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A Differential Optical Absorption Spectroscopy Method for X_{CO_2} Retrieval from Ground-Based Fourier Transform Spectrometers Measurements of the Direct Solar Beam

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ABSTRACT

A differential optical absorption spectroscopy (DOAS)-like algorithm is developed to retrieve the column-averaged dry-air mole fraction of carbon dioxide from ground-based hyper-spectral measurements of the direct solar beam. Different to the spectral fitting method, which minimizes the difference between the observed and simulated spectra, the ratios of multiple channel-pairs—one weak and one strong absorption channel—are used to retrieve X_{CO_2} from measurements of the shortwave infrared (SWIR) band. Based on sensitivity tests, a super channel-pair is carefully selected to reduce the effects of solar lines, water vapor, air temperature, pressure, instrument noise, and frequency shift on retrieval errors. The new algorithm reduces computational cost and the retrievals are less sensitive to temperature and H_2O uncertainty than the spectral fitting method. Multi-day Total Carbon Column Observing Network (TCCON) measurements under clear-sky conditions at two sites (Tsukuba and Bremen) are used to derive X_{CO_2} for the algorithm evaluation and validation. The DOAS-like results agree very well with those of the TCCON algorithm after correction of an airmass-dependent bias.

Key words: CO_2 Retrieval, ground-based measurement, hyper-spectrum, shortwave infrared band

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1. Introduction

Carbon dioxide (CO_2) is considered to be the main greenhouse gas causing current global warming (Solomon et al., 2007). However, Easterling and Wehner (2009) reported that records of surface air temperature show no warming trend or even a slight cooling trend, while greenhouse gas levels are still increasing. The disagreement about climate change is mostly due to the lack of long-term records of CO_2 measurements, especially for large area measurements and CO_2 sources and sinks (Stephens et al., 2007; Canadell et al., 2010).

It is advantageous to use satellite remote sensing to monitor atmospheric CO_2 globally. However, at present, only the satellite datasets of column-averaged dry-air mole fraction of CO_2 (X_{CO_2}) from the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) on board the Environmental Satellite (ENVISAT) (Bovensmann et al., 1999) and the Thermal and Near-infrared Sensor for Carbon Observation–Fourier Transform Spectrometer (TANSO-FTS) on board the Greenhouse Gases Observing Satellite (GOSAT) (Kuze et al., 2009), are used to estimate regional CO_2 fluxes. Both instruments use the reflected solar radiation in the shortwave infrared (SWIR) spectral region, making them sensitive to the variation of near-surface CO_2 concentrations. Unfortunately, the low spectral resolution of SCIAMACHY limits the inversion accuracy, with a single retrieval precision of about 2.5 ppm, as compared to ground-based Fourier transform spectrometer (FTS) measurements (Buchwitz et al., 2005; Reuter et al., 2011). The biases and standard deviations of the column-averaged dry-air mole fraction of carbon dioxide (X_{CO_2}) from the SWIR L2 V02.xx GOSAT retrieval algorithm reach −1.48 and 2.09

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ppm, respectively (Morino et al., 2011; Yoshida et al., 2011; Yoshida et al., 2013).

Compared with satellite observations of reflected light, ground-based observations of the direct solar beam are less influenced by surface albedo, aerosols etc. Therefore, ground-based observations can achieve higher accuracy and precision in determining the CO_2 total column amount. However, at present, the Total Carbon Column Observing Network (TCCON) is the only existing network that retrieves the total column concentration of CO_2 from ground-based FTS measurements for satellite validation. TCCON achieves a network-wide uncertainty of X_{CO_2} of better than 0.8 ppm, with 2σ after correcting for an airmass-dependent bias and calibrating to aircraft vertical profiles (Wunch et al., 2011a, b).

