3.0–4.4) at 13:30 (LT) when the

Figure 7. Relationship of $\Delta XCO_2$ with AOT, SSA at 500 nm, and AE at Saga for all seasons (a, b, c) and separate seasons (a’, b’, c’).
GOSAT passed over (Figure 11(a)). At the same time, haze and cirrus clouds can be seen in sky-view images (e.g. Figure 11(e)). During the night, stratospheric aerosols over Saga were present at the background level (Figure 11(b)). The very negative \( \Delta XCO_2 \) was likely caused by the cirrus clouds at Saga because the cirrus clouds were located at a higher altitude than the thin aerosols and their effect on the GOSAT XCO\(_2\) was therefore greater. Uchino et al. (2012a) have reached a similar conclusion. Moreover, because light scattered by cirrus clouds traverse a shorter path through the atmosphere than light that is not scattered, less of the scattered light is absorbed by CO\(_2\). The result is an underestimation
4.2.2. 26 March 2018

An unusually high AOT of 0.87 and a relatively low AE of 0.76 were observed on 26 March 2018. These values are a little more extreme than the corresponding values reported by Suzuki et al. (2004) during Eolian dust events in April 1998 and April 1999 in southern Japan, when the AOT was less than 0.84 and the AE was 0.2–0.5. Dust particles are usually large particles and are transported in the troposphere (Iwasaka et al. 2003). Indeed, there was a severe dust event during 22–27 March 2018 that resulted from dust storms over the Gobi and Sahara deserts and transport of that dust to Japan (Sugimoto et al. 2019).

Because the ΔXCO₂ at Saga was negatively correlated with AOT and AE, the maximum AOT of 0.87 on 26 March 2018 would have usually led to a very negative ΔXCO₂ whereas the relatively low AE of 0.76 on that day would usually have led to a positive ΔXCO₂. The combination of a large AOT and a small AE led to a ΔXCO₂ on 26 March 2018 of −0.21 ppm. The vertical profiles of R and Dep observed by lidar during the day of 26 March 2018 revealed three factors that could have affected the ΔXCO₂: thin cirrus clouds at an altitude of 7.5 km (R = 5, Dep = 12–19); tropospheric aerosols at 3–5 km (R = 2, Dep = 5–7); and dense, boundary-layer aerosols below 2 km (R = 6–6.5, Dep = 5–7) (Figure 11(c)). The low AE and the large Dep in the troposphere of Figure 11(c) also indicate the presence of large, non-spherical, dust-like particles at Saga on that day. Aerosols were also apparent in sky-view images (e.g. Figure 11(f)). During the night, the stratospheric aerosols over Saga were at background levels (Figure 11(d)). The maximum AOT and corresponding ΔXCO₂ of −2.18 ppm at Tsukuba (Section 4.1.2) seem to have occurred for the same reasons as the large AOT in this case—a dust event—because the depolarization ratio in the troposphere in both cases indicated the presence of dust-like particles. Therefore, the large AOT

![Figure 10. Time series of ΔXCO₂, AOT, and and AE at Saga.](image-url)
Figure 11. Vertical profiles of the backscattering ratio (R) and total depolarization ratio (Dep) at 532 nm and the sky-view images at Saga recorded by NIES and Saga University. Blue lines in panels (a)-(d) are the height of the first tropopause. LT = local time.
associated with a dust-like event might have resulted in the negative ΔXCO₂. However, the complex lidar profile on this day revealed complicated effects on the GOSAT XCO₂ values. That is, the presence of cirrus clouds and dense, boundary-layer aerosols tends to cause a large negative ΔXCO₂ but the actual ΔXCO₂ was just −0.21 ppm. Therefore, the effect of the aerosol height distribution on ΔXCO₂ (i.e. the tropospheric aerosols at 3–5 km) required further investigation to understand the pattern of ΔXCO₂ on this day.

