Reply to the comments by Referee #2

To Associate Editor and Referee #2,

We appreciate you reading our paper carefully and giving valuable comments and suggestions again. We have considered your recommendations for revisions and made the necessary changes. The major points that we deal with in the revised manuscript are as follows:

1. We have changed “V1.0” to “V1” throughout the text. The GOSAT project has released V01.00, V01.01, and V01.20 produces, but the CO2 products of all the three versions are exactly the same data.
2. We have eliminated Figure 2 of the original manuscript not to defocus the scope of this paper that discusses UTLS CO2 data. We have described that the simultaneous retrieval of surface parameters did not affect retrieved CO2 concentrations in the UTLS regions, but could increase the number of normally retrieved CO2 data.
3. Following the recommendations of the two Referees, we have investigated the effect of considering TIR CO2 averaging kernel functions on CO2 concentrations in the UTLS regions. For this purpose, we have done the two types of analysis.
   3-1. We have compared TIR and CONTRAIL CME CO2 data with and without TIR CO2 averaging kernel functions over each of the nine airports, and showed the comparison results in Figure 4 of the revised manuscript. Here, we have created CO2 vertical profiles using CME ascending/descending CO2 data below the tropopause and stratospheric CO2 concentrations taken from the Nonhydrostatic Icosahedral Atmospheric Model (NICAM)–Transport Model (TM) (Niwa et al., 2011) that introduced CONTRAIL CO2 data to the inverse model (Niwa et al., 2012), and then applied TIR CO2 averaging kernel functions to the created profiles.
   3-2. Keeping the detailed evaluation of 2-1 in mind, we assumed a CO2 vertical profile on the basis of the combination of CONTRAIL CME level flight CO2 data (“CONTRAIL (raw)”) and CarbonTracker CT2013B monthly-mean CO2 profiles (Peters et al., 2007) at each of the CME level flight measurement locations, applied TIR CO2 averaging kernel functions to the assumed profiles, and then compared the CO2 data with averaging kernels (“CONTRAIL (AK)”) with TIR CO2 data in the UTLS regions. In Figure 5, 6, and 7 of the revised manuscript, we showed the comparison results of both the CONTRAIL (raw) and CONTRAIL (AK) data.
4. We have eliminated Figure 5 of the original manuscript. This is because we have evaluated the effect of considering TIR CO2 averaging kernel functions on TIR and CONTRAIL CME CO2 comparisons quantitatively in the revised manuscript.
5. Following the suggestion of Referee #1, we have showed the comparison results for each latitude band, instead of showing the comparison results for each airline route, in Figure 7 of the revised manuscript. We have also modified Table 2 to show the bias values of TIR CO2 data against CONTRAIL (AK) CO2 data.
6. We have eliminated Figure 10 of the original manuscript to avoid speculative discussion.

Individual responses to the two Referees’ comments are listed below.

Reply to Referee #2

General comments:

Authors compare GOSAT TIR CO2 retrievals to carbon dioxide observations by CONTRAIL flights, both to vertical profiles and to level flight data. The comparison study is an important step towards improving the TIR CO2 retrieval product and should present a valuable source of information to potential users of CO2 product. However, current version of the manuscript requires revisions that are likely to alter the results and conclusion before the paper can be considered fully suitable for publication.

The reported results include TIR CO2 comparison with vertical profiles around Narita airport and with
level flight data observed around the world. Comparisons for Narita profiles are made with averaging 
kernel (AK) smoothing applied and reveal low bias (Fig. 2) with respect to CONTRAIL data in mid 
troposphere along with a significant random error at the range of several ppm. This part of comparison 
looks valid. In the following sections the TIR CO2 retrievals are compared to the level flight data, but it is 
done without applying corrections with averaging kernel (as presented in Eq. 5). Comparison without 
applying correction is, however, of a limited value for this type of remote sensing product. As authors 
state in multiple occasions the TIR retrieval does show strong dependence on the retrieval prior, 
indicating large weight of the prior in the retrieval. Potential users of the product for inverse modeling 
applications have to apply corrections themselves according to established practice, otherwise they would 
end up using mostly prior model simulation instead of observations by GOSAT TIR. Which is not what they 
intend. For the same reason there are few or no known published attempts to compare similar TIR product to 
level flight data that do not resolve vertical concentration profile. Accordingly, revision of the level flight 
part is strongly recommended. As the manuscript title suggests the comparison at UTLS level making a 
major contribution to the study results, a major revision is required.

Reply:
We appreciate your comments. We have seriously taken your comments, and made 
comparisons between TIR and CONTRAIL CME level flight CO2 data with considering TIR CO2 
averaging kernel functions in the revised manuscript. As described above, we have used 
CarbonTracker CT2013B monthly-mean CO2 profiles (Peters et al., 2007) to assume a CO2 
vertical profile at each of the CME level flight measurement locations. Then, we applied TIR 
CO2 averaging kernel functions to the assumed profiles to smooth them to the vertical resolution 
of TANSO-FTS TIR observations, and defined them as “CONTRAIL (AK).” Then, we extracted 
CONTRAIL (AK) data that corresponded to the TIR retrieval layers where TIR CO2 data were 
compared with original CONTRAIL CME data (“CONTRAIL (raw)”), and compared their 
averages with the TIR CO2 averages in the several layers. In Figure 5, 6, and 7 of the revised 
manuscript, we showed the comparison results of both the CONTRAIL (raw) and CONTRAIL 
(AK) CO2 data.

Specific comments:
1. On Page 13006, Line 19 authors state they did not apply “TIR CO2 averaging kernels to CONTRAIL 
CME CO2”, and in the following discussion provide mostly verbal argument that skipping the correction is 
justified. Instead the reader would expect to see results of numerical tests supporting authors’ position. 
For products like GOSAT SWIR XCO2 comparison with uncorrected values produce essentially same result, 
but in TIR case the value of averaging kernel is well below 1 (Fig. 9) which implies large weight of the 
prior in the product. As follows from Eq 5, application of the said correction with relatively small values 
of averaging kernel (in the order of 0.2 or less as shown on Fig 9) may attract the corrected value strongly 
to prior concentration, reducing difference between CONTRAIL and prior; from what is shown on Figs 6 
and 7 by several times and compromising much of discussion in Section 6. From the reviewer’s standpoint, 
revision and improvement of comparison with CONTRAIL level flight data is essential, which could be most 
easily done by extending the level flight data vertically using modeled or climatological profiles, and 
applying the averaging kernel afterwards. Importance of the correction given by Eq. 5 is emphasized by its 
use as a regular practice in the inverse modeling applications, where the model vs observation difference 
is estimated using same equation. Use of the Eq. 5 by modelers in calculating model to observation misfit 
effectively implies replacing the retrieval prior by model simulated profile. Comparison with uncorrected data, 
such as shown on Fig 7, may divert potential users from understanding strong and weak points of TIR L2 
data.

Reply:
We appreciate your comments. In the revised manuscript, we have quantitatively discussed the 
effect of considering TIR CO2 averaging kernel functions on CO2 concentrations in the UTLS 
regions. Over the nine airports, we have created CO2 vertical profiles using CONTRAIL CME 
ascending-descending CO2 data below the tropopause and stratospheric CO2 concentrations taken 
from the Nonhydrostatic Icosahedral Atmospheric Model (NICAM)-Transport Model (TM) 
(Niwa et al., 2011), and applied TIR CO2 averaging kernel functions to the created profiles. Then, 
we have compared TIR and CONTRAIL CME CO2 data with and without TIR CO2 averaging 
kernel functions over each of the nine airports, and showed the comparison results in Figure 4 of 
the revised manuscript. The NICAM-TM CO2 data used in this evaluation were calculated on the 
basis of surface flux data that were estimated in the inverse model that introduced CONTRAIL.
CO₂ data in addition to surface CO₂ data; therefore, we think they are appropriate datasets to evaluate the impact of considering TIR CO₂ averaging kernel functions on the CONTRAIL CME UTLS CO₂ data. Furthermore, we have assumed a CO₂ vertical profile on the basis of the combination of CONTRAIL CME level flight CO₂ data and CarbonTracker CT2013B monthly-mean CO₂ profiles (Peters et al., 2007) at each of the CME level flight measurement locations, applied TIR CO₂ averaging kernel functions to the assumed profiles, and then compared the CO₂ data with averaging kernels with TIR CO₂ data in the UTLS regions. The CarbonTracker CT2013B CO₂ data are available to the public, so that readers can refer to the dataset that we used as a CO₂ climatological dataset. We have also modified Table 2 to show the latitudinal dependence of the bias of TIR CO₂ data against CONTRAIL CO₂ data with averaging kernels, so that it should be useful for users to correct the TIR CO₂ data.

2. In the abstract (P12994 L24) and other locations authors mention the retrieval prior CO₂ they use have several biases. Another source of retrieval biases is spectral bias. It is not clear how large is relative contribution of these two. Comparison of the TIR product made with another prior or bias corrected prior appears desirable, given contamination of the product with prior biases. Also, from the absence of the prior contributors in the coauthors list and acknowledgements one can suspect that there wasn’t enough contact between prior developers and retrieval team.

Reply: Following your suggestions, we have evaluated the effect of L1B spectral bias on retrieved CO₂ concentrations in the UTLS regions by the equation of \( \frac{dCO₂}{dspec} = G_{CO₂} d_{spec} \). \( G_{CO₂} \) is a gain matrix for CO₂ retrieval, \( d_{spec} \) is a spectral bias vector that was created assuming that the V161.160 spectra had the same bias as V130.130 L1B spectra reported in Kataoka et al. (2014), and \( dCO₂ \) is a vector of bias errors in retrieved CO₂ concentrations attributable to the spectral bias. In order to evaluate the effect of a priori uncertainty on retrieved CO₂ concentrations, we arbitrarily decreased a priori concentration by 1% in test TIR CO₂ retrieval, and then compared the retrieved CO₂ concentrations with those retrieved using the original a priori data. Please see the second and third paragraphs of Section 7 of the revised manuscript for further details. We had reported a negative bias seen in the a priori UTLS CO₂ data with Dr. Saeki and Dr. Maksyutov who are responsible to create the a priori data for the TANSO-FTS retrieval processing, and discussed this problem with them. As you suggested, we should have acknowledged them in this paper. They have a plan to recreate a priori data for the future reprocessing of both TANSO-FTS SWIR and TIR L2 data.

Technical corrections:

P12994 L2. The first sentence better to rewrite to avoid using construct as “thermal infrared (TIR) band . . . has been observing carbon dioxide . . .”. Clearer phrase could sound as “TANSO-FTS has been observing carbon dioxide in thermal infrared (TIR) band”. Similar wording appears later as well. Authors write on P12996 L10 “. . . (TES) has retrieved CO₂ concentrations . . .”. One may argue that TES can measure or observe radiances, but retrieval would be done on the ground. The paper should be checked again to correct places with somewhat tentative language.

Reply: We have rewritten the text following your suggestions. We greatly appreciate your suggestions.
Validation of GOSAT/TANSO-FTS TIR UTLS CO₂ data
(Version 1.0) using CONTRAIL measurements

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Abstract

The thermal infrared (TIR) band of the Thermal and Near Infrared Sensor for Carbon Observation (TANSO)–Fourier Transform Spectrometer (FTS) on board the Greenhouse Gases Observing Satellite (GOSAT) has been observing carbon dioxide (CO₂) concentrations in several atmospheric layers in the thermal infrared (TIR) band since its launch. This study compared TANSO-FTS TIR V1.0V1 CO₂ data and CO₂ data obtained in the Comprehensive Observation Network for TRace gases by AIrLiner (CONTRAIL) project in the upper troposphere and lower stratosphere (UTLS), where the TIR band of TANSO-FTS is most sensitive to CO₂ concentrations, to validate the quality of the TIR V1.0V1 UTLS CO₂ data from 287 to 162 hPa. We first evaluated the impact of considering TIR CO₂ averaging kernel functions on CO₂ concentrations using CO₂ profile data obtained by the CONTRAIL Continuous CO₂ Measuring Equipment (CME), and found that the impact at around the CME level flight altitudes (~11 km) was on average less than 0.5 ppm in low latitudes and less than 1 ppm in middle and high latitudes. From a comparison made during flights between Tokyo and Sydney, the averages of the TIR upper atmospheric CO₂ data were within 0.1% of the averages of the CONTRAIL CME CO₂ data with and without TIR CO₂ averaging kernels for all seasons in the Southern Hemisphere agreed well with the averages of the data obtained by
the CONTRAIL Continuous CO₂-Measuring Experiment (CME) within 0.1% for all of the
seasons in the Southern Hemisphere. The results of a-comparisons for all of the eight airline
routes showed that the agreements of TIR and CME CO₂ data were worse in spring and
summer than in fall and winter in the Northern Hemisphere in the upper troposphere. While
the differences between TIR and CME CO₂ data were on average within 1 ppm in fall and
winter, TIR CO₂ data had a negative bias up to 2.4 ppm against CME CO₂ data with TIR CO₂
averaging kernels in the northern low and middle latitudes in spring and summer, the
agreement between the TIR and CONTRAIL CO₂ data was within 0.5% on average in the
Northern Hemisphere, which was better than the agreement between a priori and CONTRAIL
CO₂ data. The quality of TIR lower stratospheric CO₂ data depends largely on the information
content, and therefore has a seasonal dependence. In high latitudes, TIR V1.0 lower
stratospheric CO₂ data are only valid in the summer. The magnitude of bias in the TIR upper
atmospheric CO₂ data did not have a clear longitudinal dependence. The comparison results
for flights in northern low and middle latitudes showed that the agreement between TIR and
CONTRAIL CO₂ data in the upper troposphere was worse in the spring and summer than in
the fall and winter. This could be attributed to a larger negative bias in the upper atmospheric
a-priori CO₂ data in the spring and summer and a seasonal dependence of spectral bias in
TANSO-FTS TIR Level 1B (L1B) radiance data. The negative bias in the northern middle
latitudes resulted in the maximum of TIR CO₂ concentrations being lower than that of
CME-CONTRAIL CO₂ concentrations, which led to an underestimate of the amplitude of
CO₂ seasonal variation.

1 Introduction

Carbon dioxide (CO₂) in the atmosphere is a well-known strong greenhouse gas (IPCC, 2013,
and references therein), with concentrations that have been observed both in situ and by
satellite sensors. Its long-term observation began in Mauna Loa, Hawaii, and the South Pole
in the late 1950s (Keeling et al., 1976a, 1976b, 1996). Since then, comprehensive CO₂
observations in the atmosphere have been conducted worldwide in several observatories and
tall towers (Bakwin et al., 1998), by aircraft flask sampling (e.g., Crevoisier et al., 2010), and
via the AirCore sampling system (Karion et al., 2010) in the framework of researches by the
National Oceanic and Atmospheric Administration (NOAA). Atmospheric CO₂
concentrations have gradually increased at a globally averaged annual rate of 1.7±0.5 ppm
from 1998 to 2011, although its growth rate has relatively large interannual variation (IPCC, 2013). Upper atmospheric CO₂ observations have been made in many areas by several projects using commercial airliners, such as the Comprehensive Observation Network for TRace gases by AIrLiner (CONTRAIL) project (Machida et al., 2008) and the Civil Aircraft for the Regular Investigation of the atmosphere Based on an Instrument Container (CARIBIC) project (Brenninkmeijer et al., 2007). Continuous long-term measurements of CO₂ made by several airplanes of Japan Airlines (JAL) in the CONTRAIL project have revealed details of its seasonal variation and interhemispheric transport in the upper atmosphere (Sawa et al., 2012) and interannual and long-term trends of its latitudinal gradients (Matsueda et al., 2015).

Atmospheric CO₂ observations by satellite sensors are categorized into two types: those utilizing CO₂ absorption bands in the shortwave infrared (SWIR) regions at around 1.6 and 2.0 μm, and those in the thermal infrared (TIR) regions at around 4.6, 10, and 15 μm. The Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) on the Environmental Satellite (ENVISAT) first observed CO₂ column-averaged dry-air mole fractions (XCO₂) from spectra at 1.57 μm (Buchwitz et al., 2005; Barkley et al., 2006). The Thermal and Near Infrared Sensor for Carbon Observation (TANSO)–Fourier Transform Spectrometer (FTS) on board the Greenhouse Gases Observing Satellite (GOSAT), which was launched in 2009 (Yokota et al., 2009), has observed XCO₂ with high precision by utilizing the 1.6 and/or 2.0 μm CO₂ absorption bands (Yoshida et al., 2011, 2013; O’Dell et al., 2012; Butz et al., 2011; Cogan et al., 2012). The Orbiting Carbon Observatory 2 (OCO-2) was successfully launched in 2014, and started regular observations of XCO₂ with high spatial resolution. Satellite CO₂ observations at TIR absorption bands have a longer history beginning with the High-Resolution Infrared Sounder (HIRS) (Chédin et al., 2002, 2003, 2005). The Atmospheric Infrared Sounder (AIRS) has achieved more accurate observations of middle and upper tropospheric CO₂ concentrations (Crevoisier et al., 2004; Chahine et al., 2005; Maddy et al., 2008; Strow and Hannon, 2008). The Tropospheric Emission Spectrometer (TES) has observed CO₂ concentrations in several vertical layers with high accuracy by taking advantage of its high wavelength resolution (Kulawik et al., 2010, 2013). The Infrared Atmospheric Sounding Interferometer (IASI) has observed upper atmospheric CO₂ amounts from its TIR spectra (Crevoisier et al., 2009). TANSO-FTS also has a TIR band in addition to its three SWIR bands, and obtains vertical information of CO₂ concentrations in addition to XCO₂ in the same field of view (Saitoh et al., 2009).
Rayner and O’Brien (2001) and Pak and Prather (2001) showed the utility of global CO\textsubscript{2} data obtained by satellite sensors for estimating its source and sink strength, and many studies of CO\textsubscript{2} inversion have been conducted using a huge amount of satellite data since the 2000s. Chevallier et al. (2005) first used satellite CO\textsubscript{2} data, observed with the Operational Vertical Sounder (TOVS), to estimate CO\textsubscript{2} surface fluxes. They reported that a regional bias in satellite CO\textsubscript{2} data hampers the outcomes. Nassar et al. (2011) demonstrated that the wide spatial coverage of satellite CO\textsubscript{2} data is beneficial to CO\textsubscript{2} surface flux inversion through the combined use of TES and surface flask CO\textsubscript{2} data, particularly in regions where surface measurements are sparse. In addition to CO\textsubscript{2} surface inversion results using TIR observations, global XCO\textsubscript{2} data observed with the SWIR bands of TANSO-FTS have been actively used for estimating CO\textsubscript{2} source and sink strength (Maksyutov et al., 2013; Saeki et al., 2013; Chevallier et al., 2014; Basu et al., 2013, 2014; Takagi et al., 2014). One of the important things to consider when incorporating satellite data in CO\textsubscript{2} inversion is the accuracy of the data, as suggested by Basu et al. (2013). Uncertainties in satellite CO\textsubscript{2} data should be assessed seasonally and regionally to determine the seasonal and regional characteristics of the satellite CO\textsubscript{2} bias.