A Chinese satellite for CO_2 monitoring is planned for launch in 2015 (Liu et al., 2013). To validate the satellite retrievals, a surface observation network has been set up to measure the hyper-spectrum of the direct solar beam in the SWIR bands. To derive the total column amount of CO_2 from these spectral measurements, a retrieval algorithm is needed. In this paper, a new DOAS-like algorithm is developed, in which multiple pairs of CO_2 absorption ratios (one in the weak CO_2 absorption channel and one in the strong CO_2 absorption channel) are used to derive the column CO_2. More importantly, both channels in the pair are carefully selected to reduce their sensitivity to the surface pressure, air temperature, water vapor, noise and frequency shift. Compared with the spectral fitting method, DOAS-like retrievals are less sensitive to temperature and H_2O uncertainty.

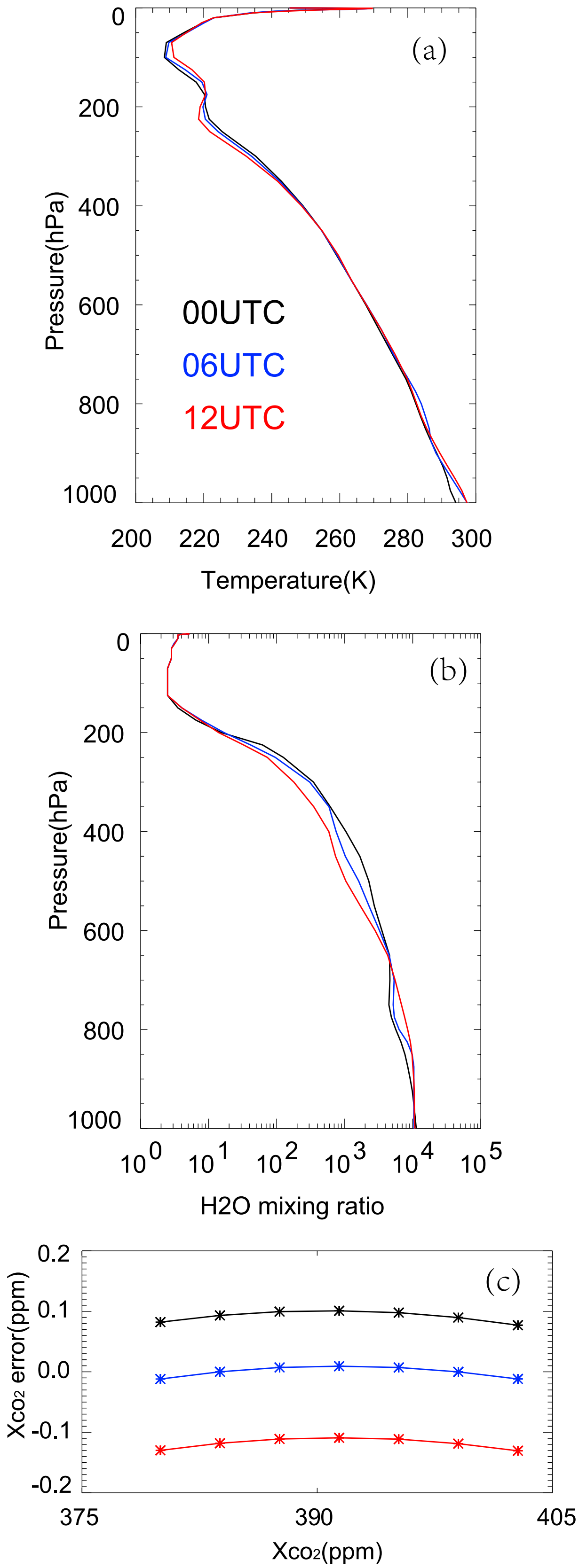

Fig. 1. (a) Temperature and (b) water vapor profiles at 0000 (black), 0600 (blue) and 1200 UTC (red) in one day, and (c) inversion errors with same coefficients *a* and *b*.