### 4.3. Lauder site

Unlike the air at the Tsukuba and Saga sites, the air at Lauder was clean. The AOT in this area is generally small (<0.2), and this site has many days of clear sky (i.e. cloud-free conditions with few aerosols in the atmosphere). We have calculated the mean ± standard deviation of the clear-sky ΔXCO₂ and ΔXCH₄ at Lauder to be −0.23 ± 1.45 ppm and −1.69 ± 9.70 ppb, respectively. For all the data, the mean ± standard deviation for ΔXCO₂ and ΔXCH₄ at this site are −0.80 ± 1.83 (ppm) and −5.27 ± 10.79 (ppb) with correlation coefficients r between GOSAT and TCCON of 0.94 and 0.83, respectively.

We also performed regression analyses at Lauder of ΔXCO₂ and ΔXCH₄ versus either AOT, SSA at 500 nm or AE for all seasons and for separate seasons. The regression slope was −24.34 ppm between ΔXCO₂ and AOT with an r of −0.32 and a P of 3.3 × 10⁻⁵ (Figure 12(a)). There was no significant relationship between ΔXCO₂ and either SSA or AE at Lauder (P > 0.05, Figures 12(b) and (c)). The slope of the regression of ΔXCH₄ versus AOT at Lauder was −82.64 ppb (Figure 13(a)); the r and associated P values were −0.19 and 0.02, respectively. In addition, the slope of a regression of ΔXCH₄ versus SSA was −40.27 ppb (Figure 13(b)); the r and associated P values were −0.26 and 9 × 10⁻⁴, respectively. The slopes of all the regressions of ΔXCO₂ and ΔXCH₄ versus AOT or SSA were much larger in magnitude at Lauder than at Tsukuba and Saga.

Some AOT outliers seemed to have strong effects on ΔXCO₂ and ΔXCH₄. Examining the relationship between AOT and either ΔXCO₂ or ΔXCH₄ with inclusion of these outliers may have been misleading. The unusual and unnaturally large magnitudes of the slopes at Lauder compared to Saga and Tsukuba may indicate that these slopes do not reflect typical conditions at Lauder. To get a better sense of the typical conditions at Lauder, we reanalyzed the dataset after excluding the data on 7 October 2015 (the maximum AOT discussed in Section 4.3.2.1). Table 2 shows the results of this reanalysis. The regression slope between ΔXCO₂ and AOT was −13.37 ppm (P = 0.059, r = −0.15). The one-sided t-test was therefore significant at P = 0.059/2 = 0.029. The slope of the regression between ΔXCH₄ and SSA was −43.87 ppb (P = 1.3 × 10⁻⁴, r = −0.3). These results provide convincing evidence that ΔXCO₂ at Lauder was negatively correlated with the AOT, whereas ΔXCH₄ was negatively correlated with the SSA.

Without the large AOT datum on 7 October 2015, there was no significant correlation between either ΔXCO₂ or ΔXCH₄ and AOT during the SON season (Table 2). The significance of these correlations was strongly dependent on the large AOT on 7 October 2015.

The seasonal means of both the ΔXCO₂ and ΔXCH₄ were negative during all seasons (Figure 14) though they were all less than one standard deviation in magnitude. Figure 15 clearly shows that the AOT was significantly smaller at Lauder than at Tsukuba and Saga. The large seasonal AOT means during SON (−0.07 ± 0.04) and DJF (0.05 ± 0.02) corresponded to large negative ΔXCO₂ means during SON (−1.23 ± 2.85) and DJF (−1.38 ± 1.55).
This pattern is consistent with the negative correlations between $\Delta X_{CO_2}$ and AOT at Lauder during these seasons. In addition, the largest seasonal mean SSA (.90 during JJA) corresponded to the largest negative $\Delta X_{CH_4}$ seasonal mean ($-10.08 \pm 10.09$) (Figure 14). The mean AE for all seasons was 1.45 at Lauder (Figure 15), hence small particles such as biomass burning aerosols were dominant at this site (Liley et al. 2001; Liley and Forgan 2009, 2002).