The importance of upper atmospheric CO\textsubscript{2} data in the inversion analysis of CO\textsubscript{2} surface fluxes was discussed in Niwa et al. (2012). They used CONTRAIL CO\textsubscript{2} data in conjunction with surface CO\textsubscript{2} data to estimate surface flux, and demonstrated that adding middle and upper tropospheric data observed by the aircraft could greatly reduce the posteriori flux errors, particularly in tropical Asian regions. Middle and upper tropospheric and lower stratospheric CO\textsubscript{2} concentrations and column amounts of CO\textsubscript{2} can be simultaneously observed in the same field of view with TANSO-FTS on board GOSAT. Provided that the quality of upper atmospheric CO\textsubscript{2} data simultaneously obtained with TANSO-FTS is proven to be comparable to that of TANSO-FTS XCO\textsubscript{2} data (Yoshida et al., 2013; Inoue et al., 2013), the combined use of upper atmospheric CO\textsubscript{2} and XCO\textsubscript{2} data observed with TANSO-FTS could be a useful tool for estimating CO\textsubscript{2} surface flux.

GOSAT, which is the first satellite to be dedicated to greenhouse gas monitoring, was launched on January 23, 2009. As described above, the TIR band of TANSO-FTS on board GOSAT has been observing CO\textsubscript{2} concentrations in several vertical layers in the TIR band. In this study, we focused on CO\textsubscript{2} concentrations in the upper troposphere and lower stratosphere (UTLS), where the TIR band of TANSO-FTS is most sensitive. We validated these data by
comparison with upper atmospheric CO$_2$ data obtained in a wide spatial coverage in the
CONTRAIL project. Sections 2 and 3 explain the GOSAT and CONTRAIL measurements,
respectively. Section 4 details the retrieval algorithm used in the latest version 1.0 (V1.0) CO$_2$
level 2 (L2) product of the TIR band of TANSO-FTS. Section 5 describes the methods of
comparing TANSO-FTS TIR V1 L2 and CONTRAIL CO$_2$ data. Sections 6 and 7 show and
discuss the results of the comparisons between TANSO-FTS TIR V1.0 L2 and
CONTRAIL CO$_2$ data. Section 8 summarizes this study.

2 GOSAT observations

GOSAT is a joint satellite project of the National Institute for Environmental Studies (NIES),
Ministry of the Environment (MOE), and Japan Aerospace Exploration Agency (JAXA) for
the purpose of making global observations of greenhouse gases such as CO$_2$ and CH$_4$
(Hamazaki et al., 2005; Yokota et al., 2009). It was launched on January 23, 2009, from the
Tanegashima Space Center, and has continued its observations for more than six years.
GOSAT is equipped with the TANSO-FTS for greenhouse gas monitoring and the TANSO-
Cloud and Aerosol Imager (CAI) to detect clouds and aerosols in the TANSO-FTS field of
view (Kuze et al., 2009). TANSO-FTS consists of three bands in the SWIR region and one
band in the TIR region. The SWIR bands observe column amounts of greenhouse gases and the TIR band observes vertical information of gas
concentrations (Yoshida et al., 2011, 2013; Saitoh et al., 2009, 2012; Ohyama et al., 2012, 2013).
Kuze et al. (2012) provided a detailed description of the methods used for the processing and
calibration of level 1B (L1B) spectral data from TANSO-FTS. They explained the algorithm
for the version 150.151 (V150.151) L1B spectral data. The TIR V1.0 L2 CO$_2$ product we
focused on in this study was created from a later version, V161.160, of L1B spectral data. The
following modifications were made to the algorithm from V150.151 to V161.160: improving
the TIR radiometric calibration through the improvement of calibration parameters, turning
off the sampling interval non-uniformity correction, modifying the spike noise criteria of the
quality flag, and reevaluating the misalignment between the GOSAT satellite and TANSO-
FTS sensor. Kataoka et al. (2014) reported that the bias accuracies of TANSO-FTS TIR
V130.130 L1B radiance spectra based on comparisons with the Scanning High-resolution
Interferometer Sounder (S-HIS) spectra for warm scenes were 0.5 K at 800–900 cm$^{-1}$ and
700–750 cm⁻¹, 0.1 K at 980–1080 cm⁻¹, and more than 2 K at 650–700 cm⁻¹. Although the magnitude of the spectral bias evaluated on the basis of V130.130 L1B data would change in V161.160 L1B data, the issue of L1B spectral bias still remains. The spectral bias inherent in TIR L1B spectra would be mainly because of uncertainty of polarization correction. Another possible cause was discussed in Imasu et al. (2010). When retrieving CO₂ concentrations from the TIR band of TANSO-FTS, the spectral bias that is predominant in CO₂ absorption bands should be considered (Ohyama et al., 2013).

3 CONTRAIL Continuous Measurement Equipment (CME) observations

We used CO₂ data obtained in the CONTRAIL project to validate the quality of TANSO-FTS TIR V1.0V1 L2 CO₂ data. CONTRAIL is a project to observe atmospheric trace gases such as CO₂ and CH₄ using instruments installed on commercial aircraft operated by JAL. Observations of trace gases in this project began in 2005. Two types of measurement instruments, the Automatic Air Sampling Equipment (ASE) and the Continuous CO₂ Measuring Equipment (CME), have been installed on several JAL aircraft to measure trace gases over a wide area (Machida et al., 2008).

This study used CO₂ data obtained with CME on several airline routes from Narita Airport, Japan. CO₂ observations with CME use a LI-COR LI-840 instrument that utilizes a nondispersive infrared absorption (NDIR) method (Machida et al., 2008). In the observations, two different standard gases, with CO₂ concentration of 340 ppm and 390 ppm based on NIES09 scale, are regularly introduced into the NDIR for calibration. The accuracy of CME CO₂ measurements is 0.2 ppm. See Machida et al. (2008), Matsueda et al. (2008), and Machida et al. (2011) for details of the CME CO₂ observations and their accuracy and precision.

4 Retrieval algorithm of TANSO-FTS TIR V1.0V1 CO₂ data

4.1 Basic retrieval settings

Saitoh et al. (2009) provided an algorithm for retrieving CO₂ concentrations from the TIR band of TANSO-FTS. The first version, V00.01, of the L2 CO₂ product of the TIR band of TANSO-FTS was basically processed by the algorithm described in Saitoh et al. (2009). The
V1.0V1 L2 CO2 product that we focused on in this study also adopted a non-linear maximum a posteriori (MAP) method with linear mapping, as was the case for the V00.01 product. We utilized the following expressions in TIR CO2 retrieval:

\[ \hat{z}_{i+1} = W^* \hat{x}_a + G[y - F(\hat{x}_i)] + K_i W(W^* \hat{x}_i - W^* x_a)] \]

\[ G = [W^T K_i S_{\varepsilon}^{-1} K_i W + (W^* S_a W^* T_{\varepsilon})^{-1}] W^T K_i S_{\varepsilon}^{-1} \]

where \( x_a \) is an a priori vector, \( S_a \) is a covariance matrix of the a priori vector, \( S_{\varepsilon} \) is a covariance matrix of measurement noise, \( K_i \) is a CO2 Jacobian matrix calculated using the \( i^{th} \) retrieval vector \( \hat{x}_i \) on full grids, \( F(\hat{x}_i) \) is a forward spectrum vector based on \( \hat{x}_i \), \( y \) is a measurement spectrum vector, and \( \hat{z}_{i+1} \) is the \( i+1^{th} \) retrieval vector defined on retrieval grids. \( W \) is a matrix that interpolates from retrieval grids onto full grids. \( W^* \) is the generalized inverse matrix of \( W \).

The full grids are vertical layer grids for radiative transfer calculation, and the retrieval grids are defined as a subset of the full grids. In the V1.0V1 L2 CO2 retrieval algorithm, linear mapping between retrieval grids and full grids was also applied, but the number of full grid levels was 78 instead of 110 in the V00.01 algorithm. The determination of retrieval grids in the V1.0V1 algorithm basically followed the method of the V00.01 algorithm. It was based on the areas of a CO2 averaging kernel matrix in the tropics, but the retrieval grid levels were fixed for all of the retrieval processing, as presented in Table 1. Averaging kernel matrix \( A \) is defined (Rodgers, 2000) as

\[ A = GKW \]  

(2)

Figure 1 shows typical averaging kernel functions of TIR V1.0V1 L2 CO2 retrieval in middle latitudes in summer. The degrees of freedom (DF) in these cases (trace of the matrix \( A \)) were (a) 2.22, (b) 1.81, and (c) 1.36, respectively. The seasonally averaged DF values of TIR V1.0V1 CO2 data ranged from 1.12 to 2.35. In the low and middle latitudes between 35°N and 35°S, almost all the CO2 DF values were around exceeded 2.0 or more; this means that observations by the TIR band of TANSO-FTS can provide information on CO2 concentrations in more than two vertical layers, one of which we focused on in this study.
A priori and initial values for CO$_2$ concentrations were taken from the outputs of the NIES transport model (NIES-TM05) (Saeki et al., 2013). A priori and initial values for temperature and water vapor were obtained from Japan Meteorological Agency (JMA) Grid Point Value (GPV) data. Basically, the retrieval processing of TANSO-FTS was only conducted under clear-sky conditions, which was judged based on a cloud flag from TANSO-CAI in the daytime (Ishida and Nakajima, 2009; Ishida et al., 2011) and on a TANSO-FTS TIR spectrum in the nighttime.

4.2 Improvements in the TIR V1.0V1 CO$_2$ algorithm

The following conditions are the improvements made in the TANSO-FTS TIR V1.0V1 L2 CO$_2$ algorithm from the V00.01 algorithm. The V1.0V1 algorithm used the CO$_2$ 10 μm absorption band in addition to the CO$_2$ absorption band at around 15 μm band; the wavelength regions of 690–750 cm$^{-1}$, 790–795 cm$^{-1}$, 930–990 cm$^{-1}$, and 1040–1090 cm$^{-1}$ were used in the CO$_2$ retrieval. We did not apply any channel selection. In these wavelength regions, temperature, water vapor, and ozone concentrations were retrieved simultaneously with CO$_2$ concentration. Moreover, surface temperature and surface emissivity were simultaneously derived as a correction parameter of the spectral bias inherent in TANSO-FTS TIR V161.160 L1B spectra at the above-mentioned CO$_2$ absorption bands. We assumed that the spectral bias could be divided into two components: a wavelength-dependent bias whose amount varied depending on wavelength and a wavelength-independent bias whose amount was uniform in a certain wavelength region. We tried to correct such a wavelength-independent component of the spectral bias by adjusting the value of surface temperature. Similarly, a wavelength-dependent component of the spectral bias was corrected by adjusting the value of surface emissivity in each wavelength channel. Therefore the matrices of $K$ and $S_a$ of expression (1) are as follows:

$$K = \begin{pmatrix} K_{CO2} & K_{H2O} & K_{O3} & K_T & k_{S1} & k_{S2} & k_{S3} & k_{S4} & k_{S5} & k_{E1} & k_{E2} & k_{E3} & k_{E4} & k_{E5} \end{pmatrix}, \ (3)$$
where $K_{CO2}$, $K_{H2O}$, $K_{O3}$, and $K_T$ are Jacobian matrices of CO2, water vapor, ozone, and temperature on full grids, respectively, and $S_{CO2}$, $S_{H2O}$, $S_{O3}$, and $S_T$ are a priori covariance matrices of CO2, water vapor, ozone, and temperature on full grids, respectively. The vectors $k_{ST_1}$, $k_{ST_2}$, $k_{ST_3}$, $k_{ST_4}$, and $k_{ST_5}$ are the Jacobian vectors of surface temperature in the wavelength regions of 690–715 cm$^{-1}$, 715–750 cm$^{-1}$, 790–795 cm$^{-1}$, 930–990 cm$^{-1}$, and 1040–1090 cm$^{-1}$, respectively. The vectors $k_{SE_1}$, $k_{SE_2}$, $k_{SE_3}$, $k_{SE_4}$, and $k_{SE_5}$ are the Jacobian vectors of surface emissivity in each of the five wavelength regions, respectively. The elements of the Jacobian vectors of surface parameters that were defined for each of the five wavelength regions were set to be zero in the other wavelength regions. The values $S_{ST_1}$, $S_{ST_2}$, $S_{ST_3}$, $S_{ST_4}$, and $S_{ST_5}$ and $S_{SE_1}$, $S_{SE_2}$, $S_{SE_3}$, $S_{SE_4}$, and $S_{SE_5}$ are a priori variances of surface temperature and surface emissivity in each of the five wavelength regions, respectively. Simultaneous retrieval of the surface parameters in the V1.0V1 algorithm was conducted just for the purpose of correcting the TIR V161.160 L1B spectral bias; it had no physical meaning. We estimated the surface parameters separately in each of the five wavelength regions to consider differences in the amount of spectral bias in each wavelength region. The matrices $S_a$ for CO2, temperature, water vapor, and ozone were diagonal matrices with vertically fixed diagonal elements with a standard deviation of 2.5%, 3 K, 20%, and 30%, respectively. Here, a priori and initial values for ozone were obtained from the climatological data for each latitude bin for each month given by MacPeters et al. (2007). We assumed rather
large values as a priori variances of the surface parameters (a standard deviation of 10 K for
surface temperature), which could allow more flexibility in the L1B spectral bias correction
by the surface parameters. The a priori and initial values for surface emissivity were
calculated by linear regression analysis using the Advanced Space-borne Thermal Emission
Reflection Radiometer (ASTER) Spectral Library (Baldridge et al., 2009) using on the basis
of land-cover classification, vegetation, and wind speed information. The a priori and initial
values for surface temperature were estimated using radiance data in several channels around
900 cm\(^{-1}\) of the TIR V161.160 L1B spectra.

4.3 Effects of spectral bias on CO\(_2\) retrieval

In the TIR V1.0V1 L2 algorithm, we estimated surface temperature and surface emissivity to
correct the spectral bias inherent in the TANSO-FTS TIR L1B spectra (Kataoka et al., 2014).
The existence of a relatively large spectral bias around the CO\(_2\) 15 μm absorption band in
TANSO-FTS TIR L1B spectra (Kataoka et al., 2014) resulted in a decrease in the number of
normally retrieved CO\(_2\) profiles. This is probably because the TIR L1B spectral bias in the
CO\(_2\) 15 μm absorption band was sometimes too large for the L2 retrieval calculation to
converse in a limited iteration. The correction of the TIR L1B spectral bias through the
simultaneous retrieval of the surface parameters did not affect retrieved CO\(_2\) concentrations in
the UTLS regions, which was the focus of this study, but it altered the number of normally
retrieved CO\(_2\) profiles. Here, we evaluated the impact of the correction of the TIR L1B
spectral bias through the simultaneous retrieval of the surface parameters on the TIR L2 CO\(_2\)
retrieval. Figure 2 shows comparisons between several types of TIR CO\(_2\) profiles retrieved by
changing the treatment of the surface parameters in the retrieval and coincident CONTRAIL
CME CO\(_2\) profiles over Narita airport. Criteria for the coincident pairs of a 100 km distance
from Narita airport, a time difference in 2 hours, and a day difference within ±1 day yielded a
total of 141 coincident profile pairs in 2010. In the comparisons, we applied averaging kernel
functions of TIR CO\(_2\) data to corresponding CONTRAIL CME CO\(_2\) profiles, as follows
(Rodgers and Connor, 2003):

\[
x_{\text{obs-CO2}} = x_{\text{a priori}} + A(x_{\text{CONTRAIL}} - x_{\text{a priori}}) + (\text{5})
\]

Here, \(x_{\text{CONTRAIL}}\) and \(x_{\text{a priori}}\) are CONTRAIL CME and a priori CO\(_2\) profiles. Figure 2a shows
a comparison of the V1.0 L2 CO\(_2\) product (i.e., the result of a comparison of CO\(_2\) retrievals
based on TANSO-FTS TIR L1B spectra corrected through the simultaneous retrieval of both

surface temperature and surface emissivity). Figure 2b and 2c show the results of a comparison of CO₂ retrievals that used TIR L1B spectra corrected only by surface temperature and surface emissivity, respectively. Figure 2d shows the result of a comparison of CO₂ retrievals from uncorrected original TIR L1B spectra. The existence of a relatively large spectral bias around the CO₂–15 μm absorption band in TANSO-FTS TIR L1B spectra (Kataoka et al., 2014) resulted in a decrease in the number of normally retrieved CO₂ profiles. In the V1.0 case (Figure 2a), CO₂ profiles were normally retrieved for 74 of the 114 coincident pairs. The comparison between Figure 2a and 2c (Figure 2b and 2d) demonstrated that the correction of the TIR L1B spectral bias through the simultaneous retrieval of surface temperature could increase the number of normally retrieved CO₂ profiles (in this case, from 48 to 74). This implies that a wavelength-independent component of the spectral bias in CO₂ absorption bands could be reduced by adjusting the value of surface temperature at the bands.

In contrast, the comparisons between Figure 2a and 2b and Figure 2c and 2d showed that the spectral bias correction of the TIR L1B spectral bias through the simultaneous retrieval of surface emissivity did not increase the number of normally retrieved CO₂ profiles had a relatively small impact on TIR L2 CO₂ retrieval. If the TIR L1B spectral bias has a wavelength dependence, nevertheless, surface emissivity, which has a wavelength dependence, could be effective for correcting such a wavelength-dependent bias in wavelength-dependent L1B spectral bias. A more effective method of L1B spectral bias correction based on surface emissivity should be considered in the next version of the TIR L2 CO₂ retrieval algorithm, if a future version of the TIR L1B spectral data still has a bias.

5 Comparison methods of TANSO-FTS TIR V1.0 upper atmospheric CO₂ data with CME CO₂ data

5.1 Area comparisons

Here, we used the level flight CO₂ data of CONTRAIL CME observations in 2010 to validate the quality of UTLS CO₂ data from the TANSO-FTS TIR V1.0 V1 L2 CO₂ product. The level flight data obtained from the following eight airline routes of the CONTRAIL CME observations were used in this study: Tokyo–Amsterdam (NRT–AMS) and Tokyo–Moscow (NRT–DME), Tokyo–Vancouver (NRT–VYR), Tokyo–Honolulu (NRT–HNL), Tokyo–Bangkok (NRT–BKK), Tokyo–Singapore (NRT–SIN) and Tokyo–Jakarta (NRT–CGK), and
Tokyo–Sydney (NRT–SYD). We merged the level flight data of Tokyo–Amsterdam and Tokyo–Moscow into “Tokyo–Europe”, and the data of Tokyo–Singapore and Tokyo–Jakarta into “Tokyo–East Asia”. Figure 23 shows the flight tracks of all of the CONTRAIL CME observations in 2010 used in this study. As shown in the figure, we divided the CONTRAIL CME level flight data into 40 areas following Niwa et al. (2012), and compared them with TANSO-FTS TIR CO2 data in each area in each season. The level flight data in each area were averaged for each season (MAM, JJA, SON, and JF/DJF). The amount of level flight data varied depending on the area and season. The largest amount of data was obtained in area 15 over Narita Airport, where 4,694–9,306 data points were obtained. A relatively small amount of level flight data, 79–222 data points, was obtained in area 1 over Amsterdam. In all 40 areas, we collected sufficient level flight data to undertake comparison analysis based on the average values, except for seasons and regions with no flights.

