Atmos. Chem. Phys. Discuss., 11, 29883–29914, 2011 www.atmos-chem-phys-discuss.net/11/29883/2011/ doi:10.5194/acpd-11-29883-2011 © Author(s) 2011. CC Attribution 3.0 License.



This discussion paper is/has been under review for the journal Atmospheric Chemistry and Physics (ACP). Please refer to the corresponding final paper in ACP if available.

# Influence of aerosols and thin cirrus clouds on the GOSAT-observed CO<sub>2</sub>: a case study over Tsukuba

O. Uchino<sup>1</sup>, N. Kikuchi<sup>1</sup>, T. Sakai<sup>2</sup>, I. Morino<sup>1</sup>, Y. Yoshida<sup>1</sup>, T. Nagai<sup>2</sup>, A. Shimizu<sup>1</sup>, T. Shibata<sup>3</sup>, A. Yamazaki<sup>2</sup>, A. Uchiyama<sup>2</sup>, N. Kikuchi<sup>1</sup>, S. Oshchepkov<sup>1</sup>, A. Bril<sup>1</sup>, and T. Yokota<sup>1</sup>

<sup>1</sup>National Institute for Environmental Studies, 16-2 Onogawa, Tsukuba, Ibaraki 305-8506, Japan

<sup>2</sup>Meteorological Research Institute, 1-1 Nagamine, Tsukuba, Ibaraki 305-0052, Japan <sup>3</sup>Graduate School of Environmental studies, Nagoya University, D2-1(510) Furo-cho, Chikusa-ku, Nagoya 464-8601, Japan

Received: 12 October 2011 - Accepted: 27 October 2011 - Published: 8 November 2011

Correspondence to: O. Uchino (uchino.osamu@nies.go.jp)

Published by Copernicus Publications on behalf of the European Geosciences Union.

Discussion Pa	AC 11, 29883–2	<b>ACPD</b> 11, 29883–29914, 2011							
per   Discussion	Influence of aerosols and thin cirrus clouds O. Uchino et al.								
Pape	Title	Page							
Υ.	Abstract	Introduction							
	Conclusions	References							
iscuss	Tables	Figures							
sion P	I	۶I							
aper	•	•							
_	Back	Close							
Discu	Full Scre	een / Esc							
ussion	Printer-frier	ndly Version							
Pap	Interactive	Discussion							
er	6								

#### Abstract

Lidar observations of vertical profiles of aerosols and thin cirrus clouds were made at Tsukuba (36.1° N, 140.1° E), Japan, to investigate the influence of aerosols and thin cirrus clouds on the column-averaged dry-air mole fraction of carbon dioxide (XCO<sub>2</sub>) retrieved from observation data of the Thermal And Near-infrared Sensor for carbon Observation Fourier Transform Spectrometer, measured in the Short-Wavelength InfraRed band (TANSO-FTS SWIR), onboard the Greenhouse gases Observing SATellite (GOSAT). The lidar system measured the backscattering ratio, depolarization ratio, and/or the wavelength exponent of atmospheric particles. The lidar observations and ground-based high-resolution FTS measurements at the Tsukuba Total Carbon Column Observing Network (Tsukuba TCCON) site were recorded simultaneously during passages of GOSAT over Tsukuba.

GOSAT SWIR XCO<sub>2</sub> data (version 01.xx) released in August 2010 were compared with the lidar and Tsukuba TCCON data. High-altitude aerosols and thin cirrus clouds
 had a large impact on the GOSAT SWIR XCO<sub>2</sub> results. By taking into account the observed aerosol/cirrus vertical profiles and using a more adequate solar irradiance database in the GOSAT SWIR retrieval, the difference between the GOSAT SWIR XCO<sub>2</sub> data and the Tsukuba TCCON data was greatly reduced.

#### 1 Introduction

- <sup>20</sup> The concentration of carbon dioxide ( $CO_2$ ) increased from about 280 ppm in preindustrial times (before 1750) to 386.8 ppm in 2009, primarily because of emissions from combustion of fossil fuels and land-use changes (IPCC, 2007; WMO, 2010). Because  $CO_2$  absorbs infrared radiation from the earth's surface, increased  $CO_2$  concentrations lead to a rise in the earth's surface temperature. These changes in temperature
- <sup>25</sup> influence the biosphere, and the biosphere changes can have a feedback effect on CO<sub>2</sub> concentrations (Cox et al., 2000). To accurately predict future atmospheric CO<sub>2</sub> con-



centrations and their impacts on climate, it is necessary to clarify the global distribution and variations of  $CO_2$  sources and sinks.

Current CO<sub>2</sub> flux estimates obtained by inverse modeling rely mainly on groundbased observation data. Errors in the estimated regional fluxes in Siberia, Africa, Australia, and South America are particularly large because ground-based monitor-

- <sup>5</sup> Australia, and South America are particularly large because ground-based monitoring stations are sparse in those regions (WMO, 2010). Spectroscopic remote sensing from space is capable of acquiring data that cover the globe and hence is expected to reduce errors in the CO<sub>2</sub> flux estimation obtained by using inverse modeling (Rayner and O'Brien, 2001; Chevallier et al., 2009; Hungershoefer et al., 2010).
- To improve regional CO<sub>2</sub> flux estimates, the Greenhouse gases Observing SATellite (GOSAT) was launched on 23 January 2009 (Kuze et al., 2009) to observe global distributions of CO<sub>2</sub> and methane (CH<sub>4</sub>) concentrations from space. Column-averaged dry-air mole fractions of CO<sub>2</sub> and CH<sub>4</sub> (XCO<sub>2</sub> and XCH<sub>4</sub>) are retrieved from the Short-Wavelength InfraRed (SWIR) observation data of the Thermal And Near-infrared Sen-
- <sup>15</sup> sor for carbon Observation Fourier Transform Spectrometer (TANSO-FTS) onboard GOSAT (Yoshida et al., 2011). Morino et al. (2011) preliminarily validated the GOSAT SWIR XCO<sub>2</sub> and XCH<sub>4</sub> results by comparing them with reference data obtained by a ground-based high-resolution FTS of the Total Carbon Column Observing Network (TCCON; Wunch et al., 2011). They found that the GOSAT SWIR XCO<sub>2</sub> and XCH<sub>4</sub> (version 01.xx) values were systematically underestimated by 8.85±4.75 ppm
- $(2.29 \pm 1.23\%)$  and  $20.4 \pm 18.9$  ppb  $(1.15 \pm 1.06\%)$ , respectively. To improve the accuracy of the retrieval results, the causes of these biases (systematic errors) need to be investigated.