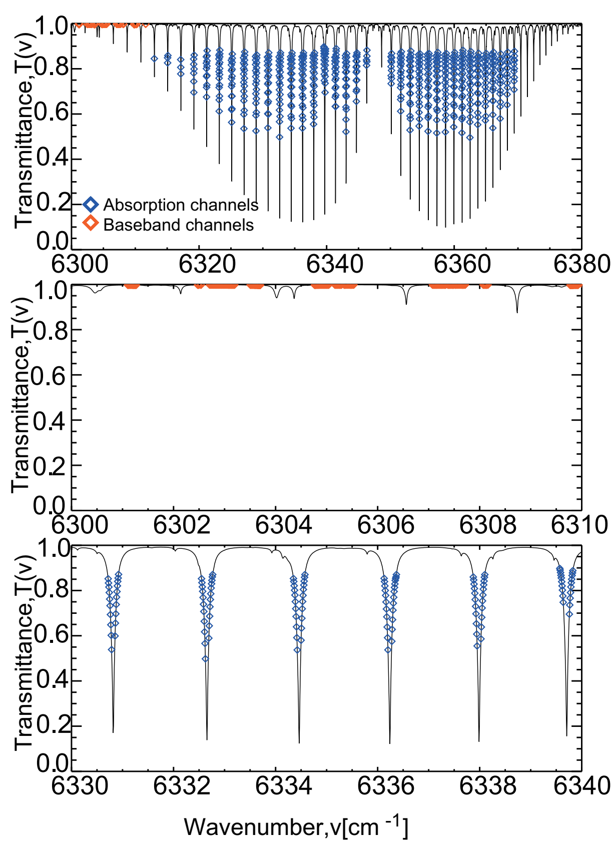

Fig. 2. Channels used in the retrieval algorithm.

2. Retrieval algorithm

2.1. *Physical basis*

Our retrieval algorithm is based on the fact that the photon path lengths within a narrow spectral range are equal. Therefore, the ratio of the channel pair is proportional to X_{CO_2} if the surface pressure, temperature profile and water vapor are known. Based on the Lambert–Beer law, a ground-based measurement of the direct solar beam for a fixed wavelength can be expressed as

$$I_\lambda = I_{\mathrm{sca},\lambda} + I_{0,\lambda} e^{-\tau m} , \tag{1}$$

where I_λ is the downward radiance measured at the bottom of the atmosphere for wavelength λ, $I_{\mathrm{sca},\lambda}$ is the forward scattering contribution in the incident direction, and I_0 is the incoming solar radiance at the top of the atmosphere. m is the air mass factor. τ is the optical depth in the vertical optical path, which can be written as

$$\tau = \tau_{CO_2} + \tau_{H_2O} + \tau_{\mathrm{aer}} + \tau_{\mathrm{Ray}} , \tag{2}$$

where the right-hand terms represent optical depth of CO_2 absorption, water vapor absorption, aerosol extinction, and Rayleigh scattering, respectively. The scattering term of $I_{\mathrm{sca},\lambda}$ in Eq. (1) is negligible due to a very small field of view (FOV) (∼2.4 mrad) of the spectrometer, particularly for small aerosol particles and small aerosol optical depths (Min et al., 2004; Min and Duan, 2005; Wunch et al., 2011a). Therefore, the radiance can be simplified as

$$I_\lambda = I_0 e^{-(\tau_{CO_2} + \tau_{H_2O} + \tau_{\mathrm{aer}} + \tau_{\mathrm{Ray}}) m} . \tag{3}$$