5.2 Comparisons of CME profiles with and without averaging kernels

In comparisons of TIR V1 L2 CO2 data with the CONTRAIL CME level flight data, it is difficult to smooth the CME data by applying TIR CO2 averaging kernels, because CO2 concentrations below and above the CME flight levels were not observed. Here, we evaluated the impact of considering averaging kernel functions on CO2 concentrations using the CME profile data. We regarded the CME data obtained during the ascent and descent flights over the nine airports as part of CO2 vertical profiles, and investigated differences between TIR and CME CO2 data with and without applying averaging kernel functions in the altitude regions around the CME level flight observations. We assumed the CME ascending/descending CO2 concentration at the uppermost altitude level to be constant up to the tropopause height, following the method proposed by Araki et al. (2010). We used stratospheric CO2 data taken from the Nonhydrostatic Icosahedral Atmospheric Model (NICAM)–Transport Model (TM) (Niwa et al., 2011; 2012) to create whole CO2 vertical profiles over the airports. The NICAM-TM CO2 data used here introduced CONTRAIL CO2 data to the inverse model in addition to surface CO2 data, and therefore could simulate upper atmospheric CO2 concentrations well (Niwa et al., 2012). We determined the stratospheric CO2 profile by assuming the CO2 concentration gradients, calculated on the basis of the NICAM-TM CO2 data above the tropopause height.
To compare these CME CO₂ profiles with TIR CO₂ data, we calculated a weighted average of all the CME CO₂ data included in each of the 28 retrieval grid layers with respect to altitude, and defined the CO₂ data in the 28 layers as “CONTRAIL (raw)” data. Then, we selected TIR CO₂ data that coincided with each of the CONTRAIL (raw) profiles. The criteria for the coincident pairs were a 300 km distance from Narita airport, and a 3-day difference of each other observation. We applied TIR CO₂ averaging kernel functions to the corresponding CONTRAIL (raw) profile, as follows (Rodgers and Connor, 2003):

\[
x_{\text{CONTRAIL (AK)}} = x_{\text{a priori}} + A(x_{\text{CONTRAIL (raw)}} - x_{\text{a priori}}).
\]

Here, \(x_{\text{CONTRAIL (raw)}}\) and \(x_{\text{a priori}}\) are CONTRAIL (raw) and a priori CO₂ profiles. We defined the CONTRAIL (raw) data with TIR CO₂ averaging kernel functions as “CONTRAIL (AK)” data.

### 5.3 Level flight comparisons

In this study, we made comparisons between TIR and CONTRAIL CME level flight CO₂ data in two ways. The first was a direct comparison with original CME CO₂ data, i.e., CONTRAIL (raw) data. The second was a comparison with CONTRAIL (AK) data in the altitude regions around the CME level flight observations that were based on “assumed CO₂ profiles” created at each of the measurement locations of all the CME level flight data. In the first comparison with CONTRAIL (raw) data, the CME level flight data in each of the 40 areas were averaged for each season (MAM, JJA, SON, and JF/DJF). In all 40 areas, we collected an enough amount of level flight data to undertake a comparative analysis based on the average values, except for seasons and regions with no flights. The average altitude of all of the CONTRAIL CME level flight data used here was 11.245 km. The airline routes of Tokyo–Europe, Tokyo–Vancouver, and Tokyo–Honolulu contained both tropospheric and stratospheric data in the areas along their routes; therefore, we calculated the average and standard deviation values separately. Here, we differentiated between the tropospheric and stratospheric level flight data on the basis of temperature lapse rates from the JMA GPV data that were interpolated to the CONTRAIL CME measurement locations. The average altitudes of the tropospheric and stratospheric level flight data from the airline route between Tokyo and Europe were 10.84 km and 11.18 km, respectively.

In the comparison with CONTRAIL (raw) data, Next, we selected TANSO-FTS TIR V4.0V1 L2 CO₂ data that were in the altitude range within ±1 km of the average altitude of the
CME-CONTRAIL level flight data for each area for each season, and calculated their averages and standard deviations. Similarly, we calculated the averages and standard deviations of the corresponding a priori CO₂ data for each area for each season. For the airline routes of Tokyo–Europe, Tokyo–Vancouver, and Tokyo–Honolulu, the averages and standard deviations of TIR V1.0 V1 CO₂ data and the corresponding a priori CO₂ data were calculated separately for the tropospheric and stratospheric data. In this calculation, we first selected TIR V1.0 V1 CO₂ data that were collected in a range within ±1 km of the average altitudes of the CONTRAIL tropospheric and stratospheric CO₂ data for each area. Then, we classified each of the selected TIR CO₂ data points into tropospheric and stratospheric data on the basis of the temperature lapse rates from the JMA GPV data that were interpolated to the TANSO-FTS measurement locations, and calculated the seasonal averages and standard deviations for the reselected tropospheric and stratospheric TIR CO₂ data. This procedure was required for two reasons: (1) One was that the tropopause height at each TANSO-FTS measurement location should differ on a daily basis, and (2) the other was that because TANSO-FTS TIR CO₂ data were selected within the range of 2 km, some tropospheric TIR CO₂ data were selected on the basis of the CONTRAIL stratospheric level flight data, and vice versa. Figure 34 shows the number of TANSO-FTS TIR CO₂ data points that were finally selected in each retrieval layer for each of the airline routes. The TIR CO₂ data used in the comparative analysis were mainly from layers 9 and layer 10 (from 287 to 196 hPa) for the tropospheric comparison and from layers 10 and layer 11 (from 237 to 162 hPa) for the stratospheric comparison.

In the second comparison, we assumed a CO₂ vertical profile on the basis of CONTRAIL (raw) data at each of the CONTRAIL CME level flight locations, and applied TIR CO₂ averaging kernel functions to the assumed profiles. For this purpose, realistic CO₂ vertical profiles were required along the eight airline routes. In this study, we created a CO₂ profile at each CME level flight measurement location from CarbonTracker CT2013B monthly-mean CO₂ data (Peters et al., 2007). The CarbonTracker CT2013B CO₂ data are available to the public, and therefore readers can refer to the dataset that we used as a CO₂ climatological dataset. The method for creating a CO₂ vertical profile from the CONTRAIL (raw) and CarbonTracker CT2013B data is as follows. We first averaged all of the CarbonTracker CT2013B monthly-mean data included in each of the 40 areas to create area-averaged CarbonTracker CT2013B profiles. Then, we shifted the area-averaged CarbonTracker CT2013B profile so that its concentration fit to each of the CONTRAIL (raw) data at CME
level flight altitude. Finally, we applied area-averaged TIR CO$_2$ averaging kernel functions to each of the shifted area-averaged CO$_2$ profiles, and created profiles of CONTRAIL (AK) at all the CME level flight measurement locations.

We compared the CONTRAIL (AK) data with TIR CO$_2$ data at the altitude regions around the CME level flight observations for each area in each season. We extracted CONTRAIL (AK) data that corresponded to the TIR retrieval layers where TIR CO$_2$ data were compared to CONTRAIL (raw) data, and averaged them for each area for each season. For the airline routes of Tokyo–Europe, Tokyo–Vancouver, and Tokyo–Honolulu, we separately averaged CONTRAIL (AK) data created from tropospheric and stratospheric CONTRAIL (raw) data, and defined the averages as tropospheric and stratospheric CONTRAIL (AK) data, respectively. As shown in Figure 3, the CONTRAIL (AK) data used for the comparison during flights between Tokyo and Sydney consisted of CO$_2$ concentrations in layers 9 and 10 of the CONTRAIL (AK) profiles. For the flights between Tokyo and Europe, the CONTRAIL (AK) data used for the tropospheric and stratospheric comparisons were based on CO$_2$ concentrations in layers 9 and 10 and in layers 10 and 11 of CONTRAIL (AK) profiles, respectively.

We did not apply TIR CO$_2$-averaging kernels to CONTRAIL CME CO$_2$ data in the following UTLS analysis. Because CO$_2$ concentrations below and above the CONTRAIL CME flight levels were not observed except over airports, assuming a CO$_2$ vertical profile for each of CONTRAIL CME level flight data points and applying averaging kernels to the assumed CONTRAIL CO$_2$-profiles would increase the uncertainty in the CONTRAIL CO$_2$ data. Here, we assess the effect of not applying averaging kernels to CONTRAIL CME level flight data. Figure 5a shows the means of the averaging kernels of each of the three layers 9, 10, and 11 of all of the TANSO-FTS TIR CO$_2$ profiles used in the comparisons in summer. In Figure 5, we show examples of area 40 in the airline route between Tokyo and Honolulu, where we had a large amount of data for comparison, and area 1 in the airline route between Tokyo and Amsterdam, where we had data for comparison both in the troposphere and stratosphere. Considering the half-value width of the averaging kernels in Figure 5a, the TANSO-FTS TIR CO$_2$ retrieval results in layers 9–11 would be affected by CO$_2$ concentrations from ~400 to ~120–130 hPa. As shown in the CONTRAIL CME ascending/descending CO$_2$ profiles in Figure 5b, the variability in the CO$_2$ concentration from ~400 to ~200 hPa was relatively small in summer; the same was true in the other three seasons. This indicates that CO$_2$
concentrations below layer 9 had a small impact on the TIR CO₂ retrieval results in layers 9 and 10, which suggests that the following results do not change much, even when considering the averaging kernels related to the layers below layer 9. Consequently, we determined not to apply TIR CO₂ averaging kernels to CONTRAIL CME CO₂ data in this study. However, because we did not have CONTRAIL CME CO₂ data above ~200 hPa, we could not evaluate the impact of the CO₂ concentration above ~200 hPa on TANSO-FTS TIR CO₂ retrieval results in layers 9–11 on the basis of observation data. Thus, we should discuss again the effect of not applying averaging kernels to CONTRAIL CME data on the following comparison results in Section 6.

5.2 Results of the comparisons

6 Comparison results

6.1 Impacts of averaging kernels on CME profiles

Figure 4 shows comparisons of the differences between TANSO-FTS TIR and CONTRAIL (raw) CO₂ data, and the differences between TIR and CONTRAIL (AK) CO₂ data in low (BKK), middle (NRT and SYD), and high (DME) latitudes in layers 9, 10, and 11. In low latitudes, the differences between CONTRAIL (raw) and CONTRAIL (AK) were mostly less than 0.5 ppm in all seasons. This is because the tropopause heights there were much higher than the altitude levels of CONTRAIL CME level flight measurements, and CO₂ concentrations did not change much in the altitude regions where we compared TIR and CONTRAIL CME data. The same was true for other airports in low latitudes. While the differences between CONTRAIL (raw) and CONTRAIL (AK) were larger in middle and high latitudes than in low latitudes, they were in most cases less than 1 ppm in all seasons. In conclusion, the impact of applying the TIR CO₂ averaging kernels on CONTRAIL CME CO₂ data at around the CME level flight altitudes (~11 km) was on average less than 0.5 ppm in low latitudes and less than 1 ppm in middle and high latitudes.

6.2 Comparisons during level flight

The airline route between Tokyo and Sydney covered a wide latitude range from the northern mid-latitudes (35°N) to southern mid-latitudes (34°S). Figure 5 shows the comparisons among CONTRAIL (raw), CONTRAIL (AK) CME level flight, TANSO-FTS TIR, and a
priori CO₂ data during flights between Tokyo and Sydney in spring. In this case, we averaged CO₂ data mainly from layers 9 and 10 of the TIR retrieval layer levels. The 1-σ values of the averages show the variability of CO₂ concentrations in these UTLS layers. The average of the TIR CO₂ data agreed better with the averages of the CONTRAIL (raw) and (AK) CO₂ data than the a priori CO₂ data in all of the latitudes. The differences between CONTRAIL (raw) and CONTRAIL (AK) were approximately 0.5 ppm, which is consistent with the result shown in Figure 4, despite the fact that CONTRAIL (AK) data here were evaluated on the basis of CarbonTracker monthly-mean data. In the Southern Hemisphere, the average of the TIR CO₂ data was within 0.1% of the average averages of the CONTRAIL – (raw) and CONTRAIL (AK) CO₂ data. In the Northern Hemisphere, the average of the TIR CO₂ data agreed with the averages of the CONTRAIL (raw) and CONTRAIL (AK) CO₂ data to within 0.5%, although their agreement was slightly worse there than in the Southern Hemisphere.

Along the airline route between Tokyo and Europe, both tropospheric and stratospheric CO₂ data were obtained in the CONTRAIL CME observations. Therefore, we were able to validate the quality of TANSO-FTS TIR CO₂ data for this route both in the upper troposphere and lower stratosphere using the UTLS UTLS-CME CONTRAIL CO₂ data. Here, we averaged CO₂ data mainly from layers 9 and 10 for the upper tropospheric comparison and from layers 10 and 11 for the lower stratospheric comparison. As shown in Figure 6, the differences between CONTRAIL (raw) and CONTRAIL (AK) were again approximately 0.5 ppm when CONTRAIL CME data were divided into the upper troposphere and lower stratosphere, which is consistent with the result shown in Figure 4. Figure 6b and 6c shows that the differences between the upper tropospheric and lower stratospheric CO₂ concentrations of CONTRAIL CME data were approximately 2–3 ppm in the winter (maximum of 4.24 ppm in area 14). The upper tropospheric and lower stratospheric CO₂ concentrations from TANSO-FTS TIR V4.0/1 data also clearly differed clearly, while the upper tropospheric and lower stratospheric CO₂ concentrations from a priori data were similar. The upper tropospheric TIR CO₂ concentrations were in a fairly good agreement within 1 ppm with the corresponding CONTRAIL (raw) and CONTRAIL (AK) CME data (Figure 6b). In the lower stratosphere in winter (Figure 6c), the averages of the CONTRAIL (raw), CONTRAIL (AK) CME level flight, TANSO-FTS TIR, and a priori CO₂ data were all within 0.5–1 ppm of each other nearly identical.
Figure 7 shows the results of all of the comparisons among CONTRAIL (raw), CONTRAIL (AK) CME, TANSO-FTS TIR, and a priori CO₂ data in the upper troposphere (left) and lower stratosphere (right) for each of the six (eight) airline routes for each season. We divided the data for all four datasets in each of the 40 areas into six latitude bands: 40°S–20°S (areas 30 and 31), 20°S–0° (areas 21, 28, and 29), 0°–20°N (areas 16, 17, 20, 22, 23, 26, and 27), 20°N–40°N (areas 15, 18, 19, 24, 25, and 37–40), 40°N–60°N (areas 1, 2, 14, and 32–36), and 60°N–70°N (areas 3–13). As for the lower stratosphere, we showed the results at northern latitudes of 40°N where an adequate amount of data was obtained. The thick and dashed lines indicate the differences between CONTRAIL CME and TANSO-FTS TIR CO₂ data (TIR ave. minus CONTRAIL ave.) and the differences between CONTRAIL CME and a priori CO₂ data (a priori ave. minus CONTRAIL ave.) for each of the areas along the airline routes. All of the results with more than three data points are presented. Overall, the thick black and gray lines (TIR ave. minus CONTRAIL (raw) ave. and TIR ave. minus CONTRAIL (AK) ave.) were closer to zero than the dashed green lines (a priori ave. minus CONTRAIL (raw) ave.), which means that TIR CO₂ data agreed better with CONTRAIL CME CO₂ data than a priori CO₂ data.

The left panels of Figure 7 show that the agreements between TIR and CONTRAIL (raw) and CONTRAIL (AK) CO₂ average data were worse in spring and summer than in fall and winter in the Northern Hemisphere in the upper troposphere. The differences between TIR and CONTRAIL (raw) and CONTRAIL (AK) CO₂ data were on average within 1 ppm in fall and winter in the northern troposphere. At 0°–40°N in summer, in contrast, the TIR and a priori CO₂ average data were 2.3 ppm lower than the CONTRAIL (AK) CO₂ average data. At 20°N–40°N in spring, the differences between TIR and CONTRAIL (AK) CO₂ average data were 2.4 ppm, although the TIR CO₂ data had a better agreement with CONTRAIL CME CO₂ data than a priori CO₂ data. On the other hand, the averages of the TIR CO₂ data were within 0–0.7 ppm of the averages of the CONTRAIL (AK) CO₂ data in the Southern Hemisphere in all seasons, as in the comparison in spring shown in Figure 5.

In the lower stratosphere, the agreements between the average TANSO-FTS TIR and CONTRAIL CME CO₂ data did not have a smaller seasonality than in the upper troposphere. The averages of TIR and CONTRAIL (raw) and CONTRAIL (AK) CO₂ data agreed with each other within 0.5% in all seasons. For the airline route between Tokyo and Europe (Figure 8a), the agreement between tropospheric TANSO-FTS TIR and CONTRAIL CME CO₂
average data seemed slightly better in winter, although comparisons among seasons in the troposphere were difficult because of the lack of CONTRAIL CME data in high latitudes in the spring and summer (Figure 8a1). In the stratosphere (Figure 8a2), the averages of TIR and CONTRAIL CO$_2$ data agreed well with each other, and their differences were within $\pm 0.5$–1 ppm in the spring, summer, and winter. The differences between the two averages were slightly larger in the fall (approximately 2 ppm). For the airline route between Tokyo and Vancouver (Figure 8b), the averages of the TIR CO$_2$ data were more similar to the averages of the CONTRAIL CO$_2$ data than the a priori CO$_2$ data both in the upper troposphere and lower stratosphere in the fall and winter; the differences between the TIR and CONTRAIL CO$_2$ average data were approximately within 1 ppm. For the airline route between Tokyo and Honolulu (Figure 8c), the agreement between TIR and CONTRAIL CO$_2$ average data did not show clear seasonal differences in the lower stratosphere (Figure 8c2), because of a small number of stratospheric data. In contrast, in the upper troposphere (Figure 8c1), the differences between the two were clearly larger in the spring and summer than in the fall and winter. In particular, both the differences between TIR and CONTRAIL CO$_2$ data and between a priori and CONTRAIL CO$_2$ data were larger in spring, as was the case for the results of the comparison for the airline route between Tokyo and Vancouver (Figure 8b1).