Houweling et al. (2005) demonstrated that systematic errors in CO<sub>2</sub> satellite remote <sup>25</sup> sensing data can be caused by aerosols by performing model calculations that showed large sensitivity of the CO<sub>2</sub> column to the vertical aerosol profile. To minimize the errors due to aerosols in SWIR CO<sub>2</sub> measurements from space, Butz et al. (2009) proposed that the amount, vertical distribution, and microphysical properties of aerosol particles should be parameterized and retrieved simultaneously with the total CO<sub>2</sub> column.



The GOSAT SWIR retrieval algorithm in Ver. 01.xx assumes that aerosols are uniformly distributed below 2 km of altitude and that no cirrus clouds are present. These assumptions are too simple; therefore, a forward spectrum error due to these assumptions may be one of the major sources of error in GOSAT SWIR  $XCO_2$  and  $XCH_4$ data. In this study, we investigated the impact of vertical aerosol profiles and thin cirrus clouds observed by lidar and sky radiometer on the GOSAT SWIR retrieval results,

- focusing on the GOSAT SWIR  $XCO_2$  results. First, we compared the GOSAT SWIR  $XCO_2$  data with reference data obtained by a ground-based high-resolution FTS at the National Institute for Environmental Studies (NIES) in Tsukuba, which is part of TC-
- CON (hereafter Tsukuba TCCON FTS). Next, we showed that GOSAT SWIR XCO<sub>2</sub> data are greatly influenced by high-altitude aerosols and thin cirrus clouds observed by lidar. Finally, we demonstrated that by taking into account the vertical aerosol profiles and thin cirrus clouds observed by lidar and sky radiometer, and by using Toon's solar irradiance database instead of Kurucz's database, the difference between the GOSAT SWIR XCO<sub>2</sub> data and the Tsukuba TCCON data becomes much less.

### 2 Comparison of GOSAT SWIR and Tsukuba TCCON XCO<sub>2</sub> data

### 2.1 GOSAT SWIR XCO<sub>2</sub>

We used GOSAT SWIR XCO<sub>2</sub> Ver. 01.xx products. The Ver. 01.xx retrieval algorithm uses TANSO-FTS Band 1 (12 900–13 200 cm<sup>-1</sup>) and Band 2 (5800–6400 cm<sup>-1</sup>) to simultaneously derive XCO<sub>2</sub> and XCH<sub>4</sub>. To suppress bias error, auxiliary parameters such as surface pressure and aerosol optical thickness (AOT) are retrieved together with XCO<sub>2</sub> and XCH<sub>4</sub>. The GOSAT SWIR Ver. 01.xx algorithm focuses on those data obtained under cloud-free conditions, and cloud-contaminated data detected by the TANSO Cloud and Aerosol Imager (TANSO-CAI) onboard GOSAT and TANSO-FTS
Band 3 (4800–5200 cm<sup>-1</sup>) data are excluded from the retrieval analysis. After the retrieval calculations, the quality of the retrieved state is checked from the viewpoints of



the convergence (number of iterations, chi-square, and mean square of the residual spectra for each retrieval sub-band), available information (degrees of freedom for signals and the signal-to-noise ratio, SNR), and the range of the retrieved AOT values. Details are described by Yoshida et al. (2011).

#### 5 2.2 Tsukuba TCCON FTS

Solar absorption spectra are measured with a Bruker IFS 120 HR FTS at NIES ( $36.0513^{\circ}$  N,  $140.1215^{\circ}$  E, 31 m a.s.l.) in Tsukuba, Japan. Direct solar light is introduced into the FTS with a solar tracker and five gold-coated flat mirrors. The solar tracker is mounted inside a dome on the roof of the building where the FTS is housed. Measurements with the high-resolution FTS are performed according to the TCCON data protocol. A CaF<sub>2</sub> beam splitter and an InGaAs detector are used for the 5500–10500 cm<sup>-1</sup> spectral region. A spectral resolution of  $0.02 \text{ cm}^{-1}$  (defined as 0.9/maximum optical path difference), an aperture size of 0.5 mm, and a scanner velocity of 10 kHz are used as standard parameters for the TCCON measurements. The pres-

sure in the FTS is kept at ~0.03 Torr by an oil-free scroll vacuum pump. The forward and backward scanned interferograms are separately integrated over a period of about 70 s. A weather station also observes meteorological data, recording surface pressure, surface temperature, relative humidity, wind direction and speed, rainfall, and solar radiation intensity at the same site. Table 1 lists the characteristics of the Tsukuba TCCON

FTS. Each measured spectrum was obtained by Fourier transform of the interferogram. Spectra measured with the Tsukuba TCCON FTS were analyzed by using the GFIT nonlinear least-squares spectral fitting algorithm, which is used for retrievals across all TCCON stations (Wunch et al., 2011).

TCCON XCO<sub>2</sub> is defined as the ratio of the CO<sub>2</sub> column amount to the dry-air column amount. To calculate the dry-air column amount, the GFIT algorithm uses the measured O<sub>2</sub> column amount divided by the known dry-air mole fraction of O<sub>2</sub> (0.2095). The O<sub>2</sub> and CO<sub>2</sub> columns are measured simultaneously using the 7751–8000 cm<sup>-1</sup>



(1250–1290 nm) spectral band. XCO<sub>2</sub> is then obtained as follows:

 $XCO_2 = 0.2095 \times (CO_2 \text{ column}/O_2 \text{ column})$ 

#### (1)

By using the  $CO_2$  to  $O_2$  ratio, systematic and correlated errors present in both retrieved columns are minimized.