Based on the time series of $\Delta X_{CO_2}$ and AOT at Lauder (Figure 16(a)), we selected 7 May 2012 as an example of a small AOT before the Calbuco volcanic eruption (April 2015), and we selected 7 and 13 October 2015, which were the days when the values of $\Delta X_{CO_2}$ and $\Delta X_{CH_4}$ were most negative after the Calbuco volcanic eruption. These were very atypical days that facilitated our evaluation of the effects of aerosols and cirrus clouds on GOSAT $X_{CO_2}$ and $X_{CH_4}$ values at this site.

**Figure 12.** Relationship between $\Delta X_{CO_2}$ and either AOT, SSA at 500 nm, or AE at Lauder for all seasons (a, b, c) and separate seasons (a’, b’, c’).
4.3.1. 7 May 2012, before the Calbuco volcanic eruption

This day was chosen to study the relationship between the small AOT of 0.02 and the corresponding ΔXCO₂ and ΔXCH₄ values of 0.77 ppm and −6.05 ppb, respectively, during clear-sky conditions at Lauder. Because the AOT value was only 0.02, we expected the ΔXCO₂ to be close to zero based on the regression line in Figure 12(a). The corresponding SSA of 0.88 on this day would have led us to expect that ΔXCH₄ would be negative based on the regression line between ΔXCH₄ and SSA in Figure 13(b). Moreover, both the ΔXCO₂ and ΔXCH₄ values on this day were within one standard deviation of the corresponding mean values of clear-sky ΔXCO₂ and ΔXCH₄ values at Lauder (Section 4.3). These values of ΔXCO₂ and ΔXCH₄ indicated that the effects of aerosols on this day on GOSAT XCO₂ and XCH₄ were not significant. In addition, the vertical profiles of the backscattering and depolarization ratios observed near the time of the GOSAT overpass at 12:57:05 local time.

Figure 13. Relationship between ΔXCH₄ and either AOT, SSA at 500 nm, or AE at Lauder for all seasons (a, b, c) and separate seasons (a’, b’, c’).
(LT) on 7 May 2012 as well as the sky view (Figure 17) indicates that the sky was clear. The effects of the small AOT on ΔXCO₂ and ΔXCH₄ at Lauder were therefore not significant on this clear-sky day.

### 4.3.2. After the Calbuco volcanic eruption

Because Uchino et al. (2018) have indicated that the increase of SAOD following the April 2015 eruption of the Calbuco volcano in Chile, which was observed with lidar at Lauder, could result in negatively biased GOSAT XCO₂, we made scatter plots of ΔXCO₂ and ΔXCH₄ versus stratospheric aerosols (Figure 18) by using SAOD data derived from Uchino et al. (2018). In this figure, the effects of SAOD on ΔXCO₂ and ΔXCH₄ at Lauder from April 2012 to July 2016 are apparent. The increase in SAOD
following the April 2015 eruption of the Calbuco volcano in Chile, which was observed with lidar at Lauder, resulted in a negative bias of the GOSAT XCO2 and XCH4 values. Although the SAOD is small ($2 \times 10^{-3} - 2.6 \times 10^{-2}$), its influence on $\Delta$XCO2 and $\Delta$XCH4 is much larger than that of tropospheric aerosols. Small SAODs have been shown to have a large impact on XCO2 over the ocean retrieved from the Orbiting Carbon Observatory-2 during the Southern Hemisphere winter (O’Dell et al. 2018). The implication is that small amounts of stratospheric aerosols have caused negative biases of GOSAT XCO2 and XCH4 data at Lauder.

4.3.2.1. 7 October 2015. A maximum AOT value of 0.15 was observed during the afternoon on 7 October 2015. Most of New Zealand experienced a dry spring in 2015 because of a strong El Niño and that persisted in the Tropical Pacific (Kogan and Guo 2017). On 7 October 2015, because of a combination of high temperature, low humidity, and very strong winds throughout South Island, the NIWA Summer Series Week reported multiple fires around South Island (NIWA 2015). The rather large AEs of 1.19–1.55 at Lauder during this time were related to biomass burning due to the dominance of small particles (Liley and Forgan 2002). Besides local burning, past studies have shown that long-range transport of biomass burning products from Southeast Asia, Africa, and South America might cause mostly aerosol enhancements in the middle troposphere over Lauder (Jones et al. 2001; Liley et al. 2001). The aerosols observed in the middle troposphere at Lauder on this day might therefore have originated from biomass burning.