Then, we focused on the results of the comparison between TANSO-FTS TIR and CONTRAIL CME upper tropospheric CO$_2$ data obtained in northern low and middle latitudes. Figure 8d shows that the agreement between TIR and CONTRAIL CO$_2$ average data was worse in the spring and summer than in the fall and winter for the airline route between Tokyo and Bangkok. The differences between TIR and CONTRAIL CO$_2$ data exceeded the $1\sigma$ standard deviations of the averages of TIR CO$_2$ data, and were larger than the differences between a priori and CONTRAIL CO$_2$ data at 23–34°N (area 20) in the summer. Similarly, the agreement between the averages of TIR and CONTRAIL CO$_2$ data was worse in the spring and summer than in the fall and winter for the airline route between Tokyo and East Asia (Figure 8e).

For the airline route between Tokyo and Sydney (Figure 8f), the average of the TANSO-FTS TIR CO$_2$ data was within 1 ppm of the average of the CONTRAIL CME CO$_2$ data in the Southern Hemisphere in all of the seasons, as in the comparison in the spring shown in Figure 6. However, in the Northern Hemisphere, the agreement between the two was not as strong in all of the seasons. In the comparisons in the northern summer, although the differences
between the average TIR and CONTRAIL CO₂ data were less than 1% (3 ppm), there was a relatively large negative bias in the TIR CO₂ data against the CONTRAIL CO₂ data compared to the other seasons. In the upper troposphere in northern summer, TIR CO₂ data showed a significantly good agreement with CONTRAIL CO₂ data compared to a priori CO₂ data in the Southern Hemisphere. However, in northern low and middle latitudes, TIR and a priori CO₂ data had a negative bias of up to 1% against CONTRAIL CO₂ data.

Discussion

As shown in Figure 7, TANSO-FTS TIR V1 L2 CO₂ data had a negative bias of 2.3–2.4 ppm against CONTRAIL CME CO₂ data in the northern low and middle latitudes in spring and summer. Uncertainties in surface parameters and temperature profiles could affect CO₂ retrieval in thermal infrared spectral regions. As described above, retrieving surface parameters simultaneously instead of using initial surface parameters did not affect there was no clear difference in CO₂ concentrations in the UTLS regions between the case of using initial surface parameters and the case of simultaneously retrieving surface parameters in the the TIR V1 CO₂ retrieval. We compared temperature profiles with a priori JMA GPV temperature profiles used as a priori data in the UTLS region, and did not find any difference. The differences between the two which could explain did not show any correlation with the largest TIR CO₂ negative bias in the northern low and middle latitudes in spring and summer. In the UTLS regions, temperature variability is relatively large, and therefore comprehensive validation analysis of both this suggests that the simultaneous retrieval of temperatures are unlikely a main cause of the TIR CO₂ negative bias of 2.3–2.4 ppm in the northern low and middle latitudes in the spring and summer, although the a priori and retrieved temperature profiles should be required validated using reliable and independent temperature data such as radiosonde data.

Uncertainty in a priori data could result in uncertainty in retrieved CO₂ data. Here, we arbitrarily decreased the a priori concentration by 1% in a test TIR CO₂ retrieval, and then compared the retrieved CO₂ concentrations with those retrieved using the original a priori data. In the northern low and middle latitudes in spring and summer where the DF values of TIR V1 CO₂ data were around 1.8 and more, a 1% negative bias in a priori data could yield up to a 0.7% negative bias in retrieved CO₂ concentrations in the altitude regions where we did comparisons between TIR and CONTRAIL CME data, although the magnitude of the bias
varied depending on retrievals. As shown by the green lines in Figure 7, a priori CO₂
concentrations were underestimated by 2–4 ppm in the northern low and middle latitudes in
spring and summer. The test TIR CO₂ retrieval demonstrated that the negative bias of a priori
CO₂ data against CONTRAIL CME data is a possible cause of the TIR CO₂ negative bias in
the UTLS regions in the northern low and middle latitudes in spring and summer.

In general, the information content of CO₂ observations made by TIR sensors is higher in
middle and high latitudes in spring and summer than in fall and winter because of the thermal
contrast in the atmosphere, with less seasonal dependence in low latitudes. Therefore, in
spring and summer, retrieved CO₂ data contain more measurement information and are less
constrained by a priori data at all latitudes. However, as shown in Figure 7, the retrieved TIR
CO₂ data in the northern low and middle latitudes did not sufficiently reduce the negative bias
of the a priori CO₂ data in the UTLS regions in spring and summer. This implies the existence
of factors that worsened CO₂ retrieval results other than the a priori data, especially in spring
and summer. Another possible factor that worsened CO₂ retrieval results is the uncertainty in
the calibration of TIR V161.160 L1B spectra. As reported in Kataoka et al. (2014), TANSO-
FTS TIR V130.130 L1B radiance spectra had a wavelength-dependent bias ranging from 0.1
to 2 K. Although the characteristics of the spectral bias in V161.160 L1B data used in TIR V1
L2 CO₂ retrievals are still under investigation, we assumed the same degree of bias in
V161.160 L1B spectra, and evaluated the effect of the L1B spectral bias on the TIR CO₂
retrieval using the following equation:

\[
\mathbf{d}_{CO₂} = \mathbf{G}_{CO₂} \mathbf{d}_{\text{spec}}.
\]  

Here, \( \mathbf{G}_{CO₂} \) is a gain matrix for CO₂ retrieval, \( \mathbf{d}_{\text{spec}} \) is a spectral bias vector based on the
evaluation by Kataoka et al. (2014), and \( \mathbf{d}_{CO₂} \) is a vector of bias errors in retrieved CO₂
concentrations attributable to the spectral bias. The result showed that a wavelength-
dependent bias comparable to V130.130 L1B spectra could yield up to 0.3% and 0.5%
uncertainties in retrieved CO₂ concentration in the UTLS regions in the northern middle
latitude in spring and in the northern low latitude in summer, respectively. Uncertainty in the
radiometric calibration of TANSO-FTS L1B spectra causes the spectral bias inherent in TIR
L1B spectra. The temperatures of the internal blackbody on board the TANSO-FTS
instrument partly reflect the environmental thermal conditions inside the instrument. The
temperatures of FTS-mechanics and aft-optics on the optical bench of the TANSO-FTS
instrument are precisely controlled at 23 °C. The difference in temperature between the environment inside the instrument and the optical bench could cause the uncertainty in the radiometric calibration of TANSO-FTS L1B spectra. Thus, the temperatures of the internal blackbody on board the TANSO-FTS instrument could be a parameter used to evaluate the TANSO-FTS TIR L1B spectral bias.

Figure 89 shows the averages of the partial degree of freedom of TANSO-FTS TIR V1.0 L2 CO2 data for each of the areas along the airline routes between Tokyo and Europe in the upper troposphere (a) and the lower stratosphere (b) for each season. The partial DF is defined as the diagonal element trace of a submatrix of the averaging kernels corresponding to a partial column of TIR CO2 data that were compared to CONTRAIL CME level flight data, which is equal to the averages of the 9th, 10th, or 11th diagonal element of matrix A. As shown in Figure 89, the average values of the partial DF of TIR lower stratospheric CO2 data were clearly lower than those of TIR upper tropospheric CO2 data for all of the fights between Tokyo and Europe. TIR upper tropospheric CO2 data were from layers 9 and 10, and TIR lower stratospheric CO2 data were from layers 10 and 11, as shown in Figure 34, which led to a clear difference in partial DF values between the TIR upper tropospheric and lower stratospheric CO2 data. The partial DF values of TIR upper tropospheric CO2 data were 0.13–0.20 in all of the areas for all seasons. In contrast, the partial DF values of TIR lower stratospheric CO2 data in the spring, fall, and winter were ~0.05 in almost all of the areas, although they were as high as 0.1–0.14 in the summer. From the results shown in Figure 6c and Figure 8, we conclude that TIR CO2 retrieval results in the lower stratosphere in winter were constrained to the relatively good a priori CO2 data due to the low information content, and consequently had a good agreement with CONTRAIL CME CO2 data. The comparisons in the areas during the airline route between Tokyo and Europe were included in the comparison results of 60°N–70°N in the right panels of Figure 7. In this region, the average differences between a priori and CONTRAIL (raw) data were 1-2 ppm in summer and fall, while they were less than 0.5 ppm in spring and winter. In summer, TIR CO2 retrievals had a relatively high information content compared to the other seasons, which led to an agreement between TIR and CONTRAIL (raw) and CONTRAIL (AK) CO2 data of within 0.5 ppm. In fall, TIR CO2 retrieval results in the lower stratosphere were more constrained to the a priori CO2 data, and therefore had a negative bias of approximately 1-2 ppm against CONTRAIL (raw) and CONTRAIL (AK) CO2 data. In conclusion, the quality of TIR V1 CO2 data in the lower stratosphere depends largely on the information content compared to the upper
troposphere. In the case of high latitude measurements, TIR V1 lower stratospheric CO\(_2\) data are only valid in summer.

The thick lines in Figure 8a2 show that the agreement of TIR and CONTRAIL CO\(_2\) data in the lower stratosphere was better in the spring, summer, and winter than in the fall. The dashed lines in Figure 8a2 also show that a priori CO\(_2\) data agreed better with CONTRAIL CME CO\(_2\) data in the spring and winter than in the summer and fall. From the results shown in Figure 8a2 and Figure 9, we conclude that TIR CO\(_2\) retrieval results in the lower stratosphere in the spring and winter were constrained to the relatively good a priori CO\(_2\) data due to the low information content, and consequently had a good agreement with CONTRAIL CO\(_2\) data. In the fall, TIR CO\(_2\) retrieval results in the lower stratosphere were constrained to the relatively poor a priori CO\(_2\) data, and therefore had a negative bias of approximately 2 ppm against CONTRAIL CO\(_2\) data. In the summer, TIR CO\(_2\) retrievals had a relatively high information content compared to the other seasons, which led to a good agreement between TIR and CONTRAIL CO\(_2\) data despite the relatively poor a priori CO\(_2\) data. In conclusion, the quality of TIR V1.0 CO\(_2\) data in the lower stratosphere depends largely on the information content compared to the upper troposphere. In the case of high latitude measurements, TIR V1.0 lower stratospheric CO\(_2\) data are only valid in the summer.

As shown in Figure 8d, 8e, and 8f, the agreement between TANSO-FTS TIR and CONTRAIL CME CO\(_2\) data was worse in the spring and summer than in the fall and winter in northern low and middle latitudes. At these latitudes, TIR upper tropospheric CO\(_2\) data had a negative bias of up to ~1% against CONTRAIL upper tropospheric CO\(_2\) data. This characteristic was also seen in the data from flights between Tokyo and Honolulu where there was a large amount of upper tropospheric data available in all of the seasons (Figure 8c1). Overall, as shown in Figure 8, a priori CO\(_2\) data used in TIR V1.0 CO\(_2\) retrievals had a negative bias against CONTRAIL CO\(_2\) data. The a priori CO\(_2\) data in the spring and summer in Figure 8d, 8e, and 8f clearly had a larger negative bias against the corresponding CONTRAIL CO\(_2\) data. The results show that a priori CO\(_2\) data taken from NIES-TM 05 underestimate the increase in the CO\(_2\) concentration in the upper atmosphere in spring and summer, which results in a larger negative bias of TIR V1.0 upper tropospheric CO\(_2\) data in the spring and summer than in the fall and winter in northern low and middle latitudes.
In general, information content of CO² observations made by TIR sensors is higher in middle and high latitudes in the spring and summer than in the fall and winter because of thermal contrast in the atmosphere, with less seasonal dependence in low latitudes. Therefore, in the spring and summer, retrieved CO² data contain more measurement information and are less constrained by a priori data in all latitudes. However, as shown in Figure 8, the retrieved TIR CO² data in the Northern Hemisphere did not sufficiently reduce the negative bias of the a priori CO² data in the spring and summer. The degree of improvement in the spring and summer was comparable to or worse than in the fall and winter. This implies the existence of factors that worsened CO² retrieval results other than the poor a priori data in the spring and summer. One of the possible factors is uncertainty in JMA GPV temperature profiles used in TIR V1.0 L2 CO² retrieval. If they have some seasonal bias, seasonally dependent bias in retrieved CO² data would be produced. However, the TIR V1.0 algorithm simultaneously retrieved temperature profiles other than CO², and therefore the effect of temperature uncertainty on retrieved CO² data should be reduced.

Another possible factor that worsened CO² retrieval results is uncertainty in the calibration of TIR V161.160 L1B spectra. This means that the amount of TIR V161.160 L1B spectral bias has some seasonal dependence. Therefore, we investigated an appropriate parameter to evaluate the uncertainty in TANSO-FTS TIR L1B spectra. The temperatures of the internal blackbody on board the TANSO-FTS instrument partly reflect the environmental thermal condition inside the instrument. The temperatures of FTS-mechanics and aft-optics on the optical bench of the TANSO-FTS instrument are precisely controlled at 23 degrees Celsius. The difference in temperature between the environment inside the instrument and the optical bench would cause the uncertainty in radiometric calibration of TANSO-FTS L1B spectra. Thus, the temperatures of the internal blackbody on board the TANSO-FTS instrument could be a parameter to evaluate the TANSO-FTS TIR L1B spectral bias.

Figure 10 is a scatter-plot of the average temperatures of the onboard internal blackbody and the average differences between TANSO-FTS TIR and CONTRAIL CME CO² data shown in Figure 8 for each area for each season. The average temperatures of the on-board internal blackbody were lower in the spring and summer than in the fall and winter in all of the areas. It can be seen from Figure 10 that the internal blackbody temperatures in the summer (diamonds) were lower than those in the other seasons (crosses). As discussed above, a priori CO² data had a larger negative bias against CONTRAIL CME CO² data particularly in...
northern low and middle latitudes in the spring and summer, which led to a larger negative bias in retrieved TIR CO\textsubscript{2} data at these latitudes. In addition, retrieved TIR CO\textsubscript{2} data had a larger bias in summer when the internal blackbody temperatures were lower, even if the amount of negative bias in a priori CO\textsubscript{2} data in summer was comparable to that in the other seasons, as shown in Figure 10. As stated above, the temperature of the onboard internal blackbody could be a candidate for evaluating the spectral bias. At this moment, however, there is no definite evidence of a clear correlation between the temperatures of the onboard internal blackbody and the bias in TANSO-FTS V1.0 L1B spectra. The internal blackbody temperature could be a factor in reducing the spectral bias, but it was not found in the current analysis. The correlation between the average temperatures of the onboard internal blackbody and the average differences between TIR and CONTRAIL CO\textsubscript{2} data is not very strong.

The TANSO-FTS TIR V1.0 L2 CO\textsubscript{2} algorithm simultaneously retrieves surface temperature and surface emissivity as a corrective parameter of the bias in TIR L1B spectra. Therefore, the uncertainty in these surface parameters would have a large impact on retrieved TIR CO\textsubscript{2} profiles. Figure 7d and 7e in Saitoh et al. (2009) show that the uncertainty of retrieved UTLS CO\textsubscript{2} concentrations in layers 9–11 is much less than 1% when the surface parameters have 1% uncertainty, although the uncertainty of CO\textsubscript{2} concentrations at ~400 hPa reaches 3% for the same condition. We conclude that the uncertainty of the surface parameters has a relatively small impact on the TIR UTLS CO\textsubscript{2} concentrations that were the focus of this study. The uncertainties in surface parameters and water vapor in lower atmosphere largely affect lower and middle tropospheric TIR CO\textsubscript{2} data, and therefore should be discussed when validating the quality of TIR CO\textsubscript{2} data in the lower and middle troposphere, which is beyond the scope of this paper.

We compared TANSO-FTS TIR V1.0 L2 upper-tropospheric and lower-stratospheric CO\textsubscript{2} data that were mainly from layers 9 and 10 and from layers 10 and 11 with the corresponding CONTRAIL CME tropospheric and stratospheric CO\textsubscript{2} data without applying the TIR CO\textsubscript{2} averaging kernels to the CONTRAIL CO\textsubscript{2} data. As discussed above, CO\textsubscript{2} concentrations below layer 9 have a small impact on TIR CO\textsubscript{2} retrieval results in layers 9 and 10, because the variability in the CO\textsubscript{2} concentration from ~400 to ~200 hPa was relatively small in all of the seasons. However, in layer 11, TIR CO\textsubscript{2} retrieval results could be overestimated by the effect of the CO\textsubscript{2} concentration below layer 9, if the atmosphere in layer 11 is stratospheric air with relatively low CO\textsubscript{2} concentrations. On the other hand, TIR CO\textsubscript{2} retrieval results in layers 9–11...
could also be affected by CO$_2$ concentration from ~200 to ~120–130 hPa, judging from the half-value width of the averaging kernels in Figure 5a. If the atmosphere from ~200 to ~120–130 hPa is stratospheric air with low CO$_2$ concentrations, retrieved TIR CO$_2$ concentrations in layers 9–11 could be underestimated. In summary, retrieved TIR CO$_2$ concentrations could be underestimated in layers 9–10, and face the conflicting possibility of being overestimated and/or underestimated in layer 11. However, because we did not have CO$_2$-observation data below and above the CONTRAIL CME flight levels, we cannot reach a definite conclusion.

As shown in Figure 8, TIR upper tropospheric CO$_2$ data had a slightly negative bias against CONTRAIL CME CO$_2$ data. In the comparison of the airline route of Tokyo–Sydney as shown in Figure 6, the differences between the averages of TIR CO$_2$ data and the averages of CONTRAIL CO$_2$ data were slightly larger in the Northern Hemisphere (0.5%) than in the Southern Hemisphere (0.1%). The difference between upper tropospheric and lower stratospheric CO$_2$ concentrations is larger in the Northern Hemisphere in spring (Sawa et al., 2012), which would cause a slightly larger negative bias in the Northern Hemisphere than in the Southern Hemisphere. The effect of lower CO$_2$ concentrations from ~200 to ~120–130 hPa on TIR CO$_2$ retrieval results in layers 9–10 could be one of the causes of a negative bias in retrieved CO$_2$ data other than the negative bias of a priori CO$_2$ data and the spectral bias of TIR V161.160 L1B spectra.