The precision of the FTS measurement of XCO<sub>2</sub> is better than 0.2% under clear 5 sky conditions (Washenfelder et al., 2006; Ohyama et al., 2009; Messerschmidt et al., 2010; Wunch et al., 2011). All TCCON XCO<sub>2</sub> data are corrected for airmass-dependent artifacts (Wunch et al., 2011). Aircraft profiles obtained over many of these sites are used to empirically scale the TCCON data according to the WMO standard reference scale. The scaling factor of TCCON XCO<sub>2</sub> is 1.011. The uncertainty of TCCON XCO<sub>2</sub> 10 associated with the FTS measurement after scaling by 1.011 has been estimated

to be 0.8 ppm (~0.2%) by comparing TCCON retrievals with many different aircraftmeasured profiles (Wunch et al., 2010).

In 2010, Tsukuba TCCON FTS data were calibrated against data from three aircraft flights and tower measurements of CO<sub>2</sub> concentrations, and an additional bias of 15  $-1.32 \pm 0.46$  ppm (1 $\sigma$ ) was demonstrated after airmass-dependent artifact correction and 1.011 scaling (Tanaka et al., 2011). Here we use these bias-corrected Tsukuba TCCON XCO<sub>2</sub> data.

#### 2.3 Comparison of GOSAT SWIR XCO<sub>2</sub> and Tsukuba TCCON data

- We compared GOSAT SWIR XCO<sub>2</sub> data obtained over Tsukuba on 9 days between 20 September 2009 and March 2010 with Tsukuba TCCON data, using the mean values measured at Tsukuba within 30 min of the GOSAT overpass time (around 12:54 LT) (Fig. 1; Table 2). About half of GOSAT SWIR XCO<sub>2</sub> data were rejected because they did not meet the quality control criteria. The GOSAT SWIR XCO<sub>2</sub> data obtained on 14
- February 2010 did not converge within the pre-determined maximum iteration number 25 of 20, so we used the XCO<sub>2</sub> value obtained at the 20th iteration. The average difference between GOSAT SWIR XCO<sub>2</sub> and Tsukuba TCCON XCO<sub>2</sub> was 3.42%, based on all



data summarized in Table 2. On the other hand, the average difference when data not meeting quality control criteria were excluded was 2.34 %, which is nearly equal to the value of 2.29 % obtained by an extended comparison over 9 TCCON sites around the world (Morino et al., 2011). Thus, the quality control procedure adequately rejects
 outlying data points. Next we investigated these results by comparing them with lidar data obtained simultaneously with the GOSAT and Tsukuba TCCON FTS data.

## 3 Lidar observations of aerosols and thin cirrus clouds over Tsukuba and the influence of high-altitude particles on GOSAT SWIR XCO<sub>2</sub>

A compact lidar, based on a Nd:YAG laser, was developed to observe vertical distributions of thin cirrus clouds and aerosols and evaluate the influence of these particles on GOSAT SWIR XCO<sub>2</sub> data. Two laser wavelengths of 1064 nm ( $\lambda_1$ ) and 532 nm ( $\lambda_2$ ) are transmitted into the atmosphere through a beam expander. The backscattered light from the upper atmosphere is collected by a telescope and then divided into  $\lambda_1$  and  $\lambda_2$  by a dichroic mirror, and  $\lambda_2$  is further divided into a parallel (*P*) and a perpendicular component (*S*) by a polarizer.  $\lambda_1$  is detected by an avalanche photodiode (APD) and  $\lambda_2$  by photomultiplier tubes (PMTs). The output signals are processed by transient recorders with an analog/digital converter (A/D) and a photon counter (PC). Table 3 summarizes the characteristics of the lidar (Uchino et al., 2010).

The backscattering ratio R is defined as

 $_{20}$  R = (BR + BA)/BR

where BR and BA are the Rayleigh and Mie backscattering coefficients, respectively. Backscattering ratio profiles are derived by the inversion method (Fernald, 1984). We assumed the lidar ratio (extinction to backscatter ratio) to be 50 sr for aerosols and 20 sr for cirrus clouds (Sakai et al., 2003). To calculate BR, we used the atmospheric molec-

<sup>25</sup> ular density profiles obtained by operational radiosondes at the Tsukuba Aerological Observatory of the Japan Meteorological Agency (JMA) (36.06° N, 140.13° E).



(2)

The total depolarization ratio (Dep) is defined as

 $Dep = S/(P + S) \times 100(\%)$ 

where P and S are the parallel and perpendicular components of the backscattered signals. Dep indicates whether the particles are spherical or non-spherical, with large values indicating non-spherical particles. The wavelength exponent, Alp, which shows whether the Mie particles are small or large, is defined by

 $BA(\lambda) \propto \lambda^{-Alp}$ 

Larger values of Alp indicate smaller particles.

Figure 2 shows vertical profiles of *R*, Dep, and Alp observed on 14, 20, and 23
February 2010. The lidar observations were made during a period of about 10 min as GOSAT passed over Tsukuba. The vertical resolution used for the analysis was 150 m. On 14 February 2010, there were thin cirrus clouds at altitudes of 6.1–10.9 km and aerosols below 3 km. The partial optical thickness at altitudes of 0.4–30 km, Tau (0.4–30 km), was 0.24 at 532 nm (Fig. 2). The optical thickness from the surface to the top of the atmosphere could not be obtained below 0.4 km because the beam overlap

- between the lidar transmitter and receiver was not perfect. Lidar measurements of stratospheric aerosols above 15 km were observed at night (Uchino et al., 2010). In contrast to 14 February, 20 February 2010 was a comparatively clear day with aerosols in the boundary layer, and Tau (0.4–30 km) was estimated to be 0.1. On 23 February,
- the high-altitude aerosols observed at altitudes of 1–5 km were likely dust particles, because Dep was large, indicating non-spherical particles. Tau (0.4–30 km) was 0.16. The difference between GOSAT SWIR XCO<sub>2</sub> and Tsukuba TCCON XCO<sub>2</sub> values was the largest (19.01 ppm or 4.86 %) on 14 February 2010 (Table 2). The difference was small (4.86 ppm or 1.24 %) on 20 February, and it was somewhat large (12.00 ppm
- or 3.07 %) on 23 February. The cirrus clouds on 14 February 2010 might have influenced the GOSAT retrieval. There were also thin cirrus clouds around 10.9–11.2 km altitude on 11 September 2009, when the difference was also relatively large (2.99 %).