If we focus solely on the data during the SON season, during which the maximum AOT occurred on 7 October 2015, the correlations between $\Delta$XCO2 and $\Delta$XCH4 versus AOT were clearly negative. The slopes of the corresponding regression lines were $-43.47$ ppm ($P = 0.004$, Figure 12 (a')) and $-204.61$ ppb ($P = 0.003$, Figure 13 (a')).

The AOT value of 0.15 that was observed on 7 October 2015 corresponded to a negative $\Delta$XCO2 of $-5.97$ ppm and the most negative $\Delta$XCH4 at Lauder, $-36.43$ ppb. Both $\Delta$XCO2 and $\Delta$XCH4 were negatively correlated with AOT during the SON season, and the associated regression lines indicated that the values of $\Delta$XCO2 and $\Delta$XCH4 associated with the AOT of...
0.15 would be very negative. Furthermore, the vertical profile of R and Dep observed by lidar (Figure 19(a)) near the time of the GOSAT overpass revealed a tropospheric aerosol layer at an altitude <7 km (R = 1.1−1.5, Dep = 2.3−5.1) and a stratospheric aerosol layer at an altitude of 13−19 km (R = 1.4, Dep = 1.5−5.1) due to the influence of the Calbuco volcanic eruption (Uchino et al. 2018). Bègue et al. (2017) also confirmed a long-range transport of stratospheric aerosols in the Southern Hemisphere following the 2015 Calbuco eruption. These observations suggest that the Calbuco volcanic eruption was a source of aerosols in addition to biomass burning on this day. Both tropospheric and stratospheric aerosols caused effects on negative ΔXCO₂ and ΔXCH₄. The very negative values of ΔXCO₂ and ΔXCH₄ on this day are expected to be associated with a very positive AOT. However, the maximum AOT value on this day was still small. The stratospheric aerosols present at altitude of 13−19 km (Figure 19(a)) might have caused a negative bias of the GOSAT XCO₂ and XCH₄ values (e.g. Figure 18). Stratospheric aerosols may therefore have contributed to the negative bias of the GOSAT XCO₂ and XCH₄ in this case.

Figure 16. Time series of (a) ΔXCO₂ with AOT and (b) ΔXCH₄ with SSA at Lauder.
4.3.2.2. **13 October 2015.** This day was chosen because it was associated with the most negative \( \Delta XCO_2 \) value (−7.88 ppm) and a very negative \( \Delta XCH_4 \) value (−25.56 ppb), although the corresponding AOT value was only 0.06, and the SSA was 0.86. The regression line between \( \Delta XCO_2 \) and AOT (Figure 12(a)) at Lauder suggests that the \( \Delta XCO_2 \) would be only about −1.0 ppm at an AOT of 0.06. Similarly, the regression lines in Figures 13(a) and (b) between \( \Delta XCH_4 \) and the AOT and SSA at Lauder suggest that the \( \Delta XCH_4 \) should have been about −6 ppb at an AOT of 0.06 and the SSA of 0.86. The actual \( \Delta XCO_2 \) and \( \Delta XCH_4 \) were much more negative than the values suggested by the regression lines. Another factor may therefore have contributed to the very negative values of \( \Delta XCO_2 \) and \( \Delta XCH_4 \) on this day. In fact, the lidar profile at the nearest GOSAT overpass time (13:57:16 LT) (Figure 19(b)) reveals...
the presence of stratospheric aerosols at altitudes $>11$ km ($R = 1.4–1.6$, $Dep = 1.4–2.1$) and thin cirrus clouds at an altitude of 7 km ($R = 3$, $Dep = 15$). Sky-view images also showed cirrus clouds on 13 October 2015 (Figure 19(c)). We hypothesized that stratospheric aerosols accounted for the large negative values of $\Delta XCO_2$ and $\Delta XCH_4$ at Lauder on 7 October 2015 (Section 4.3.2.1). The large negative $\Delta XCO_2$ and $\Delta XCH_4$ on this day at Lauder might therefore have been caused by stratospheric aerosols, and the presence of cirrus clouds in this case might have further reduced the $\Delta XCO_2$ values.