We investigated the differences between TIR and CONTRAIL CO$_2$ comparison results in layers 9–11 with and without applying averaging kernel functions although in limited areas over the nine airports where CO$_2$ vertical profiles were observed during ascent and descent. In the northern middle latitudes in spring (NRT in Figure 4), CONTRAIL (AK) was on average 0.2 and 1.2 ppm lower than CONTRAIL (raw) in layers 9 and 10. In contrast, the tendency was the opposite in the southern middle latitudes in spring (SYD in Figure 4); CONTRAIL (AK) was on average 1.1 and 0.4 ppm higher than CONTRAIL (raw) in layers 9 and 10. This means that CO$_2$ concentrations in layers 9 and 10 were more affected by stratospheric air with relatively low CO$_2$ concentrations in the northern middle latitude in spring, when considering averaging kernels. This is consistent with the result of Sawa et al. (2012) showing that the difference between upper tropospheric and lower stratospheric CO$_2$ concentrations was larger in the Northern Hemisphere in spring. Following the method proposed by Araki et al. (2010), we used CONTRAIL ascending/descending CO$_2$ data below a tropopause and stratospheric CO$_2$ concentrations taken from the Nonhydrostatic Icosahedral Atmospheric Model (NICAM)–Transport Model (TM) (Niwa et al., 2011) to create CO$_2$
profiles over airports. In northern middle latitudes in the spring (over NRT airport),
considering averaging kernel functions by using Expression (5) decreased a negative bias in
TIR CO$_2$ data in layers 9 and 10 by ~1 ppm. On the other hand, the same tendency was not
seen in southern middle latitudes in the spring (SYD) when considering averaging kernel
functions. This is consistent with the above discussion related to Sawa et al. (2012). In the
summer and fall in middle latitudes in both hemispheres, the effect of considering averaging
kernel functions on TIR and CONTRAIL CO$_2$ comparison results was negligible (less than
~0.5 ppm), although CONTRAIL CO$_2$ data in layers 9 and 10 with averaging kernel functions
became slightly larger there. In low latitudes (BKK, SIN, and CGK), differences between TIR
and CONTRAIL CO$_2$-comparison results in layers 9 and 10 with and without considering
averaging kernel functions were also negligible in every season. In northern high latitudes
(AMS and YVR), bias of TIR lower stratospheric CO$_2$-data against CONTRAIL CO$_2$-data in
layers 10 and 11 tended to diminish when considering averaging kernel functions, and the
effect of considering averaging kernel functions on TIR and CONTRAIL upper tropospheric
CO$_2$-comparison results in layers 9 and 10 was again negligible.

Using CONTRAIL CME level flight observations that covered wide spatial areas
allowed us to discuss the longitudinal differences in the characteristics of TIR UTLS
CO$_2$ data. In the comparison results of the airline routes of Tokyo–Europe (Figure 6) and
Tokyo–Vancouver (not shown here) shown in Figure 8a and 8b, the magnitudes of the
differences between TIR and CONTRAIL (raw) and (AK) CO$_2$ data did not have a clear
longitudinal dependence. Table 2 summarizes the latitudinal dependence of the magnitudes of
the differences between TIR and CONTRAIL (AK) CO$_2$ data were similar in every longitude
in the fall and winter in the upper troposphere and in every season in the lower stratosphere,
although there is little logic to discuss the longitudinal differences in the spring and summer
in the upper troposphere because of a small number of the data. On the other hand, in the
comparison results of Tokyo–Honolulu, differences between TIR and CONTRAIL CO$_2$-data
became larger toward ~165°W (195° in Figure 8c) in the spring. This area is located at 25°N,
and differences between TIR and CONTRAIL CO$_2$-data were also large in area 20 (in the
airline route of Tokyo–Bangkok), area 27 (Tokyo–East Asia), and area 28 (Tokyo–Sydney)
located in the same latitude region, which implies that these biases depended on latitude, not
on longitude. We conclude that the data quality of TIR V1.0 L2 UTLS CO$_2$-data does not
have a clear longitudinal dependence. Finally, we evaluated bias values of TIR V1.0 CO$_2$-data
against CONTRAIL CME CO$_2$-data for each season for each of the latitude regions: 60–70°N
(areas 3–13), 40–60°N (areas 1, 2, 14, 35, 37, 41–43), 20–40°N (areas 15, 20, 21, 27, 28, 36, 38–40), 0–20°N (areas 18, 22, 19, 25, 26, 29, 30), 0–20°S (areas 23, 31, 32), and 20–40°S (areas 33, 34). The bias values are the weighted averages of differences between TIR and CONTRAIL averaged CO₂ data of the areas located in each latitude region with considering the number of TIR CO₂ data in each of the areas. In the upper troposphere in 20–60°N, negative biases in TIR CO₂ data against CONTRAIL CME CO₂ data rangeding from 12.2 to 2.47 ppm and from 1.2 to 1.6 ppm were seen in the spring and summer, respectively, when applying averaging kernels to the assumed CME CO₂ profiles created based on CarbonTracker CT2013B monthly-mean profiles as summarized in Table 2. Although the evaluation on the basis of NICAM-TM stratospheric CO₂-concentrations in limited areas over several airports, considering averaging kernel functions decreased a negative bias in TIR CO₂ data by ~1 ppm in the spring and slightly increased a negative bias in the summer in northern middle latitudes. Thus, the following negative biases should be considered when incorporating TIR V1.0 upper tropospheric CO₂ data in inverse models which usually consider averaging kernel functions: ~2.0 ppm in both spring and summer in 20–40°N, ~1.0 ppm in spring and ~1.5 ppm in summer in 40–60°N. In northern low latitudes (0–20°N), the negative bias of 2.0 ppm should be taken into account in summer, as presented in Table 2. In the lower stratosphere in northern high latitudes, bias of TIR CO₂ data against CONTRAIL CME CO₂ data tended to diminish when considering averaging kernel functions. It is the negative biases in the northern low and middle latitudes that we should in particular mainly be concerned about when using TIR V1.0 L2 CO₂ data in any scientific analysis. In the upper troposphere in the northern middle latitudes, CO₂ concentrations reach the maximum from spring through early summer. The negative biases in TIR CO₂ data resulted in the maximum of TIR CO₂ concentrations being lower than that of the CONTRAIL CME CO₂ concentrations, which led to an underestimate of the amplitude of seasonal variation when using TIR CO₂ data without taking their negative biases into account.

78 **Summary**

In this study, we conducted a comprehensive validation of the UTLS CO₂ concentrations from the GOSAT/TANSO-FTS TIR V1.0 L2 CO₂ product. The TIR V1.0 L2 CO₂ algorithm used both the CO₂ 10 μm and 15 μm absorption bands (690–750 cm⁻¹, 790–795 cm⁻¹, 930–990 cm⁻¹, and 1040–1090 cm⁻¹), and simultaneously retrieved vertical profiles of CO₂, water
vapor, ozone, and temperature in these wavelength regions. Because the TANSO-FTS TIR V161.160 L1B radiance data used in the TIR V1.0 L2 CO2 retrieval had a spectral bias, we simultaneously derived surface temperature and surface emissivity in the same wavelength regions just as a corrective parameter, other than temperature and gas profiles, to correct the spectral bias. The simultaneous retrieval of surface temperature greatly increased the number of normally retrieved CO2 profiles.

To validate the quality of TIR V1.0 upper atmospheric CO2 data, we compared them with the level flight CO2 data of the CONTRAIL CME observations along the following airline routes in 2010: Tokyo–Europe (Amsterdam and Moscow), Tokyo–Vancouver, Tokyo–Honolulu, Tokyo–Bangkok, Tokyo–East Asia (Singapore and Jakarta), and Tokyo–Sydney. For the CONTRAIL data obtained during the northern high latitude flights, we made comparisons among CONTRAIL, TIR, and a priori CO2 data separately in the upper troposphere and in the lower stratosphere. The TIR upper tropospheric and lower stratospheric CO2 data that were compared were mainly from layers 9 and 10 (287–196 hPa) and from layers 10 and 11 (237–162 hPa), respectively. In this study, we evaluated the impact of considering TIR CO2 averaging kernel functions on CO2 concentrations using the CME profile data over the nine airports; the impact at around the CME level flight altitudes (~11 km) was on average less than 0.5 ppm in low latitudes and less than 1 ppm in middle and high latitudes.

In the Southern Hemisphere, the averages of TANSO-FTS TIR V1.1-upper atmospheric CO2 data were within 0.1% of the averages of CONTRAIL CO2 data with and without TIR CO2 averaging kernels for all of the seasons, from the limited comparisons made during flights between Tokyo and Sydney, while in the Northern Hemisphere, TIR CO2 data had a better agreement with CONTRAIL CO2 data than a priori CO2 data, with the agreement being on average within 0.5% in the Northern Hemisphere. The northern high latitude comparisons suggest that the quality of TIR lower stratospheric CO2 data depends largely on the information content. In high latitudes, TIR V1.0-lower stratospheric CO2 data are only valid in the summer when their information content is highest. Overall, the agreements of TIR and CONTRAIL CME CO2 data were worse in spring and summer than in fall and winter in the Northern Hemisphere in the upper troposphere. TIR CO2 data had a negative bias up to 2.4 ppm against CONTRAIL CO2 data with TIR CO2 averaging kernels in the northern low and middle latitudes in spring and summer. This is in the northern low and middle latitudes, the
agreement between TIR and CONTRAIL CO₂ data in the upper troposphere was worse in the spring and summer than in the fall and winter, partly because of the larger negative bias in the a priori CO₂ data in the spring and summer than in the fall and winter. The seasonal dependence of the spectral bias inherent to TANSO-FTS TIR L1B radiance data could cause a negative bias in retrieved CO₂ concentrations, particularly in summer. TIR sensors can make more observations than SWIR sensors. When using the TIR UTLS CO₂ data, the combined use of TIR UTLS CO₂ data and XCO₂ data from the SWIR bands of TANSO-FTS can be useful for studies of CO₂ surface flux inversion and atmospheric transport, provided that the seasonally and regionally dependent negative biases of the TIR V1.0 V1 L2 CO₂ data presented here should be taken into account.

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References


Ohyama, H., Kawakami, S., Shiomi, K., Morino, I., and Uchino, O.: Atmospheric Temperature and Water Vapor Retrievals from GOSAT Thermal Infrared Spectra and Initial Validation with Coincident Radiosonde Measurements, SOLA, 9, 143-147, 2013.


Saeki, T., Saito, R., Belikov, D., and Maksyutov, S.: Global high-resolution simulations of CO2 and CH4 using a NIES transport model to produce a priori concentrations for use in satellite data retrievals, Geosci. Model Dev., 6, 81-100, 2013.


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1 Strow, L. L. and Hannon, S. E.: A 4-year zonal climatology of lower tropospheric CO2
derived from ocean-only Atmospheric Infrared Sounder observations, J. Geophys. Res., 113,

2 Takagi, H. et al.: Influence of differences in current GOSAT XCO2 retrievals on surface flux

3 Yokota, T., Yoshida, Y., Eguchi, N., Ota, Y., Tanaka, T., Watanabe, H., and Maksyutov, S.:
Global Concentrations of CO2 and CH4 Retrieved from GOSAT: First Preliminary Results,

4 Yoshida, Y., Ota, Y., Eguchi, N., Kikuchi, N., Nobuta, K., Tran, H., Morino, I., and Yokota,
T.: Retrieval algorithm for CO2 and CH4 column abundances from short-wavelength infrared
spectral observations by the Greenhouse gases observing satellite, Atmos. Meas. Tech., 4,
717-734, 2011.

5 Yoshida, Y. et al.: Improvement of the retrieval algorithm for GOSAT SWIR XCO2 and
Table 1. Retrieval grid layers of GOSAT/TANSO-FTS TIR CO$_2$ V1.0V1 data.

<table>
<thead>
<tr>
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<th>Upper pressure level (hPa)</th>
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<td>10.00</td>
</tr>
<tr>
<td>26</td>
<td>10.00</td>
<td>5.62</td>
</tr>
<tr>
<td>27</td>
<td>5.62</td>
<td>1.00</td>
</tr>
<tr>
<td>28</td>
<td>1.00</td>
<td>0.10</td>
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Table 2. Bias values of GOSAT/TANSO-FTS TIR V1.0V1 CO₂ data against CONTRAIL (AK)-CME CO₂ data for each season and each latitude region in the upper troposphere and lower stratosphere in the unit of ppm. Significant bias values larger than ±21.0 ppm are indicated by boldface.

<table>
<thead>
<tr>
<th>UT</th>
<th>LS</th>
<th>MAM</th>
<th>JJA</th>
<th>SON</th>
<th>JF</th>
</tr>
</thead>
<tbody>
<tr>
<td>60–70°N</td>
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<td>-0.8</td>
<td>-0.2</td>
<td>-0.5</td>
<td>-1.0</td>
</tr>
<tr>
<td>40–60°N</td>
<td>-1.7</td>
<td>0.3</td>
<td>-1.6</td>
<td>-1.3</td>
<td>-1.1</td>
</tr>
<tr>
<td>20–40°N</td>
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<td>-2.3</td>
<td>-1.1</td>
<td>0.3</td>
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</tr>
<tr>
<td>0–20°N</td>
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<td>-2.3</td>
<td>-0.5</td>
<td>0.5</td>
<td></td>
</tr>
<tr>
<td>20°S–0</td>
<td>-0.1</td>
<td>-0.6</td>
<td>0.4</td>
<td>0.0</td>
<td></td>
</tr>
<tr>
<td>40–20°S</td>
<td>-0.2</td>
<td>-0.4</td>
<td>-0.7</td>
<td>-0.5</td>
<td></td>
</tr>
</tbody>
</table>

The evaluation of the bias values does not consider TIR CO₂ averaging kernel functions.

<table>
<thead>
<tr>
<th>UT</th>
<th>LS</th>
<th>MAM</th>
<th>JJA</th>
<th>SON</th>
<th>JF (DJF)</th>
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</thead>
<tbody>
<tr>
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<td>0.2</td>
<td>-0.3</td>
<td>-0.6</td>
</tr>
<tr>
<td>40–60°N</td>
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<td>-1.2</td>
<td>-1.2</td>
<td>-0.7</td>
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<tr>
<td>20–40°N</td>
<td>-2.7</td>
<td>-1.6</td>
<td>-0.4</td>
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<tr>
<td>0–20°N</td>
<td>-0.8</td>
<td>-2.0</td>
<td>-0.2</td>
<td>0.8</td>
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<tr>
<td>20°S–0</td>
<td>0.3</td>
<td>-0.2</td>
<td>0.5</td>
<td>0.4</td>
<td></td>
</tr>
<tr>
<td>40–20°S</td>
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<td>-0.1</td>
<td>-0.5</td>
<td>0.1</td>
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</tr>
</tbody>
</table>
Figure 1. Averaging kernel functions of GOSAT/TANSO-FTS TIR V1.0 CO₂ retrieval in the 28 retrieval grid layers shown in Table 1: (a) low latitudes in summer, (b) mid-latitudes in spring, and (c) high latitudes in winter. Solid orange, yellow, and green lines indicate averaging kernel functions of each of the three layer levels 9, 10, and 11, respectively.
Figure 2. Differences between GOSAT/TANSO-FTS TIR retrieved CO₂ profiles and the corresponding CONTRAIL CME ascending/descending data over Narita airport with considering TIR CO₂ averaging kernel functions. Thin black lines show individual comparisons. Thick gray lines and horizontal bars show the means and 1-σ standard deviations of the comparisons. The upper right number of each panel indicates the number of all of the GOSAT/TANSO-FTS TIR CO₂ profiles among the 141 pairs that were normally retrieved under each retrieval condition: (a) Ton & Eon, (b) Ton & Eoff, (c) Toff & Eon, and (d) Toff & Eoff. See the text for details of the retrieval conditions.
Figure 23. Flight tracks of all of the CONTRAIL CME observations in 2010 used in this study. A number next to a box area indicates each area number.
Figure 34. The number of GOSAT/TANSO-FTS TIR CO₂ data points compared to the CONTRAIL CME level flight data for each retrieval grid layer level for each flight. The numbers of TIR CO₂ data points in the troposphere (“T”) and stratosphere (“S”) are shown separately for the Tokyo–Europe (NRT_DME_AMS), Tokyo–Vancouver (NRT_YVR), and Tokyo–Honolulu (NRT_HNL) flight routes.
Figure 4. Scatter plots of GOSAT/TANSO-FTS TIR and CONTRAIL (raw) CO$_2$ differences and GOSAT/TANSO-FTS TIR and CONTRAIL (AK) CO$_2$ differences in layers 9, 10, and 11 for each season.