(3)

(4)

Our results indicate that the retrieval of GOSAT SWIR  $XCO_2$  data is greatly influenced by high-altitude aerosols and thin cirrus clouds.

The current version of the retrieval algorithm (Ver. 01.xx) assumes that atmospheric aerosols are uniformly distributed from the ground surface to 2 km altitude. Next we show that GOSAT SWIR XCO<sub>2</sub> data were improved when the vertical distribution of the optical thicknesses of aerosols and the thin cirrus clouds observed by lidar and sky radiometer were taken into account.

#### 4 Improvement of GOSAT SWIR XCO<sub>2</sub> retrieval

#### 4.1 Vertical profiles of aerosol species and cirrus clouds

- <sup>10</sup> Vertical profiles and optical properties of aerosols and cirrus clouds used in the retrieval analysis were prepared based on the lidar and sky radiometer measurements. The sky radiometer can measure aerosol optical thickness and single scattering albedo at four wavelengths (400, 500, 675, and 870 nm), and the Angstrom exponent can be estimated from the optical thickness at these four wavelengths (Shiobara et al., 1991; <sup>15</sup> Kobayashi et al., 2006). The single scattering albedo is defined as the ratio of the scattering coefficient to the scattering coefficient plus the absorption coefficient of the particles. If the single scattering albedo is 1.0, then the particles are non-absorbing. A large value of the Angstrom exponent indicates small particles. Table 4 summarizes the aerosol optical thickness at 500 nm ( $\tau_{500}$ ), the single scattering albedo at 500 nm ( $\omega_{500}$ ), and the Angstrom exponent ( $\alpha$ ) at the GOSAT overpass times; the optical thick-
- ness at 532 nm ( $\tau_{532}$ ), calculated from the lidar measurement by extrapolating the value of BA at 0.4 km down to the ground surface, is also shown. The optical thickness of cirrus clouds is not included in  $\tau_{532}$ , and it is approximately the same as  $\tau_{500}$ . The Angstrom exponent of aerosols over Tsukuba was large except on 14 February, 23
- <sup>25</sup> February, and 22 March 2010 (Table 4). In addition, the values of  $\omega_{500}$  were close to unity, indicating that the aerosol particles were small and non-absorbing (Table 4). The



relatively small value of  $\alpha$  on 14 February 2010 might reflect contamination by cirrus clouds, because the Dep value of the lidar measurement does not indicate the presence of large, non-spherical aerosol particles. We therefore assumed that, except on 23 February and 22 March 2010, the aerosols over Tsukuba were sulfate because the particles were small and non-absorbing.

On 23 February and 22 March, the vertical Dep profiles indicate the presence of large, non-spherical dust-like particles at 2–4 km altitude. We assumed small, non-absorbing aerosols to be sulfate and large particles to be dust. We calculated the optical properties of sulfate aerosols following Takemura et al. (2002), but using a reduced width in the size distribution as suggested by Schutgens et al. (2010). For the dust aerosol model, we used the mineral-transported component of the model of Hess et al. (1998). Using these aerosol models, we determined the dry-mass fraction of sulfate such that the Angstrom exponent of the sulfate–dust mixture agreed with that derived from the sky radiometer observations.

The vertical profiles of the extinction coefficient and the optical thicknesses of sulfate particles and cirrus clouds on 14 February 2010 are shown in Fig. 3, and those of sulfate and dust particles on 23 February 2010 are shown in Fig. 4. Similarly, we obtained vertical profiles of aerosols and cirrus clouds for the other days by using lidar and sky radiometer data observed at Tsukuba.

## 20 4.2 Revised XCO<sub>2</sub> retrieved using the vertical profiles of particles observed by lidar and sky radiometer

We retrieved XCO<sub>2</sub> (revised XCO<sub>2</sub>) by taking account of the vertical profiles of the two types of aerosols and cirrus clouds determined from lidar and sky radiometer data (Table 5, Case 1). In Case 1, we modified the operational Ver. 01.xx algorithm as follows. The uniform aerosol distribution up to 2 km altitude was replaced by the vertical profile derived from lidar measurements, as shown in Figs. 3 and 4. The aerosol optical thickness was then retrieved by scaling the vertical profile. Then we used Mie theory to derive the aerosol optical properties by assuming a mixture of sulfate and dust; for the



operational algorithm we adopted aerosol optical properties estimated by the aerosol transport model SPRINTARS (Ver. 3.54) (Takemura et al., 2000). In addition, cirrus clouds were included in the forward model on 11 September 2009 and 14 February 2010, when lidar measurements showed that they were present. The optical thickness

of the cirrus clouds was retrieved by scaling the vertical profile observed by lidar. To estimate the optical properties of ice crystals in the cirrus clouds, we adopted the Cirrus 3 model of Hess et al. (1998).

We plotted these retrieved values as the revised  $XCO_2$  against the Tsukuba TCCON values (Fig. 5). The difference between the revised  $XCO_2$  and the Tsukuba TCCON  $XCO_2$  data was 7.40 ppm or 1.90 %; thus, these revised  $XCO_2$  data are closer to the

- 10 XCO<sub>2</sub> data was 7.40 ppm or 1.90 %; thus, these revised XCO<sub>2</sub> data are closer to the TCCON data than the SWIR Ver. 01.xx results shown in Fig. 1. In particular, the data for 11 September 2009 and 14 February 2010, when aerosol optical thickness was large and cirrus clouds were present, and on 23 February and 22 March 2010, when aerosol optical thickness was large, were greatly improved. Nevertheless, although the negative bias in XCO<sub>2</sub> was reduced to one half that obtained with the operational
- the negative bias in XCO<sub>2</sub> was reduced to one half that obtained with the operational algorithm, it was not eliminated.

#### 4.3 Solar irradiance database

Although a high-resolution solar irradiance database is needed to simulate a TANSO-FTS measured spectrum, few such solar irradiance databases are available. The 20 GOSAT SWIR retrieval analysis used the high-resolution solar irradiance database (0.004 to 0.01 cm<sup>-1</sup>) of R. Kurucz (http://kurucz.harvard.edu/sun/irradiance2008/). This database was created from spectra measured with a ground-based highresolution FTS at Kitt Peak National Observatory (Arizona, USA) by removing the absorption structure due to the earth's atmosphere. However, we noticed a CO<sub>2</sub> absorp-

tion structure in the spectral residual between the measured spectrum and the spectrum simulated by the forward spectral model, whereas when we used a solar spectrum database provided by G. C. Toon (Toon et al., 1999, personal communication), we confirmed no  $CO_2$  absorption structure in the spectral residuals. We thus decided to use



Toon's solar irradiance database. We also applied the low-frequency baseline correction in the current Ver. 01.xx retrieval to Toon's solar irradiance database.