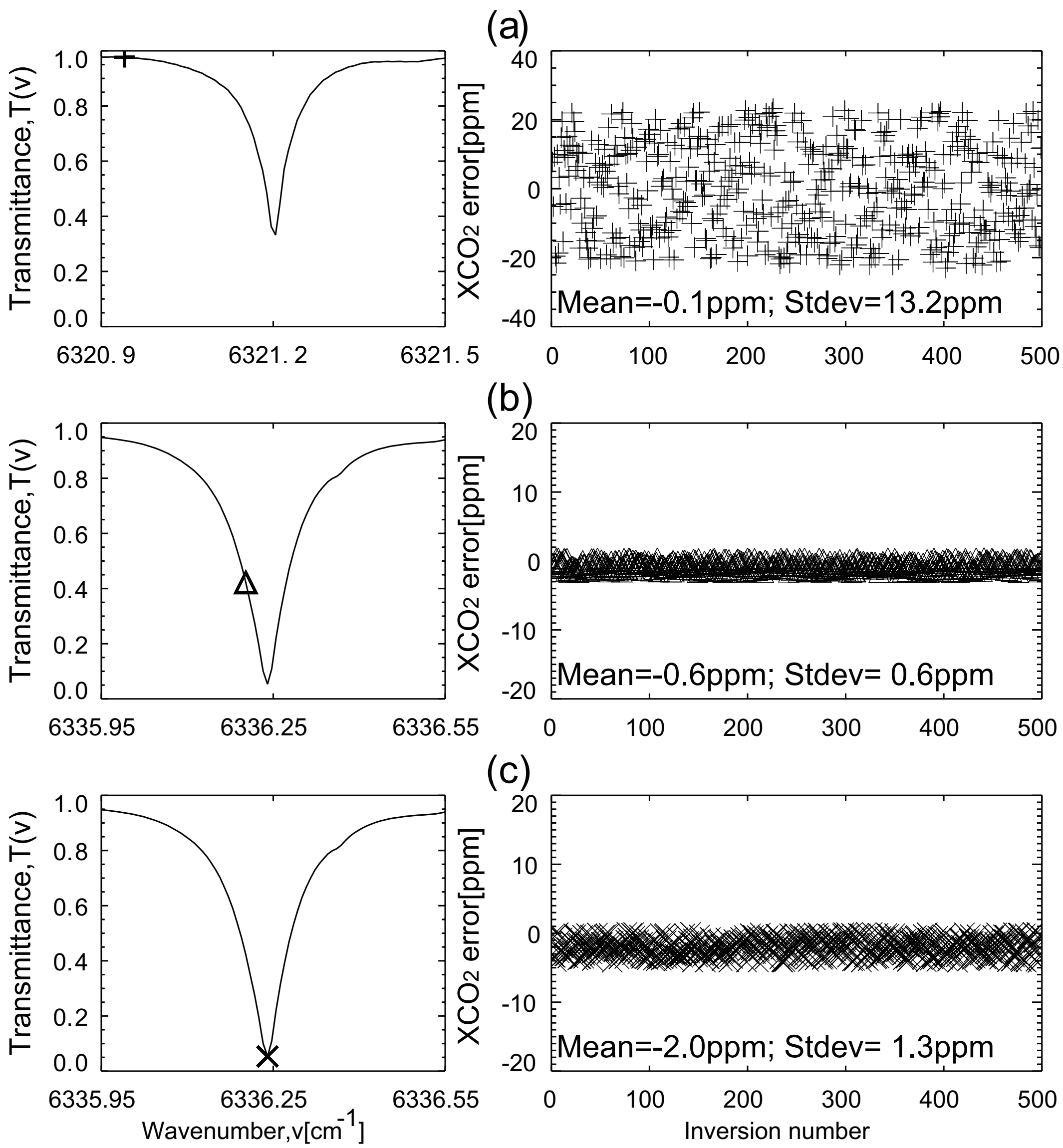

Fig. 3. Error analysis of X_{CO_2} retrieval when the strong absorption channel is located at the (a) far-wing (plus symbol), (b) in the middle (triangle symbol), and (c) center (cross symbol) of the absorption line. Five-hundred retrievals with random noise under the assumption of SNR of 500 are illustrated. Left-hand panels represent the spectrum of a single absorption line, and right-hand panels the errors of X_{CO_2} retrievals.

In a very limited spectral range, the variation of τ_{aer} and τ_{Ray} across the spectral range can be ignored. Therefore the ratio of the selected channel pair is insensitive to the loading of aerosol and Rayleigh scattering. Hence, we have

$$\frac{I_{\lambda_1}}{I_{\lambda_2}} = \frac{I_{0,\lambda_1}}{I_{0,\lambda_2}} e^{-[(\tau_{CO_2,\lambda_1} - \tau_{CO_2,\lambda_2}) - (\tau_{H_2O,\lambda_1} - \tau_{H_2O,\lambda_2})]m} . \quad (4)$$