Figure 5. (Left panel) Averaging kernel function of each of the three layer levels 9, 10, and 11, shown by solid, dashed, and dotted lines, respectively. Black and gray lines show the means of averaging kernel functions of all of the GOSAT/TANSO-FTS TIR CO$_2$ profiles used in the comparisons made in areas 1 and 40 in summer (JJA), respectively. (Right panel) Mean profiles with $1\sigma$ standard deviations of CONTRAIL CME ascending/descending CO$_2$ data over Amsterdam (located in area 1) and Honolulu (located in area 40) airports in summer (JJA), shown by black and gray lines, respectively.
Figure 56. Comparisons among CONTRAIL (raw), CONTRAIL (AK), CONTRAIL CME level flight, GOSAT/TANSO-FTS TIR, and a priori (NIES TM 05) CO₂ data during flights between Tokyo and Sydney (NRT_Syd) in spring (MAM), shown by black, gray, red, and green lines, respectively. The means and their 1-σ standard deviations were calculated in each area during the flight for all three datasets.
Figure 67. Same as Figure 56, but for flights between Tokyo and Europe (NRT_DME_AMS) in winter (JF). (a) All of the data, (b) only data in the troposphere, and (c) only data in the stratosphere. See the text for the classification of tropospheric and stratospheric data.
Figure 78. Differences between GOSAT/TANSO-FTS TIR and CONTRAIL (raw) averaged CO₂ data (TIR ave. minus CONTRAIL (raw) ave.), TIR and CONTRAIL (AK) averaged CO₂ data (TIR ave. minus CONTRAIL (AK) ave.), and a priori (NIES TM 05) and CONTRAIL (raw) averaged CO₂ data (a priori ave. minus CONTRAIL (raw) ave.) for each season for each latitude band (40°S–20°S, 20°S–0°, 0°–20°N, 20°N–40°N, 40°N–60°N, 60°N–70°N), shown by black, gray, and green lines, respectively. Left and right panels show the differences in the upper troposphere and lower stratosphere, respectively. The 1-σ standard deviations of the latitudinal averages of TANSO-FTS TIR CO₂ data are shown by vertical bars. Differences between GOSAT/TANSO-FTS TIR and CONTRAIL CME averaged CO₂ data (TIR ave. minus CONTRAIL ave.) and a priori (NIES TM 05) and CONTRAIL CME averaged CO₂ data (a priori ave. minus CONTRAIL ave.) for each season and each area of all of the six flight routes, shown by thick and dashed lines, respectively: (a) Tokyo–Europe (NRT_DME_AMS), (b) Tokyo–Vancouver (NRT_YVR), (c) Tokyo–Honolulu (NRT_HNL), (d) Tokyo–Bangkok (NRT_BKK), (e) Tokyo–East Asia (NRT_SIN_CGK), and (f) Tokyo–Sydney (NRT_SYD). The means of the differences were calculated for each of the areas in spring (MAM), summer (JJA), fall (SON), and winter (JF/DJF), as shown by the pink, red, light blue, and blue lines, respectively. The 1-σ standard deviations of the averages of TANSO-FTS TIR CO₂ data are shown by vertical bars. For the airline routes of Tokyo–Europe, Tokyo–Vancouver, and Tokyo–Honolulu, the results only for the tropospheric data (a1, b1, and c1) and only for the stratospheric data (a2, b2, and c2) are shown separately. Data in December 2010 were used only in the comparisons for the flight between Tokyo and Vancouver.
Figure 89. Partial degree of freedom (DF) for GOSAT/TANSO-FTS TIR CO₂ data in the upper troposphere (a) and the lower stratosphere (b) for each area of the flight between Tokyo and Europe (NRT_DME_AMS). The means and their 1-σ standard deviations of the partial DF data were calculated in spring (MAM), summer (JJA), fall (SON), and winter (JF), as shown by the pink, red, light blue, and blue lines, respectively.
Figure 10. Correlations between the mean temperatures of the internal blackbody (BBT) on board the GOSAT/TANSO-FTS instrument and the differences between GOSAT/TANSO-FTS TIR and CONTRAIL CME averaged CO₂ data (TIR ave. minus CONTRAIL ave.) for each area of all flights for each of the four seasons. All of the data are categorized according to the differences between corresponding a priori (NIES TM 05) and CONTRAIL CME averaged CO₂ data (a priori ave. minus CONTRAIL ave.): less than 1 ppm (light blue), 1–2 ppm (green), 2–3 ppm (orange), 3–4 ppm (pink), and 4–5 ppm (red). Regression lines of the “1–2 ppm” dataset, the “2-3 ppm” dataset and all of the datasets are shown by green, orange, and black lines, respectively. The correlation coefficients of the green, orange, and black lines are -0.49, -0.56, and -0.41, respectively.
To Associate Editor and Referees,

The following attachment is the final revised manuscript.
Validation of GOSAT/TANSO-FTS TIR UTLS CO₂ data  
(Version 1) using CONTRAIL measurements

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Abstract

Thermal and Near Infrared Sensor for Carbon Observation (TANSO)–Fourier Transform Spectrometer (FTS) on board the Greenhouse Gases Observing Satellite (GOSAT) has been observing carbon dioxide (CO₂) concentrations in several atmospheric layers in the thermal infrared (TIR) band since its launch. This study compared TANSO-FTS TIR V1 CO₂ data and CO₂ data obtained in the Comprehensive Observation Network for TRace gases by AirLiner (CONTRAIL) project in the upper troposphere and lower stratosphere (UTLS), where the TIR band of TANSO-FTS is most sensitive to CO₂ concentrations, to validate the quality of the TIR V1 UTLS CO₂ data from 287 to 162 hPa. We first evaluated the impact of considering TIR CO₂ averaging kernel functions on CO₂ concentrations using CO₂ profile data obtained by the CONTRAIL Continuous CO₂ Measuring Equipment (CME), and found that the impact at around the CME level flight altitudes (~11 km) was on average less than 0.5 ppm in low latitudes and less than 1 ppm in middle and high latitudes. From a comparison made during flights between Tokyo and Sydney, the averages of the TIR upper atmospheric CO₂ data were within 0.1% of the averages of the CONTRAIL CME CO₂ data with and without TIR CO₂ averaging kernels for all seasons in the Southern Hemisphere. The results of comparisons for all of the eight airline routes showed that the agreements of TIR and CME CO₂ data were...
worse in spring and summer than in fall and winter in the Northern Hemisphere in the upper
troposphere. While the differences between TIR and CME CO₂ data were on average within 1
ppm in fall and winter, TIR CO₂ data had a negative bias up to 2.4 ppm against CME CO₂
data with TIR CO₂ averaging kernels in the northern low and middle latitudes in spring and
summer. The negative bias in the northern middle latitudes resulted in the maximum of TIR
CO₂ concentrations being lower than that of CME CO₂ concentrations, which led to an
underestimate of the amplitude of CO₂ seasonal variation.

1 Introduction

Carbon dioxide (CO₂) in the atmosphere is a well-known strong greenhouse gas (IPCC, 2013,
and references therein), with concentrations that have been observed both in situ and by
satellite sensors. Its long-term observation began in Mauna Loa, Hawaii, and the South Pole
in the late 1950s (Keeling et al., 1976a, 1976b, 1996). Since then, comprehensive CO₂
observations in the atmosphere have been conducted worldwide in several observatories and
tall towers (Bakwin et al., 1998), by aircraft flask sampling (e.g., Crevoisier et al., 2010), and
via the AirCore sampling system (Karion et al., 2010) in the framework of researches by the
National Oceanic and Atmospheric Administration (NOAA). Atmospheric CO₂
concentrations have gradually increased at a globally averaged annual rate of 1.7±0.5 ppm
from 1998 to 2011, although its growth rate has relatively large interannual variation (IPCC,
2013). Upper atmospheric CO₂ observations have been made in many areas by several
projects using commercial airliners, such as the Comprehensive Observation Network for
TRrace gases by AIrLiner (CONTRAIL) project (Machida et al., 2008) and the Civil Aircraft
for the Regular Investigation of the atmosphere Based on an Instrument Container
(CARIBIC) project (Brenninkmeijer et a., 2007). Continuous long-term measurements of CO₂
made by several airplanes of Japan Airlines (JAL) in the CONTRAIL project have revealed
details of its seasonal variation and interhemispheric transport in the upper atmosphere (Sawa
et al., 2012) and interannual and long-term trends of its latitudinal gradients (Matsueda et al.,
2015).

Atmospheric CO₂ observations by satellite sensors are categorized into two types: those
utilizing CO₂ absorption bands in the shortwave infrared (SWIR) regions at around 1.6 and
2.0 μm, and those in the thermal infrared (TIR) regions at around 4.6, 10, and 15 μm. The
Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY)
on the Environmental Satellite (ENVISAT) first observed CO₂ column-averaged dry-air mole
fractions (XCO₂) from spectra at 1.57 μm (Buchwitz et al., 2005; Barkley et al., 2006). The
Thermal and Near Infrared Sensor for Carbon Observation (TANSO–Fourier Transform
Spectrometer (FTS) on board the Greenhouse Gases Observing Satellite (GOSAT), which
was launched in 2009 (Yokota et al., 2009), has observed XCO₂ with high precision by
utilizing the 1.6 and/or 2.0 μm CO₂ absorption bands (Yoshida et al., 2011, 2013; O’Dell et
al., 2012; Butz et al., 2011; Cogan et al., 2012). The Orbiting Carbon Observatory 2 (OCO-2)
was successfully launched in 2014, and started regular observations of XCO₂ with high spatial
resolution. Satellite CO₂ observations at TIR absorption bands have a longer history
beginning with the High-Resolution Infrared Sounder (HIRS) (Chédin et al., 2002, 2003,
2005). The Atmospheric Infrared Sounder (AIRS) has achieved more accurate observations of
middle and upper tropospheric CO₂ concentrations (Crevoisier et al., 2004; Chahine et al.,
2005; Maddy et al., 2008; Strow and Hannon, 2008). The Tropospheric Emission
Spectrometer (TES) has observed CO₂ concentrations in several vertical layers with high
accuracy by taking advantage of its high wavelength resolution (Kulawik et al., 2010, 2013).
The Infrared Atmospheric Sounding Interferometer (IASI) has observed upper atmospheric
CO₂ amounts from its TIR spectra (Crevoisier et al., 2009). TANSO-FTS also has a TIR band
in addition to its three SWIR bands, and obtains vertical information of CO₂ concentrations in
addition to XCO₂ in the same field of view (Saitoh et al., 2009).

Rayner and O’Brien (2001) and Pak and Prather (2001) showed the utility of global CO₂ data
obtained by satellite sensors for estimating its source and sink strength, and many studies of
CO₂ inversion have been conducted using a huge amount of satellite data since the 2000s.
Chevallier et al. (2005) first used satellite CO₂ data, observed with the Operational Vertical
Sounder (TOVS), to estimate CO₂ surface fluxes. They reported that a regional bias in
satellite CO₂ data hampers the outcomes. Nassar et al. (2011) demonstrated that the wide
spatial coverage of satellite CO₂ data is beneficial to CO₂ surface flux inversion through the
combined use of TES and surface flask CO₂ data, particularly in regions where surface
measurements are sparse. In addition to CO₂ surface inversion results using TIR observations,
global XCO₂ data observed with the SWIR bands of TANSO-FTS have been actively used for
estimating CO₂ source and sink strength (Maksyutov et al., 2013; Saeki et al., 2013;
Chevallier et al., 2014; Basu et al., 2013, 2014; Takagi et al., 2014). One of the important
things to consider when incorporating satellite data in CO₂ inversion is the accuracy of the
data, as suggested by Basu et al. (2013). Uncertainties in satellite CO₂ data should be assessed
seasonally and regionally to determine the seasonal and regional characteristics of the satellite CO2 bias.

The importance of upper atmospheric CO2 data in the inversion analysis of CO2 surface fluxes was discussed in Niwa et al. (2012). They used CONTRAIL CO2 data in conjunction with surface CO2 data to estimate surface flux, and demonstrated that adding middle and upper tropospheric data observed by the aircraft could greatly reduce the posteriori flux errors, particularly in tropical Asian regions. Middle and upper tropospheric and lower stratospheric CO2 concentrations and column amounts of CO2 can be simultaneously observed in the same field of view with TANSO-FTS on board GOSAT. Provided that the quality of upper atmospheric CO2 data simultaneously obtained with TANSO-FTS is proven to be comparable to that of TANSO-FTS XCO2 data (Yoshida et al., 2013; Inoue et al., 2013), the combined use of upper atmospheric CO2 and XCO2 data observed with TANSO-FTS could be a useful tool for estimating CO2 surface flux.

GOSAT, which is the first satellite to be dedicated to greenhouse gas monitoring, was launched on January 23, 2009. As described above, TANSO-FTS on board GOSAT has been observing CO2 concentrations in several vertical layers in the TIR band. In this study, we focused on CO2 concentrations in the upper troposphere and lower stratosphere (UTLS), where the TIR band of TANSO-FTS is most sensitive. We validated these data by comparison with upper atmospheric CO2 data obtained in a wide spatial coverage in the CONTRAIL project. Sections 2 and 3 explain the GOSAT and CONTRAIL measurements, respectively. Section 4 details the retrieval algorithm used in the latest version 1 (V1) CO2 level 2 (L2) product of the TIR band of TANSO-FTS. Section 5 describes the methods of comparing TANSO-FTS TIR V1 L2 and CONTRAIL CO2 data. Sections 6 and 7 show and discuss the results of the comparisons between TIR and CONTRAIL CO2 data. Section 8 summarizes this study.

2 GOSAT observations

GOSAT is a joint satellite project of the National Institute for Environmental Studies (NIES), Ministry of the Environment (MOE), and Japan Aerospace Exploration Agency (JAXA) for the purpose of making global observations of greenhouse gases such as CO2 and CH4 (Hamazaki et al., 2005; Yokota et al., 2009). It was launched on January 23, 2009, from the Tanegashima Space Center, and has continued its observations for more than six years.
GOSAT is equipped with the TANSO-FTS for greenhouse gas monitoring and the TANSO-Cloud and Aerosol Imager (CAI) to detect clouds and aerosols in the TANSO-FTS field of view (Kuze et al., 2009). TANSO-FTS consists of three bands in the SWIR region and one band in the TIR region. Column amounts of greenhouse gases are observed in the SWIR bands and vertical information of gas concentrations are obtained in the TIR band (Yoshida et al., 2011, 2013; Saitoh et al., 2009, 2012; Ohyama et al., 2012, 2013).

Kuze et al. (2012) provided a detailed description of the methods used for the processing and calibration of level 1B (L1B) spectral data from TANSO-FTS. They explained the algorithm for the version 150.151 (V150.151) L1B spectral data. The TIR V1 L2 CO₂ product we focused on in this study was created from a later version, V161.160, of L1B spectral data. The following modifications were made to the algorithm from V150.151 to V161.160: improving the TIR radiometric calibration through the improvement of calibration parameters, turning off the sampling interval non-uniformity correction, modifying the spike noise criteria of the quality flag, and reevaluating the misalignment between the GOSAT satellite and TANSO-FTS sensor. Kataoka et al. (2014) reported that the biases of TANSO-FTS TIR V130.130 L1B radiance spectra based on comparisons with the Scanning High-resolution Interferometer Sounder (S-HIS) spectra for warm scenes were 0.5 K at 800–900 cm⁻¹ and 700–750 cm⁻¹, 0.1 K at 980–1080 cm⁻¹, and more than 2 K at 650–700 cm⁻¹. Although the magnitude of the spectral bias evaluated on the basis of V130.130 L1B data would change in V161.160 L1B data, the issue of L1B spectral bias still remains. The spectral bias inherent in TIR L1B spectra would be mainly because of uncertainty of polarization correction. Another possible cause was discussed in Imasu et al. (2010). When retrieving CO₂ concentrations from the TIR band of TANSO-FTS, the spectral bias that is predominant in CO₂ absorption bands should be considered (Ohyama et al., 2013).

3 CONTRAIL Continuous Measurement Equipment (CME) observations

We used CO₂ data obtained in the CONTRAIL project to validate the quality of TANSO-FTS TIR V1 L2 CO₂ data. CONTRAIL is a project to observe atmospheric trace gases such as CO₂ and CH₄ using instruments installed on commercial aircraft operated by JAL. Observations of trace gases in this project began in 2005. Two types of measurement instruments, the Automatic Air Sampling Equipment (ASE) and the Continuous CO₂
Measuring Equipment (CME), have been installed on several JAL aircraft to measure trace gases over a wide area (Machida et al., 2008).

This study used CO₂ data obtained with CME on several airline routes from Narita Airport, Japan. CO₂ observations with CME use a LI-COR LI-840 instrument that utilizes a nondispersive infrared absorption (NDIR) method (Machida et al., 2008). In the observations, two different standard gases, with CO₂ concentration of 340 ppm and 390 ppm based on NIES09 scale, are regularly introduced into the NDIR for calibration. The accuracy of CME CO₂ measurements is 0.2 ppm. See Machida et al. (2008), Matsueda et al. (2008), and Machida et al. (2011) for details of the CME CO₂ observations and their accuracy and precision.

4 Retrieval algorithm of TANSO-FTS TIR V1 CO₂ data

4.1 Basic retrieval settings

Saitoh et al. (2009) provided an algorithm for retrieving CO₂ concentrations from the TIR band of TANSO-FTS. The first version, V00.01, of the L2 CO₂ product of the TIR band of TANSO-FTS was basically processed by the algorithm described in Saitoh et al. (2009). The V1 L2 CO₂ product that we focused on in this study also adopted a non-linear maximum a posteriori (MAP) method with linear mapping, as was the case for the V00.01 product. We utilized the following expressions in TIR CO₂ retrieval:

\[
\hat{z}_{i+1} = W^* x_a + G[y - F(\hat{x}_i) + K_i W(W^* \hat{x}_i - W^* x_a)]
\]

\[
G = [W^T K_i^T S_e^{-1} K_i W + (W^* S_a W^*)^{-1}]^{-1} W^T K_i^T S_e^{-1}
\]

where \(x_a\) is an a priori vector, \(S_a\) is a covariance matrix of the a priori vector, \(S_e\) is a covariance matrix of measurement noise, \(K_i\) is a CO₂ Jacobian matrix calculated using the \(i^{th}\) retrieval vector \(\hat{x}_i\) on full grids, \(F(\hat{x}_i)\) is a forward spectrum vector based on \(\hat{x}_i\), \(y\) is a measurement spectrum vector, and \(\hat{z}_{i+1}\) is the \(i+1^{th}\) retrieval vector defined on retrieval grids. \(W\) is a matrix that interpolates from retrieval grids onto full grids. \(W^*\) is the generalized inverse matrix of \(W\).

The full grids are vertical layer grids for radiative transfer calculation, and the retrieval grids are defined as a subset of the full grids. In the V1 L2 CO₂ retrieval algorithm, linear mapping
between retrieval grids and full grids was also applied, but the number of full grid levels was 78 instead of 110 in the V00.01 algorithm. The determination of retrieval grids in the V1 algorithm basically followed the method of the V00.01 algorithm. It was based on the areas of a CO$_2$ averaging kernel matrix in the tropics, but the retrieval grid levels were fixed for all of the retrieval processing, as presented in Table 1. Averaging kernel matrix $A$ is defined (Rodgers, 2000) as

$$A = GKW.$$  \hspace{1cm} (2)

Figure 1 shows typical averaging kernel functions of TIR V1 L2 CO$_2$ retrieval. The degrees of freedom (DF) in these cases (trace of the matrix $A$) were (a) 2.22, (b) 1.81, and (c) 1.36, respectively. The seasonally averaged DF values of TIR V1 CO$_2$ data ranged from 1.12 to 2.35. In the low and middle latitudes between 35°N and 35°S, the CO$_2$ DF values were around 2.0 or more; this means that observations by the TIR band of TANSO-FTS can provide information on CO$_2$ concentrations in more than two vertical layers, one of which we focused on in this study.

A priori and initial values for CO$_2$ concentrations were taken from the outputs of the NIES transport model (NIES-TM05) (Saeki et al., 2013). A priori and initial values for temperature and water vapor were obtained from Japan Meteorological Agency (JMA) Grid Point Value (GPV) data. Basically, the retrieval processing of TANSO-FTS was only conducted under clear-sky conditions, which was judged based on a cloud flag from TANSO-CAI in the daytime (Ishida and Nakajima, 2009; Ishida et al., 2011) and on a TANSO-FTS TIR spectrum in the nighttime.