#### 4.4 New XCO<sub>2</sub> retrieved using Toon's solar irradiance database

We retrieved  $XCO_2$  (new  $XCO_2$ ) data by using Toon's solar irradiance data instead of Kurucz's data and by taking into account the vertical profiles of the two types of aerosols and cirrus clouds determined by lidar and sky radiometer (Table 5, Case 2), and plotted these new XCO<sub>2</sub> values against the Tsukuba TCCON data (Fig. 6). The difference between the new XCO<sub>2</sub> and Tsukuba TCCON XCO<sub>2</sub> data was 2.43 ppm or 0.62 %. Thus, the new XCO<sub>2</sub> data (Fig. 6) were much closer to the Tsukuba TCCON XCO<sub>2</sub> data than the GOSAT SWIR (Ver. 01.xx) data (Fig. 1). We compared the retrieved 10 optical thickness at 532 nm with that of the a priori lidar data (Fig. 7) and found that, in general, the retrieved aerosol optical thickness was nearly equal to the a priori value. We also compared the a priori surface pressure, obtained by interpolating in both time and space the Objective Analysis Data of JMA to obtain values for Tsukuba, with the retrieved values (Fig. 8). The difference between the a priori and retrieved surface 15 pressure was small except on 11 October 2009 and 6 January 2010. Therefore, it is reasonable to infer that the new retrieval results (new XCO<sub>2</sub> data) are reliable.

#### 4.5 Discussion

The GOSAT SWIR XCO<sub>2</sub> was underestimated by  $2.29 \pm 1.23\%$  compared with the reference data obtained at 9 TCCON sites (Morino et al., 2011). In this study, we demonstrated that the negative bias of 3.42% in the GOSAT SWIR XCO<sub>2</sub> data at the Tsukuba TCCON site could be reduced to 1.90% by taking into account the vertical profiles of aerosols and cirrus clouds observed by lidar and sky radiometer. The negative bias in XCO<sub>2</sub> was then further reduced to 0.62% by using Toon's solar irradiance data instead of Kurucz's data.



These results show that vertical profiles of aerosol species and cirrus clouds must be considered in the retrieval algorithm in order to improve the data quality of the global GOSAT SWIR XCO<sub>2</sub>when lidar observations are not available. One of the simplest ways to improve the treatment of aerosols would be to incorporate vertical profiles of aerosols obtained from SPRINTARS in the forward model. Aerosol vertical profiles simulated by SPRINTARS, however, are not sufficient, as shown by comparing the SPRINTARS aerosol profile with that observed by lidar (Fig. 9). Therefore, as the first step, we simultaneously retrieved XCO<sub>2</sub> (simulated XCO<sub>2</sub>; Table 5, Case 3) and the vertical profile of aerosol optical thickness based on the a priori aerosol optical thickness profile calculated by SPRINTARS. In Case 3, the optical thickness and cloud-top pressure of the cirrus clouds were also retrieved simultaneously. The cloud-bottom pressure was modeled as a linear function of the cloud-top pressure, as suggested by N. Eguchi (Eguchi et al., 2007, personal communication), and the cirrus clouds

were assumed to be distributed uniformly in the vertical direction. In addition, Band 3 spectra (4790–4910 cm<sup>-1</sup>) were also utilized in Case 3 for higher retrieval accuracy of the vertical aerosol profiles.

The simulated and Tsukuba TCCON  $XCO_2$  values are shown in Fig. 10. Although information on the vertical profiles of aerosols and cirrus clouds observed by lidar was not used in retrieving the simulated  $XCO_2$ , the simulated values were considerably closer to the Tsukuba TCCON  $XCO_2$  values than current retrievals by GOSAT SWIR  $XCO_2$  (Fig. 1). We also found that use of Band 3 increased  $XCO_2$  by about 2 ppm, but we have not yet identified the origin of this difference. Aerosol optical properties derived

20

from SPRINTARS were used in both Case 3 and the current operational algorithm. The simulated  $XCO_2$  results shown in Fig. 10 are satisfactory, but the aerosol optical

thickness thus obtained could be a source of bias in XCO<sub>2</sub> for retrievals at sites other than the Tsukuba TCCON site. Therefore, it would be better to use a new SPRINTARS model in which AERONET observations are assimilated (Schutgens et al., 2010). Furthermore, SPRINTARS is being further improved by assimilation of lidar network and CALIOP data (Shimizu et al., 2004; Winker et al., 2007; Sekiyama et al., 2010).



#### 5 Concluding remarks

Version 01.xx GOSAT SWIR XCO<sub>2</sub> data, released in August 2010, were compared with Tsukuba TCCON data. Comparison of lidar and sky radiometer observations with the GOSAT SWIR XCO<sub>2</sub> data clearly showed that high-altitude aerosols and thin cir<sup>5</sup> rus clouds had a large impact on GOSAT SWIR XCO<sub>2</sub>. The current retrieval algorithm (Ver. 01.xx) for XCO<sub>2</sub> and XCH<sub>4</sub> from the GOSAT TANSO-FTS SWIR observation data assumes that atmospheric aerosols are uniformly distributed from the ground surface to 2 km altitude. By taking into account the actual vertical distributions of aerosols determined by lidar and sky radiometer over Tsukuba, and by using Toon's solar irra<sup>10</sup> diance database instead of Kurucz's database, the difference between GOSAT SWIR XCO<sub>2</sub> data and the Tsukuba TCCON XCO<sub>2</sub> found in the Ver. 01.xx results was reduced. In the near future, we plan to incorporate the vertical distributions of aerosols at altitudes above 2 km in the GOSAT SWIR retrieval algorithm.