Figure 19. Vertical profiles of the backscattering ratio ($R$) and total depolarization ratio ($Dep$) at 532 nm at Lauder after the Calbuco volcanic eruption as well as the sky view on 13 October 2015. The Blue lines ... in panels (a) and (b) are the first tropopause height determined by the United States National Oceanic and Atmospheric Association. LT = local time.
5. Conclusions

We used the AOT, AE, and SSA data recorded by the sky radiometer and vertical profiles of lidar data to analyze the influences of thin cirrus clouds and aerosols on the XCO$_2$ and XCH$_4$ retrieved from GOSAT at three TCCON sites: Tsukuba, Saga, and Lauder. The slope of the regression line relating $\Delta$XCO$_2$ to AOT at these sites was statistically significant.

At Tsukuba and Saga, there was a negative slope between $\Delta$XCO$_2$ and AOT at 500 nm. That is, the values of XCO$_2$ were smaller for GOSAT than for TCCON during the seasons of MAM and JJA, when the AOT was large. In particular, dust-like events were observed when the AOT values were large. Very large AOTs in the lower troposphere significant influence on negative $\Delta$XCO$_2$. In addition, we found no significant correlations between $\Delta$XCH$_4$ with AOT, SSA, and AE at these sites.

The AOT was generally small at Lauder. However, $\Delta$XCO$_2$ was negatively correlated with the AOT at 500 nm. During the SON season, the negative correlations between both $\Delta$XCO$_2$ and $\Delta$XCH$_4$ and AOT were highly significant. In addition, stratospheric aerosols caused large negative biases of $\Delta$XCO$_2$ and $\Delta$XCH$_4$, even though the SAOD was small.

The most negative $\Delta$XCO$_2$ values at the sites ranged from $-4.40$ ppm at Saga to $-7.88$ ppm at Lauder. Aerosol properties varied considerably between the three TCCON sites. The negative correlations between the SSA and $\Delta$XCO$_2$ at Tsukuba as well as $\Delta$XCH$_4$ at Lauder were statistically significant at $P < 0.05$. In addition, several lidar vertical profiles and sky-view images at the study sites confirmed the strong effects of cirrus clouds and dense aerosols on negative $\Delta$XCO$_2$ values. Supplementary materials provide details of cirrus clouds and aerosols presence in several cases.

A consistent result of the studies at Tsukuba, Saga, and Lauder was the statistically significant, negative correlation between $\Delta$XCO$_2$ and AOT at 500 nm. The regression lines at these sites between $\Delta$XCO$_2$ and AOT at 500 nm can be used to correct the bias in XCO$_2$ values estimated via GOSAT observations. Cirrus clouds and dense, boundary-layer aerosols caused very negative $\Delta$XCO$_2$ values at these sites, even when the AOT values are small, and stratospheric aerosols associated with small SAODs also caused large negative $\Delta$XCO$_2$ and $\Delta$XCH$_4$ over Lauder.

With the hypothesis that the $\Delta$XCO$_2$ and $\Delta$XCH$_4$ were caused partly by thin cirrus clouds and aerosols, this study has confirmed that the effects of cirrus clouds and aerosols on GOSAT XCO$_2$ and XCH$_4$ data were statistically significant. It is therefore crucial to further improve the effects of aerosols and cirrus clouds when retrieving GOSAT XCO$_2$ and XCH$_4$ data.

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Disclosure statement

No potential conflict of interest was reported by the author(s).
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