4.2 Improvements in the TIR V1 CO$_2$ algorithm

The following conditions are the improvements made in the TANSO-FTS TIR V1 L2 CO$_2$ algorithm from the V00.01 algorithm. The V1 algorithm used the CO$_2$ 10 μm absorption band in addition to the CO$_2$ absorption band at around 15 μm band; the wavelength regions of 690–750 cm$^{-1}$, 790–795 cm$^{-1}$, 930–990 cm$^{-1}$, and 1040–1090 cm$^{-1}$ were used in the CO$_2$ retrieval. We did not apply any channel selection. In these wavelength regions, temperature, water vapor, and ozone concentrations were retrieved simultaneously with CO$_2$ concentration. Moreover, surface temperature and surface emissivity were simultaneously derived as a
correction parameter of the spectral bias inherent in TANSO-FTS TIR V161.160 L1B spectra at the above-mentioned CO₂ absorption bands. We assumed that the spectral bias could be divided into two components: a wavelength-dependent bias whose amount varied depending on wavelength and a wavelength-independent bias whose amount was uniform in a certain wavelength region. We tried to correct such a wavelength-independent component of the spectral bias by adjusting the value of surface temperature. Similarly, a wavelength-dependent component of the spectral bias was corrected by adjusting the value of surface emissivity in each wavelength channel. Therefore the matrices of \( K \) and \( S_a \) of expression (1) are as follows:

\[
K = (K_{CO2} K_{H2O} K_{O3} K_T k_{ST_1} k_{ST_2} k_{ST_3} k_{ST_4} k_{ST_5} k_{SE_1} k_{SE_2} k_{SE_3} k_{SE_4} k_{SE_5}), \tag{3}
\]

\[
S_a = \begin{bmatrix}
S_{CO2} \\
S_{H2O} \\
S_{O3} \\
S_T \\
S_{ST_1} \\
S_{ST_2} \\
S_{ST_3} \\
S_{ST_4} \\
S_{ST_5} \\
S_{SE_1} \\
S_{SE_2} \\
S_{SE_3} \\
S_{SE_4} \\
S_{SE_5}
\end{bmatrix}, \tag{4}
\]

where \( K_{CO2}, K_{H2O}, K_{O3}, \) and \( K_T \) are Jacobian matrices of CO₂, water vapor, ozone, and temperature on full grids, respectively, and \( S_{CO2}, S_{H2O}, S_{O3}, \) and \( S_T \) are a priori covariance matrices of CO₂, water vapor, ozone, and temperature on full grids, respectively. The vectors \( k_{ST_1}, k_{ST_2}, k_{ST_3}, k_{ST_4}, \) and \( k_{ST_5} \) are the Jacobian vectors of surface temperature in the wavelength regions of 690–715 cm⁻¹, 715–750 cm⁻¹, 790–795 cm⁻¹, 930–990 cm⁻¹, and 1040–1090 cm⁻¹, respectively. The vectors \( k_{SE_1}, k_{SE_2}, k_{SE_3}, k_{SE_4}, \) and \( k_{SE_5} \) are the Jacobian vectors of surface emissivity in each of the five wavelength regions, respectively. The elements of the Jacobian vectors of surface parameters that were defined for each of the five wavelength regions were set to be zero in the other wavelength regions. The values \( S_{ST_1}, \)
S\textsubscript{S\textsc{T}_2}, S\textsubscript{S\textsc{T}_3}, S\textsubscript{S\textsc{T}_4}, and S\textsubscript{S\textsc{T}_5} and S\textsubscript{S\textsc{E}_1}, S\textsubscript{S\textsc{E}_2}, S\textsubscript{S\textsc{E}_3}, S\textsubscript{S\textsc{E}_4}, and S\textsubscript{S\textsc{E}_5} are a priori variances of surface temperature and surface emissivity in each of the five wavelength regions, respectively. Simultaneous retrieval of the surface parameters in the V1 algorithm was conducted just for the purpose of correcting the TIR V161.160 L1B spectral bias; it had no physical meaning. We estimated the surface parameters separately in each of the five wavelength regions to consider differences in the amount of spectral bias in each wavelength region. The matrices $S_\text{a}$ for CO$_2$, temperature, water vapor, and ozone were diagonal matrices with vertically fixed diagonal elements with a standard deviation of 2.5%, 3 K, 20%, and 30%, respectively. Here, a priori and initial values for ozone were obtained from the climatological data for each latitude bin for each month given by MacPeters et al. (2007). We assumed rather large values as a priori variances of the surface parameters (a standard deviation of 10 K for surface temperature), which could allow more flexibility in the L1B spectral bias correction by the surface parameters. The a priori and initial values for surface emissivity were calculated by linear regression analysis using the Advanced Space-borne Thermal Emission Reflection Radiometer (ASTER) Spectral Library (Baldridge et al., 2009) using land-cover classification, vegetation, and wind speed information. The a priori and initial values for surface temperature were estimated using radiance data in several channels around 900 cm$^{-1}$ of the TIR V161.160 L1B spectra.

In the TIR V1 L2 algorithm, we estimated surface temperature and surface emissivity to correct the spectral bias inherent in the TANSO-FTS TIR L1B spectra (Kataoka et al., 2014). The existence of a relatively large spectral bias around the CO$_2$ 15 µm absorption band in TANSO-FTS TIR L1B spectra (Kataoka et al., 2014) resulted in a decrease in the number of normally retrieved CO$_2$ profiles. This is probably because the TIR L1B spectral bias in the CO$_2$ 15 µm absorption band was sometimes too large for the L2 retrieval calculation to converge in a limited iteration. The correction of the TIR L1B spectral bias through the simultaneous retrieval of the surface parameters did not affect retrieved CO$_2$ concentrations in the UTLS regions, which was the focus of this study, but it altered the number of normally retrieved CO$_2$ profiles. The correction of the TIR L1B spectral bias through the simultaneous retrieval of surface temperature increased the number of normally retrieved CO$_2$ profiles. This implies that a wavelength-independent component of the spectral bias in CO$_2$ absorption bands could be reduced by adjusting the value of surface temperature at the bands. In contrast, the spectral bias correction through the simultaneous retrieval of surface emissivity did not increase the number of normally retrieved CO$_2$ profiles. If the TIR L1B spectral bias has a
wavelength dependence, surface emissivity could be effective for correcting such a wavelength-dependent bias. A more effective method of L1B spectral bias correction based on surface emissivity should be considered in the next version of the TIR L2 CO$_2$ retrieval algorithm, if a future version of the TIR L1B spectral data still has a bias.

5 Comparison methods

5.1 Area comparisons

Here, we used the level flight CO$_2$ data of CONTRAIL CME observations in 2010 to validate the quality of UTLS CO$_2$ data from the TANSO-FTS TIR V1 L2 CO$_2$ product. The level flight data obtained from the following eight airline routes of the CONTRAIL CME observations were used in this study: Tokyo–Amsterdam (NRT–AMS) and Tokyo–Moscow (NRT–DME), Tokyo–Vancouver (NRT–VYR), Tokyo–Honolulu (NRT–HNL), Tokyo–Bangkok (NRT–BKK), Tokyo–Singapore (NRT–SIN) and Tokyo–Jakarta (NRT–CGK), and Tokyo–Sydney (NRT–SYD). We merged the level flight data of Tokyo–Amsterdam and Tokyo–Moscow into “Tokyo–Europe”, and the data of Tokyo–Singapore and Tokyo–Jakarta into “Tokyo–East Asia”. Figure 2 shows the flight tracks of all of the CONTRAIL CME observations in 2010 used in this study. As shown in the figure, we divided the CONTRAIL CME level flight data into 40 areas following Niwa et al. (2012), and compared them with TANSO-FTS TIR CO$_2$ data in each area in each season. The amount of level flight data varied depending on the area and season. The largest amount of data was obtained in area 15 over Narita Airport, where 4,694–9,306 data points were obtained. A relatively small amount of level flight data, 79–222 data points, was obtained in area 1 over Amsterdam. In all 40 areas, we collected sufficient level flight data to undertake comparison analysis based on the average values, except for seasons and regions with no flights.

5.2 Comparisons of CME profiles with and without averaging kernels

In comparisons of TIR V1 L2 CO$_2$ data with the CONTRAIL CME level flight data, it is difficult to smooth the CME data by applying TIR CO$_2$ averaging kernels, because CO$_2$ concentrations below and above the CME flight levels were not observed. Here, we evaluated the impact of considering averaging kernel functions on CO$_2$ concentrations using the CME profile data. We regarded the CME data obtained during the ascent and descent flights over
the nine airports as part of CO₂ vertical profiles, and investigated differences between TIR and CME CO₂ data with and without applying averaging kernel functions in the altitude regions around the CME level flight observations. We assumed the CME ascending/descending CO₂ concentration at the uppermost altitude level to be constant up to the tropopause height, following the method proposed by Araki et al. (2010). We used stratospheric CO₂ data taken from the Nonhydrostatic Icosahedral Atmospheric Model (NICAM)–Transport Model (TM) (Niwa et al., 2011; 2012) to create whole CO₂ vertical profiles over the airports. The NICAM-TM CO₂ data used here introduced CONTRAIL CO₂ data to the inverse model in addition to surface CO₂ data, and therefore could simulate upper atmospheric CO₂ concentrations well (Niwa et al., 2012). We determined the stratospheric CO₂ profile by assuming the CO₂ concentration gradients, calculated on the basis of the NICAM-TM CO₂ data above the tropopause height.

To compare these CME CO₂ profiles with TIR CO₂ data, we calculated a weighted average of all the CME CO₂ data included in each of the 28 retrieval grid layers with respect to altitude, and defined the CO₂ data in the 28 layers as “CONTRAIL (raw)” data. Then, we selected TIR CO₂ data that coincided with each of the CONTRAIL (raw) profiles. The criteria for the coincident pairs were a 300 km distance from Narita airport, and a 3-day difference of each other observation. We applied TIR CO₂ averaging kernel functions to the corresponding CONTRAIL (raw) profile, as follows (Rodgers and Connor, 2003):

\[ x_{\text{CONTRAIL(AK)}} = x_{\text{a priori}} + A(x_{\text{CONTRAIL(raw)}} - x_{\text{a priori}}). \] (5)

Here, \( x_{\text{CONTRAIL(raw)}} \) and \( x_{\text{a priori}} \) are CONTRAIL (raw) and a priori CO₂ profiles. We defined the CONTRAIL (raw) data with TIR CO₂ averaging kernel functions as “CONTRAIL (AK)” data.

### 5.3 Level flight comparisons

In this study, we made comparisons between TIR and CONTRAIL CME level flight CO₂ data in two ways. The first was a direct comparison with original CME CO₂ data, i.e., CONTRAIL (raw) data. The second was a comparison with CONTRAIL (AK) data in the altitude regions around the CME level flight observations that were based on “assumed CO₂ profiles” created at each of the measurement locations of all the CME level flight data. In the first comparison with CONTRAIL (raw) data, the CME level flight data in each of the 40 areas were averaged for each season (MAM, JJA, SON, and JF/DJF). The average altitude of all of the CME level
flight data used here was 11.245 km. The airline routes of Tokyo–Europe, Tokyo–Vancouver, and Tokyo–Honolulu contained both tropospheric and stratospheric data in the areas along their routes; therefore, we calculated the average and standard deviation values separately. Here, we differentiated between the tropospheric and stratospheric level flight data on the basis of temperature lapse rates from the JMA GPV data that were interpolated to the CONTRAIL CME measurement locations. The average altitudes of the tropospheric and stratospheric level flight data from the airline route between Tokyo and Europe were 10.84 km and 11.18 km, respectively.

In the comparison with CONTRAIL (raw) data, we selected TANSO-FTS TIR V1 L2 CO$_2$ data that were in the altitude range within ±1 km of the average altitude of the CME level flight data for each area for each season, and calculated their averages and standard deviations. Similarly, we calculated the averages and standard deviations of the corresponding a priori CO$_2$ data for each area for each season. For the airline routes of Tokyo–Europe, Tokyo–Vancouver, and Tokyo–Honolulu, the averages and standard deviations of TIR V1 CO$_2$ data and the corresponding a priori CO$_2$ data were calculated separately for the tropospheric and stratospheric data. In this calculation, we first selected TIR V1 CO$_2$ data that were collected in a range within ±1 km of the average altitudes of the CONTRAIL tropospheric and stratospheric CO$_2$ data for each area. Then, we classified each of the selected TIR CO$_2$ data points into tropospheric and stratospheric data on the basis of the temperature lapse rates from the JMA GPV data that were interpolated to the TANSO-FTS measurement locations, and calculated the seasonal averages and standard deviations for the reselected tropospheric and stratospheric TIR CO$_2$ data. This procedure was required for two reasons: (1) tropopause height at each TANSO-FTS measurement location should differ on a daily basis, and (2) because TIR CO$_2$ data were selected within the range of 2 km, some tropospheric TIR CO$_2$ data were selected on the basis of the CONTRAIL stratospheric level flight data, and vice versa. Figure 3 shows the number of TANSO-FTS TIR CO$_2$ data points that were finally selected in each retrieval layer for each of the airline routes. The TIR CO$_2$ data used in the comparative analysis were mainly from layers 9 and 10 (from 287 to 196 hPa) for the tropospheric comparison and from layers 10 and 11 (from 237 to 162 hPa) for the stratospheric comparison.

In the second comparison, we assumed a CO$_2$ vertical profile on the basis of CONTRAIL (raw) data at each of the CONTRAIL CME level flight locations, and applied TIR CO$_2$
averaging kernel functions to the assumed profiles. For this purpose, realistic CO$_2$ vertical profiles were required along the eight airline routes. In this study, we created a CO$_2$ profile at each CME level flight measurement location from CarbonTracker CT2013B monthly-mean CO$_2$ data (Peters et al., 2007). The CarbonTracker CT2013B CO$_2$ data are available to the public, and therefore readers can refer to the dataset that we used as a CO$_2$ climatological dataset. The method for creating a CO$_2$ vertical profile from the CONTRAIL (raw) and CarbonTracker CT2013B data is as follows. We first averaged all of the CarbonTracker CT2013B monthly-mean data included in each of the 40 areas to create area-averaged CarbonTracker CT2013B profiles. Then, we shifted the area-averaged CarbonTracker CT2013B profile so that its concentration fit to each of the CONTRAIL (raw) data at CME level flight altitude. Finally, we applied area-averaged TIR CO$_2$ averaging kernel functions to each of the shifted area-averaged CO$_2$ profiles, and created profiles of CONTRAIL (AK) at all the CME level flight measurement locations.

We compared the CONTRAIL (AK) data with TIR CO$_2$ data at the altitude regions around the CME level flight observations for each area in each season. We extracted CONTRAIL (AK) data that corresponded to the TIR retrieval layers where TIR CO$_2$ data were compared to CONTRAIL (raw) data, and averaged them for each area for each season. For the airline routes of Tokyo–Europe, Tokyo–Vancouver, and Tokyo–Honolulu, we separately averaged CONTRAIL (AK) data created from tropospheric and stratospheric CONTRAIL (raw) data, and defined the averages as tropospheric and stratospheric CONTRAIL (AK) data, respectively. As shown in Figure 3, the CONTRAIL (AK) data used for the comparison during flights between Tokyo and Sydney consisted of CO$_2$ concentrations in layers 9 and 10 of the CONTRAIL (AK) profiles. For the flights between Tokyo and Europe, the CONTRAIL (AK) data used for the tropospheric and stratospheric comparisons were based on CO$_2$ concentrations in layers 9 and 10 and in layers 10 and 11 of CONTRAIL (AK) profiles, respectively.

6 Comparison results

6.1 Impacts of averaging kernels on CME profiles

Figure 4 shows comparisons of the differences between TANSO-FTS TIR and CONTRAIL (raw) CO$_2$ data, and the differences between TIR and CONTRAIL (AK) CO$_2$ data in low
(BKK), middle (NRT and SYD), and high (DME) latitudes in layers 9, 10, and 11. In low latitudes, the differences between CONTRAIL (raw) and CONTRAIL (AK) were mostly less than 0.5 ppm in all seasons. This is because the tropopause heights there were much higher than the altitude levels of CONTRAIL CME level flight measurements, and CO$_2$ concentrations did not change much in the altitude regions where we compared TIR and CONTRAIL CME data. The same was true for other airports in low latitudes. While the differences between CONTRAIL (raw) and CONTRAIL (AK) were larger in middle and high latitudes than in low latitudes, they were in most cases less than 1 ppm in all seasons. In conclusion, the impact of applying the TIR CO$_2$ averaging kernels on CONTRAIL CME CO$_2$ data at around the CME level flight altitudes (~11 km) was on average less than 0.5 ppm in low latitudes and less than 1 ppm in middle and high latitudes.

6.2 Comparisons during level flight

The airline route between Tokyo and Sydney covered a wide latitude range from the northern mid-latitudes (35°N) to southern mid-latitudes (34°S). Figure 5 shows comparisons among CONTRAIL (raw), CONTRAIL (AK), TANSO-FTS TIR, and a priori CO$_2$ data during flights between Tokyo and Sydney in spring. In this case, we averaged CO$_2$ data mainly from layers 9 and 10 of the TIR retrieval layer levels. The 1-σ values of the averages show the variability of CO$_2$ concentrations in these UTLS layers. The average of the TIR CO$_2$ data agreed better with the averages of the CONTRAIL (raw) and (AK) CO$_2$ data than the a priori CO$_2$ data in all latitudes. The differences between CONTRAIL (raw) and CONTRAIL (AK) were approximately 0.5 ppm, which is consistent with the result shown in Figure 4, despite the fact that CONTRAIL (AK) data here were evaluated on the basis of CarbonTracker monthly-mean data. In the Southern Hemisphere, the average of the TIR CO$_2$ data was within 0.1% of the averages of the CONTRAIL (raw) and CONTRAIL (AK) CO$_2$ data. In the Northern Hemisphere, the average of the TIR CO$_2$ data agreed with the averages of the CONTRAIL (raw) and CONTRAIL (AK) CO$_2$ data to within 0.5%, although the agreement was slightly worse there than in the Southern Hemisphere.

Along the airline route between Tokyo and Europe, both tropospheric and stratospheric CO$_2$ data were obtained in the CONTRAIL CME observations. Therefore, we were able to validate the quality of TANSO-FTS TIR CO$_2$ data for this route both in the upper troposphere and lower stratosphere using the UTLS CME CO$_2$ data. Here, we averaged CO$_2$ data mainly from layers 9 and 10 for the upper tropospheric comparison and from layers 10 and 11 for the
lower stratospheric comparison. As shown in Figure 6, the differences between CONTRAIL (raw) and CONTRAIL (AK) were again approximately 0.5 ppm when CONTRAIL CME data were divided into the upper troposphere and lower stratosphere, which is consistent with the result shown in Figure 4. Figure 6b and 6c shows that the differences between the upper tropospheric and lower stratospheric CO$_2$ concentrations of CONTRAIL CME data were approximately 2–3 ppm in winter (maximum of 4.24 ppm in area 14). The upper tropospheric and lower stratospheric CO$_2$ concentrations from TANSO-FTS TIR V1 data also clearly differed, while the upper tropospheric and lower stratospheric CO$_2$ concentrations from a priori data were similar. The upper tropospheric TIR CO$_2$ concentrations were in a good agreement within 1 ppm with the corresponding CONTRAIL (raw) and CONTRAIL (AK) data (Figure 6b). In the lower stratosphere in winter (Figure 6c), the averages of the CONTRAIL (raw), CONTRAIL (AK), TANSO-FTS TIR, and a priori CO$_2$ data were all within 0.5–1 ppm of each other.