Acknowledgements. We are grateful to G. C. Toon for making his solar irradiance data available to us. We also acknowledge N. Eguchi for valuable information on cirrus clouds. This research was supported in part by the Environment Research and Technology Development Fund (A-1102) of the Ministry of the Environment, Japan.

#### References

20

- Butz, A., Hasekamp, O. P., Frankenberg, C., and Aben, I.: Retrievals of atmospheric CO<sub>2</sub> from simulated space-borne measurements of backscattered near-infrared sunlight: accounting for aerosol effects, Appl. Opt., 48, 3322–3336, 2009.
- Chevallier, F., Maksyutov, S., Bousquet, P., Bréon, F.-M., Saito, R., Yoshida, Y., and Yokota, T.: On the accuracy of the CO<sub>2</sub> surface fluxes to be estimated from the GOSAT observations, Geophys. Res. Lett., 36, L19807, doi:10.1029/2009GL040108, 2009.
- <sup>25</sup> Cox, P. M., Betts, R. A., Jones, C. D., Spall, S. A., and Totterdell, I. J.: Acceleration of global warming due to carbon-cycle feedbacks in a coupled climate model, Nature, 408, 184–187, 2000.



- Eguchi, N., Yokota, T., and Inoue, G.: Characteristics of cirrus clouds from ICESat/GLAS observations, Geophys. Res. Lett., 34, L09810, doi:10.1029/2007GL029529, 2007.
- Fernald, F. G.: Analysis of atmospheric lidar observations: some comments, Appl. Opt., 23, 652–653, 1984.
- <sup>5</sup> Hess, M., Koepke, P., and Schult, I.: Optical properties of aerosols and clouds: the software package OPAC, B. Am. Meteorol. Soc., 79, 831–844, 1998.
  - Houweling, S., Hartmann, W., Aben, I., Schrijver, H., Skidmore, J., Roelofs, G.-J., and Breon, F. M.: Evidence of systematic errors in SCIAMACHY-observed CO<sub>2</sub> due to aerosols, Atmos.
     Chem. Phys., 5, 3003–3013, doi:10.5194/acp-5-3003-2005, 2005.
- <sup>10</sup> Hungershoefer, K., Breon, F.-M., Peylin, P., Chevallier, F., Rayner, P., Klonecki, A., Houweling, S., and Marshall, J.: Evaluation of various observing systems for the global monitoring of CO<sub>2</sub> surface fluxes, Atmos. Chem. Phys., 10, 10503–10520, doi:10.5194/acp-10-10503-2010, 2010.

Intergovernmental Panel on Climate Change (IPCC), Climate change 2007: The Physical Sci-

ence Basis: Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, edited by: Solomon, S., Qin, S., Manning, M., Chen, Z., Marquis, M., Averyt, K. B., Tignor, M., and Miller, H. L., Cambridge University Press, Cambridge, UK and New York, USA, 996 pp., 2007.

Kobayashi, E., Uchiyama, A., Yamazaki, A., and Matsuse, K.: Application of the maximum likelihood method to the inversion algorithm for analyzing aerosol optical properties from sun and sky radiance measurements, J. Meteor. Soc. Japan, 84, 1047–1062, 2006.

20

- Kuze, A., Suto, H., Nakajima, M., and Hamazaki, T.: Thermal and near infrared sensor for carbon observation Fourier-transform spectrometer on the Greenhouse Gases Observing Satellite for greenhouse gases monitoring, Appl. Opt., 48, 6716–6733, 2009.
- Messerschmidt, J., Macatangay, R., Notholt, J., Petri, C., Warneke, T., and Weinzierl, C.: Side by side measurements of CO<sub>2</sub> by ground-based Fourier transform spectrometry (FTS), Tellus B, 62, 749–758, 2010.
  - Morino, I., Uchino, O., Inoue, M., Yoshida, Y., Yokota, T., Wennberg, P. O., Toon, G. C., Wunch, D., Roehl, C. M., Notholt, J., Warneke, T., Messerschmidt, J., Griffith, D. W. T.,
- Deutscher, N. M., Sherlock, V., Connor, B., Robinson, J., Sussmann, R., and Rettinger, M.: Preliminary validation of column-averaged volume mixing ratios of carbon dioxide and methane retrieved from GOSAT short-wavelength infrared spectra, Atmos. Meas. Tech., 4, 1061–1076, doi:10.5194/amt-4-1061-2011, 2011.



- Ohyama, H., Morino, I., Nagahama, T., Machida, T., Suto, H., Oguma, H., Sawa, Y., Matsueda, H., Sugimoto, N., Nakane, H., and Nakagawa, K.: Column-averaged volume mixing ratio of CO<sub>2</sub> measured with ground-based Fourier transform spectrometer at Tsukuba, J. Geophys. Res., 114, D18303, doi:10.1029/2008JD011465, 2009.
- <sup>5</sup> Rayner, P. J. and O'Brien, D. M.: The utility of remotely sensed CO<sub>2</sub> concentration data in surface source inversions, Geophys. Res. Lett., 28, 175–178, 2001.
  - Sakai, T., Nagai, T., Nakazato, M., Mano, Y., and Matsumura, T.: Ice clouds and Asian dust studied with lidar measurements of particle extinction-to-backscatter ratio, particle depolarization, and water-vapor mixing ratio over Tsukuba, Appl. Opt., 42, 7103–7116, 2003.
- Sekiyama, T. T., Tanaka, T. Y., Shimizu, A., and Miyoshi, T.: Data assimilation of CALIPSO aerosol observations, Atmos. Chem. Phys., 10, 39–49, doi:10.5194/acp-10-39-2010, 2010. Schutgens, N. A. J., Miyoshi, T., Takemura, T., and Nakajima, T.: Applying an ensemble Kalman filter to the assimilation of AERONET observations in a global aerosol transport model, Atmos. Chem. Phys., 10, 2561–2576, doi:10.5194/acp-10-2561-2010, 2010.
- <sup>15</sup> Shimizu, A., Sugimoto, N., Matsui, I., Arao, K., Uno, I., Murayama, T., Kagawa, N., Aoki, A., Uchiyama, A., and Yamazaki, A.: Continuous observations of Asian dust and other aerosols by polarization lidars in China and Japan during ACE-Asia, J. Geophys. Res., 109, D19S17, doi:10.1029/200JD003253, 2004.

Shiobara, M., Hayasaka, T., Nakajima, T., and Tanaka, M.: Aerosol monitoring using a scanning

spectral radiometer in Sendai, Japan, J. Meteor. Soc. Japan, 69, 57–70, 1991. Takemura, T., Okamoto, H., Maruyama, Y., Numaguti, A., Higurashi, A., and Nakajima, T.: Global three-dimensional simulation of aerosol optical thickness distribution of various origins, J. Geophys. Res., 105, 17853–17873, 2000.

Takemura, T., Nakajima, T., Dubovik, O. D., Holben, B. N., Kinne, S.: Single-scattering albedo

- and Radiative forcing of various aerosol species with a global three-dimensional model, J. Climate, 15, 333–352, 2002
  - Tanaka, T., Miyamoto, Y., Morino, I., Machida, T., Nagahama, T., Sawa, Y., Matsueda, H., Wunch, D., Kawakami, S., and Uchino, O.: Aircraft measurements of carbon dioxide and methane for the calibration of ground-based high-resolution Fourier Transform Spectrome-
- ters and a comparison to GOSAT data measured over Tsukuba and Moshiri, in preparation, 2011.
  - Toon, G. C., Blavier, J.-F., Sen, B., Salawitch, R. J., Osterman, G. B., Notholt, J., Rex, M., McElroy, C. T., and Russell III, J. M.: Ground-based observations of Arctic O<sub>3</sub> loss during



spring and summer 1997, J. Geophys. Res., 104(D21), 26497-26510, 1999.

- Uchino, O., Sakai, T., Nagai, T., Sakashita, T., Suzuki, K., Shibata, T., Morino, I., and Yokota, T.: Lidar observation of stratospheric aerosols increased from the 2009 Mount Sarychev volcanic eruption, J. Remote Sens. Soc. Japan, 30, 149–155, 2010 (in Japanese).
- <sup>5</sup> Washenfelder, R. A., Toon, G. C., Blavier, J.-F., Yang, Z., Allen, N. T., Wennberg, P. O., Vay, S. A., Matross, D. M., and Daube, B. C.: Carbon dioxide column abundances at the Wisconsin Tall Tower site, J. Geophys. Res., 111, D22305, doi:10.1029/2006JD007154, 2006.
  - Winker, D. M., Hunt, W. H., and McGill, M. J.: Initial performance assessment of CALIOP, Geophys. Res. Lett., 34, L19803, doi:10.1029/2007GL030135, 2007.
- <sup>10</sup> WMO: The state of greenhouse gases in the atmosphere based on global observations through 2009, WMO Greenhouse Gas Bulletin, No. 6, 2010.
  - Wunch, D., Toon, G. C., Wennberg, P. O., Wofsy, S. C., Stephens, B. B., Fischer, M. L., Uchino, O., Abshire, J. B., Bernath, P., Biraud, S. C., Blavier, J.-F. L., Boone, C., Bowman, K. P., Browell, E. V., Campos, T., Connor, B. J., Daube, B. C., Deutscher, N. M., Diao, M., Elkins, J. W., Gerbig, C., Gottlieb, E., Griffith, D. W. T., Hurst, D. F., Jiménez, R., Keppel-
- Elkins, J. W., Gerbig, C., Gottlieb, E., Griffith, D. W. I., Hurst, D. F., Jimenez, R., Keppel-Aleks, G., Kort, E. A., Macatangay, R., Machida, T., Matsueda, H., Moore, F., Morino, I., Park, S., Robinson, J., Roehl, C. M., Sawa, Y., Sherlock, V., Sweeney, C., Tanaka, T., and Zondlo, M. A.: Calibration of the Total Carbon Column Observing Network using aircraft profile data, Atmos. Meas. Tech., 3, 1351–1362, doi:10.5194/amt-3-1351-2010, 2010.
- <sup>20</sup> Wunch, D., Toon, G. C., Blavier, J.-F. L., Washenfelder, R. A., Notholt, J., Connor, B. J., Griffith, D. W. T., Sherlock, V., and Wennberg, P. O.: The Total Carbon Column Observing Network (TCCON), Phil. Trans. R. Soc. A., 369, 2087–2112, doi:10.1098/rsta.2010.0240, 2011.
   Yoshida, Y., Ota, Y., Eguchi, N., Kikuchi, N., Nobuta, K., Tran, H., Morino, I., and Yokota, T.: Retrieval algorithm for CO<sub>2</sub> and CH<sub>4</sub> column abundances from short-wavelength infrared spectral observations by the Greenhouse gases observing satellite, Atmos. Meas. Tech., 4, 717–734, doi:10.5194/amt-4-717-2011, 2011.

	AC	ACPD								
	11, 29883–29914, 2011									
-	Influence of aerosols and thin cirrus clouds									
	O. Uchino et al.									
	Title	Page								
	Abstract	Abstract Introduction								
,	Conclusions	References								
	Tables	Figures								
J	I	۶I								
	•	•								
-	Back	Close								
2	Full Scre	en / Esc								
-	Printer-frien	dly Version								
7	Interactive	Discussion								
	(C)	•								

BY

iscussion Pape

Jiscussion Pape

Discussion Pape

Jiscussion Papel

 Table 1. Characteristics of the Tsukuba TCCON FTS.

Instrument type	Bruker IFS 120 HR
Beam splitter	CaF <sub>2</sub>
Aperture size	0.5 mm
Detector	$\ln GaAs (5000-10500 \mathrm{cm}^{-1})$
	Si diode $(9200-14000 \mathrm{cm}^{-1})$
Spectral resolution	$0.02  \mathrm{cm}^{-1}$
Single-scan observation time	70 s

Discussion Pa	<b>ACPD</b> 11, 29883–29914, 2011								
per   Discussion	Influence of aerosols and thin cirrus clouds O. Uchino et al.								
Pape	Title	Page							
er	Abstract	Introduction							
	Conclusions	References							
iscussi	Tables	Figures							
on P	14	►I.							
aper	•	Þ							
_	Back	Close							
Discus	Full Scre	en / Esc							
sion	Printer-frier	dly Version							
Paper	Interactive	Discussion							
1		()							

Table 2. Comparison of GOSAT SWIR XCO <sub>2</sub> (A) with Tsukuba TCCON XCO <sub>2</sub> (B) and the
quality control items not satisfactory for data release. Aerosol optical thickness (AOT) was
defined at the wavelength of 1600 nm. SNR is signal-to-noise ratio.