Figure 7 shows the results of all of the comparisons among CONTRAIL (raw), CONTRAIL (AK), TANSO-FTS TIR, and a priori CO$_2$ data in the upper troposphere (left) and lower stratosphere (right) for each season. We divided the data for all four datasets in each of the 40 areas into six latitude bands: 40°S–20°S (areas 30 and 31), 20°S–0° (areas 21, 28, and 29), 0°–20°N (areas 16, 17, 20, 22, 23, 26, and 27), 20°N–40°N (areas 15, 18, 19, 24, 25, and 37–40), 40°N–60°N (areas 1, 2, 14, and 32–36), and 60°N–70°N (areas 1–13). As for the lower stratosphere, we showed the results at northern latitudes of 40°N where an adequate amount of data was obtained. Overall, the black and gray lines (TIR ave. minus CONTRAIL (raw) ave. and TIR ave. minus CONTRAIL (AK) ave.) were closer to zero than the green lines (a priori ave. minus CONTRAIL (raw) ave.), which means that TIR CO$_2$ data agreed better with CONTRAIL CME CO$_2$ data than a priori CO$_2$ data.

The left panels of Figure 7 show that the agreements between TIR and CONTRAIL (raw) and CONTRAIL (AK) CO$_2$ average data were worse in spring and summer than in fall and winter in the Northern Hemisphere in the upper troposphere. The differences between TIR and CONTRAIL (raw) and CONTRAIL (AK) CO$_2$ data were on average within 1 ppm in fall and winter in the northern troposphere. At 0°–40°N in summer, in contrast, the TIR and a priori CO$_2$ average data were 2.3 ppm lower than the CONTRAIL (AK) CO$_2$ average data. At 20°N–40°N in spring, the differences between TIR and CONTRAIL (AK) CO$_2$ average data were 2.4 ppm, although the TIR CO$_2$ data had a better agreement with CONTRAIL CME CO$_2$
data than a priori CO₂ data. On the other hand, the averages of the TIR CO₂ data were within 0-0.7 ppm of the averages of the CONTRAIL (AK) CO₂ data in the Southern Hemisphere in all seasons, as in the comparison in spring shown in Figure 5.

In the lower stratosphere, the agreements between the average TANSO-FTS TIR and CONTRAIL CME CO₂ data did not have a smaller seasonality than in the upper troposphere. The averages of TIR and CONTRAIL (raw) and CONTRAIL (AK) CO₂ data agreed with each other within 0.5% in all seasons.

7 Discussion

As shown in Figure 7, TANSO-FTS TIR V1 L2 CO₂ data had a negative bias of 2.3–2.4 ppm against CONTRAIL CME CO₂ data in the northern low and middle latitudes in spring and summer. Uncertainties in surface parameters and temperature profiles could affect CO₂ retrieval in thermal infrared spectral regions. As described above, retrieving surface parameters simultaneously instead of using initial surface parameters did not affect CO₂ concentrations in the UTLS regions in the TIR V1 CO₂ retrieval. We compared simultaneously retrieved temperature profiles with a priori JMA GPV temperature profiles in the UTLS region, and did not find any difference between the two which could explain the largest TIR CO₂ negative bias in the northern low and middle latitudes in spring and summer. In the UTLS regions, temperature variability is relatively large, and therefore comprehensive validation analysis of both the a priori and retrieved temperature profiles should be required using reliable and independent temperature data such as radiosonde data.

Uncertainty in a priori data could result in uncertainty in retrieved CO₂ data. Here, we arbitrarily decreased the a priori concentration by 1% in a test TIR CO₂ retrieval, and then compared the retrieved CO₂ concentrations with those retrieved using the original a priori data. In the northern low and middle latitudes in spring and summer where the DF values of TIR V1 CO₂ data were around 1.8 and more, a 1% negative bias in a priori data could yield up to a 0.7% negative bias in retrieved CO₂ concentrations in the altitude regions where we did comparisons between TIR and CONTRAIL CME data, although the magnitude of the bias varied depending on retrievals. As shown by the green lines in Figure 7, a priori CO₂ concentrations were underestimated by 2–4 ppm in the northern low and middle latitudes in spring and summer. The test TIR CO₂ retrieval demonstrated that the negative bias of a priori
CO₂ data against CONTRAIL CME data is a possible cause of the TIR CO₂ negative bias in
the UTLS regions in the northern low and middle latitudes in spring and summer.

In general, the information content of CO₂ observations made by TIR sensors is higher in
middle and high latitudes in spring and summer than in fall and winter because of the thermal
contrast in the atmosphere, with less seasonal dependence in low latitudes. Therefore, in
spring and summer, retrieved CO₂ data contain more measurement information and are less
constrained by a priori data at all latitudes. However, as shown in Figure 7, the retrieved TIR
CO₂ data in the northern low and middle latitudes did not sufficiently reduce the negative bias
of the a priori CO₂ data in the UTLS regions in spring and summer. This implies the existence
of factors that worsened CO₂ retrieval results other than the a priori data, especially in spring
and summer. Another possible factor that worsened CO₂ retrieval results is the uncertainty in
the calibration of TIR V161.160 L1B spectra. As reported in Kataoka et al. (2014), TANSO-
FTS TIR V130.130 L1B radiance spectra had a wavelength-dependent bias ranging from 0.1
to 2 K. Although the characteristics of the spectral bias in V161.160 L1B data used in TIR V1
L2 CO₂ retrievals are still under investigation, we assumed the same degree of bias in
V161.160 L1B spectra, and evaluated the effect of the L1B spectral bias on the TIR CO₂
retrieval using the following equation:

\[ \mathbf{d}_{\text{CO}_2} = \mathbf{G}_{\text{CO}_2} \mathbf{d}_\text{spec} \]  \hspace{1cm} (6)

Here, \( \mathbf{G}_{\text{CO}_2} \) is a gain matrix for CO₂ retrieval, \( \mathbf{d}_\text{spec} \) is a spectral bias vector based on the
evaluation by Kataoka et al. (2014), and \( \mathbf{d}_{\text{CO}_2} \) is a vector of bias errors in retrieved CO₂
concentrations attributable to the spectral bias. The result showed that a wavelength-
dependent bias comparable to V130.130 L1B spectra could yield up to 0.3% and 0.5%
uncertainties in retrieved CO₂ concentration in the UTLS regions in the northern middle
latitude in spring and in the northern low latitude in summer, respectively. Uncertainty in the
radiometric calibration of TANSO-FTS L1B spectra causes the spectral bias inherent in TIR
L1B spectra. The temperatures of the internal blackbody on board the TANSO-FTS
instrument partly reflect the environmental thermal conditions inside the instrument. The
temperatures of FTS-mechanics and aft-optics on the optical bench of the TANSO-FTS
instrument are precisely controlled at 23 °C. The difference in temperature between the
environment inside the instrument and the optical bench could cause the uncertainty in the
radiometric calibration of TANSO-FTS L1B spectra. Thus, the temperatures of the internal
blackbody on board the TANSO-FTS instrument could be a parameter used to evaluate the TANSO-FTS TIR L1B spectral bias.

Figure 8 shows the averages of the partial degree of freedom of TANSO-FTS TIR V1 L2 CO$_2$ data for each of the areas along the airline routes between Tokyo and Europe in the upper troposphere (a) and the lower stratosphere (b) for each season. The partial DF is defined as the diagonal element of the averaging kernels corresponding to TIR CO$_2$ data that were compared to CONTRAIL CME level flight data, which is equal to the 9$^{th}$, 10$^{th}$, or 11$^{th}$ diagonal element of matrix $A$. As shown in Figure 8, the average values of the partial DF of TIR lower stratospheric CO$_2$ data were clearly lower than those of TIR upper tropospheric CO$_2$ data for all of the fights between Tokyo and Europe. TIR upper tropospheric CO$_2$ data were from layers 9 and 10, and TIR lower stratospheric CO$_2$ data were from layers 10 and 11, as shown in Figure 3, which led to a clear difference in partial DF values between the TIR upper tropospheric and lower stratospheric CO$_2$ data. The partial DF values of TIR upper tropospheric CO$_2$ data were 0.13–0.20 in all of the areas for all seasons. In contrast, the partial DF values of TIR lower stratospheric CO$_2$ data in spring, fall, and winter were ~0.05 in almost all of the areas, although they were as high as 0.1–0.14 in summer. From the results shown in Figure 6c and Figure 8, we conclude that TIR CO$_2$ retrieval results in the lower stratosphere in winter were constrained to the relatively good a priori CO$_2$ data due to the low information content, and consequently had a good agreement with CONTRAIL CME CO$_2$ data. The comparisons in the areas during the airline route between Tokyo and Europe were included in the comparison results of 60$^{\circ}$N–70$^{\circ}$N in the right panels of Figure 7. In this region, the average differences between a priori and CONTRAIL (raw) data were 1-2 ppm in summer and fall, while they were less than 0.5 ppm in spring and winter. In summer, TIR CO$_2$ retrievals had a relatively high information content compared to the other seasons, which led to an agreement between TIR and CONTRAIL (raw) and CONTRAIL (AK) CO$_2$ data of within 0.5 ppm. In fall, TIR CO$_2$ retrieval results in the lower stratosphere were more constrained to the a priori CO$_2$ data, and therefore had a negative bias of approximately 1-2 ppm against CONTRAIL (raw) and CONTRAIL (AK) CO$_2$ data. In conclusion, the quality of TIR V1 CO$_2$ data in the lower stratosphere depends largely on the information content compared to the upper troposphere. In the case of high latitude measurements, TIR V1 lower stratospheric CO$_2$ data are only valid in summer.
We investigated the differences between TIR and CONTRAIL CO₂ comparison results in layers 9–11 with and without applying averaging kernel functions over the nine airports where CO₂ vertical profiles were observed during ascent and descent. In the northern middle latitudes in spring (NRT in Figure 4), CONTRAIL (AK) was on average 0.2 and 1.2 ppm lower than CONTRAIL (raw) in layers 9 and 10. In contrast, the tendency was the opposite in the southern middle latitudes in spring (SYD in Figure 4); CONTRAIL (AK) was on average 1.1 and 0.4 ppm higher than CONTRAIL (raw) in layers 9 and 10. This means that CO₂ concentrations in layers 9 and 10 were more affected by stratospheric air with relatively low CO₂ concentrations in the northern middle latitude in spring, when considering averaging kernels. This is consistent with the result of Sawa et al. (2012) showing that the difference between upper tropospheric and lower stratospheric CO₂ concentrations was larger in the Northern Hemisphere in spring.

Using CONTRAIL CME level flight observations that covered wide spatial areas allowed us to discuss the longitudinal differences in the characteristics of TIR UTLS CO₂ data. In the comparison results of the airline routes of Tokyo–Europe (Figure 6) and Tokyo–Vancouver (not shown here), the magnitudes of the differences between TIR and CONTRAIL (raw) and (AK) CO₂ data did not have a clear longitudinal dependence. Table 2 summarizes the latitudinal dependence of the magnitudes of the differences between TIR and CONTRAIL (AK) CO₂ data. In the upper troposphere in 0–60°N, negative biases in TIR CO₂ data against CONTRAIL CME CO₂ data ranged from 1.2 to 2.4 ppm in spring and summer, when applying averaging kernels to the assumed CME CO₂ profiles created based on CarbonTracker CT2013B monthly-mean profiles. It is the negative biases in the northern low and middle latitudes that we should in particular be concerned about when using TIR V1 L2 CO₂ data in any scientific analysis. In the upper troposphere in the northern middle latitudes, CO₂ concentrations reach the maximum from spring through early summer. The negative biases in TIR CO₂ data resulted in the maximum TIR CO₂ concentrations being lower than that of the CONTRAIL CME CO₂ concentrations, which led to an underestimate of the amplitude of the CO₂ seasonal variation when using TIR CO₂ data without taking their negative biases into account.
8 Summary

In this study, we conducted a comprehensive validation of the UTLS CO2 concentrations from the GOSAT/TANSO-FTS TIR V1 L2 CO2 product. The TIR V1 L2 CO2 algorithm used both the CO2 10 μm and 15 μm absorption bands (690–750 cm⁻¹, 790–795 cm⁻¹, 930–990 cm⁻¹, and 1040–1090 cm⁻¹), and simultaneously retrieved vertical profiles of CO2, water vapor, ozone, and temperature in these wavelength regions. Because the TANSO-FTS TIR V1 L1B radiance data used in the TIR V1 L2 CO2 retrieval had a spectral bias, we simultaneously derived surface temperature and surface emissivity in the same wavelength regions as a corrective parameter, other than temperature and gas profiles, to correct the spectral bias. The simultaneous retrieval of surface temperature greatly increased the number of normally retrieved CO2 profiles.

To validate the quality of TIR V1 upper atmospheric CO2 data, we compared them with the level flight CO2 data of CONTRAIL CME observations along the following airline routes in 2010: Tokyo–Europe (Amsterdam and Moscow), Tokyo–Vancouver, Tokyo–Honolulu, Tokyo–Bangkok, Tokyo–East Asia (Singapore and Jakarta), and Tokyo–Sydney. For the CONTRAIL data obtained during the northern high latitude flights, we made comparisons among CONTRAIL, TIR, and a priori CO2 data separately in the upper troposphere and in the lower stratosphere. The TIR upper tropospheric and lower stratospheric CO2 data that were compared were mainly from layers 9 and 10 (287–196 hPa) and from layers 10 and 11 (237–162 hPa), respectively. In this study, we evaluated the impact of considering TIR CO2 averaging kernel functions on CO2 concentrations using the CME profile data over the nine airports; the impact at around the CME level flight altitudes (~11 km) was on average less than 0.5 ppm in low latitudes and less than 1 ppm in middle and high latitudes.

In the Southern Hemisphere, the averages of TANSO-FTS TIR V1 upper atmospheric CO2 data were within 0.1% of the averages of CONTRAIL CO2 data with and without TIR CO2 averaging kernels for all seasons, from the limited comparisons made during flights between Tokyo and Sydney, while TIR CO2 data had a better agreement with CONTRAIL CO2 data than a priori CO2 data, with the agreement being on average within 0.5% in the Northern Hemisphere. The northern high latitude comparisons suggest that the quality of TIR lower stratospheric CO2 data depends largely on the information content. In high latitudes, TIR lower stratospheric CO2 data are only valid in summer when their information content is highest. Overall, the agreements of TIR and CONTRAIL CME CO2 data were worse in spring
and summer than in fall and winter in the Northern Hemisphere in the upper troposphere. TIR CO₂ data had a negative bias up to 2.4 ppm against CONTRAIL CO₂ data with TIR CO₂ averaging kernels in the northern low and middle latitudes in spring and summer. This is partly because of the larger negative bias in the a priori CO₂ data. The spectral bias inherent to TANSO-FTS TIR L1B radiance data could cause a negative bias in retrieved CO₂ concentrations, particularly in summer. TIR sensors can make more observations than SWIR sensors. When using the TIR UTLS CO₂ data, the seasonally and regionally dependent negative biases of the TIR V1 L2 CO₂ data presented here should be taken into account.

Acknowledgements

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References


Ohyama, H., Kawakami, S., Shiomi, K., Morino, I., and Uchino, O.: Atmospheric Temperature and Water Vapor Retrievals from GOSAT Thermal Infrared Spectra and Initial Validation with Coincident Radiosonde Measurements, SOLA, 9, 143-147, 2013.


Saeki, T., Saito, R., Belikov, D., and Maksyutov, S.: Global high-resolution simulations of CO2 and CH4 using a NIES transport model to produce a priori concentrations for use in satellite data retrievals, Geosci. Model Dev., 6, 81-100, 2013.


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Table 2. Bias values of GOSAT/TANSO-FTS TIR V1 CO₂ data against CONTRAIL (AK) CO₂ data for each season and each latitude region in the upper troposphere and lower stratosphere in the unit of ppm. Significant bias values larger than ±2 ppm are indicated by boldface.

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Figure 1. Averaging kernel functions of GOSAT/TANSO-FTS TIR V1 CO$_2$ retrieval in the 28 retrieval grid layers shown in Table 1: (a) low latitudes in summer, (b) mid-latitudes in spring, and (c) high latitudes in winter. Solid orange, yellow, and green lines indicate averaging kernel functions of each of the three layer levels 9, 10, and 11, respectively.
Figure 2. Flight tracks of all of the CONTRAIL CME observations in 2010 used in this study.
A number next to a box area indicates each area number.
Figure 3. The number of GOSAT/TANSO-FTS TIR CO$_2$ data points compared to the CONTRAIL CME level flight data for each retrieval grid layer level for each flight. The numbers of TIR CO$_2$ data points in the troposphere (“T”) and stratosphere (“S”) are shown separately for the Tokyo–Europe (NRT_DME_AMS), Tokyo–Vancouver (NRT_YVR), and Tokyo–Honolulu (NRT_HNL) flight routes.
Figure 4. Scatter plots of GOSAT/TANSO-FTS TIR and CONTRAIL (raw) CO$_2$ differences and GOSAT/TANSO-FTS TIR and CONTRAIL (AK) CO$_2$ differences in layers 9, 10, and 11 for each season.
Figure 5. Comparisons among CONTRAIL (raw), CONTRAIL (AK), GOSAT/TANSO-FTS TIR, and a priori (NIES TM 05) CO$_2$ data during flights between Tokyo and Sydney (NRT_Syd) in spring (MAM), shown by black, gray, red, and green lines, respectively. The means and their 1-σ standard deviations were calculated in each area during the flight for all four datasets.
Figure 6. Same as Figure 5, but for flights between Tokyo and Europe (NRT_DME_AMS) in winter (JF). (a) All of the data, (b) only data in the troposphere, and (c) only data in the stratosphere. See the text for the classification of tropospheric and stratospheric data.
Figure 7. Differences between GOSAT/TANSO-FTS TIR and CONTRAIL (raw) averaged CO₂ data (TIR ave. minus CONTRAIL (raw) ave.), TIR and CONTRAIL (AK) averaged CO₂ data (TIR ave. minus CONTRAIL (AK) ave.), and a priori (NIES TM 05) and CONTRAIL (raw) averaged CO₂ data (a priori ave. minus CONTRAIL (raw) ave.) for each season for each latitude band (40°S–20°S, 20°S–0°, 0°–20°N, 20°N–40°N, 40°N–60°N, 60°N–70°N), shown by black, gray, and green lines, respectively. Left and right panels show the differences in the upper troposphere and lower stratosphere, respectively. The 1-σ standard deviations of the latitudinal averages of TANSO-FTS TIR CO₂ data are shown by vertical bars.
Figure 8. Partial degree of freedom (DF) for GOSAT/TANSO-FTS TIR CO₂ data in the upper
troposphere (a) and the lower stratosphere (b) for each area of the flight between Tokyo and
Europe (NRT_DME_AMS). The means and their 1-σ standard deviations of the partial DF
data were calculated in spring (MAM), summer (JJA), fall (SON), and winter (JF), as shown
by the pink, red, light blue, and blue lines, respectively.