Date	A (ppm)	<i>B</i> (ppm)	(A - B)/B (%)	Quality control
11 Sep 2009	371.02	382.44	-2.99	AOT = 1.09
11 Oct 2009	376.58	385.62	-2.34	
6 Jan 2010	376.34	389.68	-3.42	SNR = 94.5 at band 1
27 Jan 2010	381.31	391.20	-2.53	
5 Feb 2010	380.33	390.20	-2.53	
14 Feb 2010	372.42	391.43	-4.86	not converged AOT = 0.93
20 Feb 2010	386.41	391.27	-1.24	
23 Feb 2010	379.41	391.41	-3.07	
22 Mar 2010	380.66	390.10	-2.42	AOT = 1.01



Table 3. Characteristics of the two-wavelength polarization lidar.

Transmitter		
Laser	Nd:YA	٩G
Wavelength	532 nm	1064 nm
Pulse energy	150 mJ	150 mJ
Pulse repetition rate	10 H	z
Beam divergence	0.2 mi	rad
Receiver		
Telescope type	Ritchy-Chretier	n (advanced)
Telescope diameter	30.50	cm
Field of view (full angle)	1.0 mi	rad
Interference filter (FWHM)	0.28 nm	0.38 nm
Transmission	58 %	58%
Polarization measurement	Yes	No
Number of receiving channel	3 ( <i>P</i> : 2, <i>S</i> : 1)	1
Detector	PMT (R3234-01)	APD (Silicon)
Transient recorder	12bit A/D + PC	12 bit A/D
Minimum time resolution	1 mi	n
Minimum altitude resolution	7.5 r	n



Table 4.	Optical	thicknes	s at 50	0 nm (	$(\tau_{500}),$	single-	scatt	ering a	lbedo	at 5	500 nm	( <i>w</i> <sub>500</sub>	), and
Angstrom	expone	ent ( $\alpha$ ) ob	oserved	by sky	y radio	ometer,	and	aeroso	ol optic	al th	nicknes	s at 5	32 nm
$(\tau_{532})$ dete	ermined	by lidar a	at Tsuk	uba.									

Date	Sky radiometer			
	$ au_{500}$	@ <sub>500</sub>	α	$ au_{532}$
11 Sep 2009	0.276	0.956	2.102	0.201
11 Oct 2009	0.087	0.840	2.095	0.098
6 Jan 2010	0.079	1.0	2.120	0.091
27 Jan 2010	no data	no data	no data	0.160
5 Feb 2010	0.093	1.0	2.364	0.146
14 Feb 2010	0.230	1.0	1.534	0.165
20 Feb 2010	0.109	0.999	2.130	0.116
23 Feb 2010	0.279	0.997	1.416	0.181
22 Mar 2010	0.180	0.999	0.785	0.187



Discussion Pa	<b>ACPD</b> 11, 29883–29914, 2011							
per   Discussion	Influence of aerosols and thin cirrus clouds O. Uchino et al.							
Pap	Title	Title Page						
<u>e</u>	Abstract	Introduction						
	Conclusions	References						
iscussi	Tables	Figures						
on Pa	14	►I.						
aper	•	•						
_	Back	Close						
Discussion	Full Scree Printer-frier	en / Esc Idly Version						
n Paper	Interactive Discussion							

**Table 5.** Physical parameters currently used for retrieval (Ver. 01.xx) and three case studies showing decreased biases of GOSAT SWIR  $XCO_2$  data.

Ver. 01.xx	Aerosol vertical profile 0~2 km	Aerosol optical characteristics SPRINTARS	Cirrus No	Solar irradiance database Kurucz	XCO <sub>2</sub> SWIR
Case 1	lidar	sulfate and dust	Yes	Kurucz	revised
Case 2	lidar	sulfate and dust	Yes	Toon	new
Case 3	Retrieved (SPRINTARS as a priori data)	SPRINTARS	Yes	Toon	simulated



Fig. 1. Comparison of TANSO-FTS SWIR  $XCO_2$  (ver. 01.xx) data with the Tsukuba TCCON  $XCO_2$  results.







**Fig. 2.** Vertical profiles of the backscattering ratio *R*, total depolarization ratio Dep, and wavelength exponent Alp, observed by lidar on 14, 20, and 23 February 2010. Tau (0.4-30 km) is the partial optical thickness at altitudes of 0.4-30 km at 532 nm.





**Fig. 3.** Vertical profiles of the optical thicknesses (left panel) and extinction coefficients (right panel) of sulfate and cirrus cloud particles at 532 nm on 14 February 2010. The vertical scale is pressure normalized to surface pressure. The values of 0.5 and 0.1 correspond to altitudes of about 5.5 and 16 km, respectively.



**Fig. 4.** Vertical profiles of optical thicknesses (left panel) and extinction coefficients (right panel) of sulfate and dust particles at 532 nm on 23 February 2010.



29908



**Fig. 5.** Comparison between the revised  $XCO_2$  data, retrieved using Kurucz's solar irradiance data and taking into account the vertical profiles of two types of aerosols and cirrus clouds determined from lidar and sky radiometer, and the Tsukuba TCCON  $XCO_2$  data (Table 5, Case 1).





**Fig. 6.** Comparison of the new  $XCO_2$  data, using Toon's solar irradiance data and taking into account the vertical profiles of two types of aerosols and cirrus clouds determined from lidar and sky radiometer, with the Tsukuba TCCON  $XCO_2$  data (Table 5, Case 2).





Fig. 7. Comparison of retrieved optical thickness at 532 nm (Case 2) with a priori values estimated from lidar measurements.





Fig. 8. Comparison of retrieved surface pressure (Case 2) with a priori pressure.







29913

Interactive Discussion



**Fig. 10.** Comparison of simulated  $XCO_2$  data, obtained by retrieving the aerosol profile based on the a priori vertical profile and fixed aerosol optical characteristics given by the SPRINTARS model, with the Tsukuba TCCON  $XCO_2$  data (Table 5, Case 3).