Letting $r = I_{\lambda_1}/I_{\lambda_2}$ and $r_0 = I_{0,\lambda_1}/I_{0,\lambda_2}$, Eq. (4) can be rewritten as

$$r = r_0 e^{-[(\tau_{CO_2,\lambda_1} - \tau_{CO_2,\lambda_2}) - (\tau_{H_2O,\lambda_1} - \tau_{H_2O,\lambda_2})]m} . \quad (5)$$

By taking the logarithm of Eq. (5), we have

$$\ln(r) = \ln(r_0) - [(\tau_{CO_2,\lambda_1} - \tau_{CO_2,\lambda_2}) - (\tau_{H_2O,\lambda_1} - \tau_{H_2O,\lambda_2})]m . \quad (6)$$

The optical depth τ_{CO_2} is proportional to the total number of molecules of CO_2 per surface area, which is positively correlated to X_{CO_2} if the surface pressure, air temperature and CO_2 volume mixing ratio (VMR) profile are assumed to be known. Furthermore, only channel pairs with weak H_2O absorption interference are selected. Therefore, the difference associated with water vapor is small and can be treated as a correction coefficient. Then, Eq. (6) is simplified as

$$X_{CO_2} = a \ln(r) + b . \quad (7)$$

Through the about pair selection procedure, the retrieval, i.e., Eq. (7), is weakly sensitive to the atmospheric state uncertainty (temperature and water vapor). Nonetheless, the coefficients of *a* and *b* are weakly dependent on temperature and water vapor in the atmosphere. To further reduce the error associated with the atmospheric state, we can calculate both

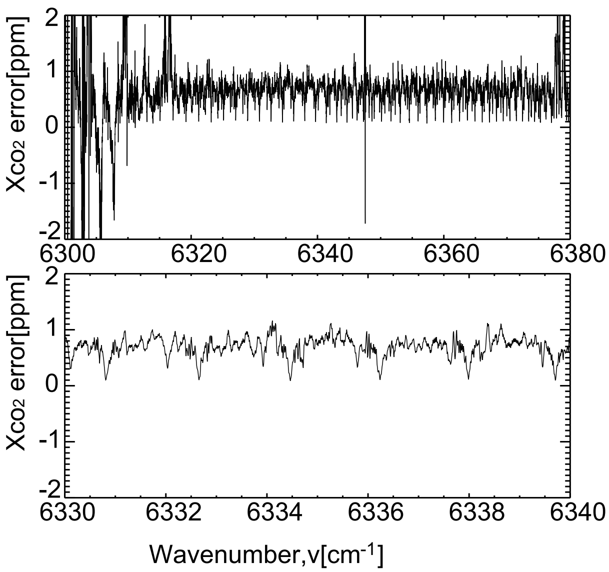

Fig. 4. The X_{CO_2} errors of each channel for +1 hPa bias of the surface pressure when the channel is used as the only strong absorption channel in the super channel-pair.

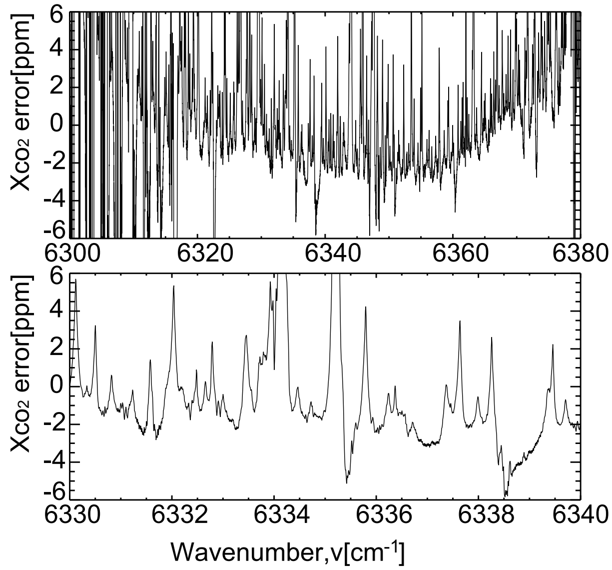

Fig. 5. The X_{CO_2} errors of each channel for a +1 K bias of temperature profile when the channel is used as the only strong absorption channel in the super channel-pair.

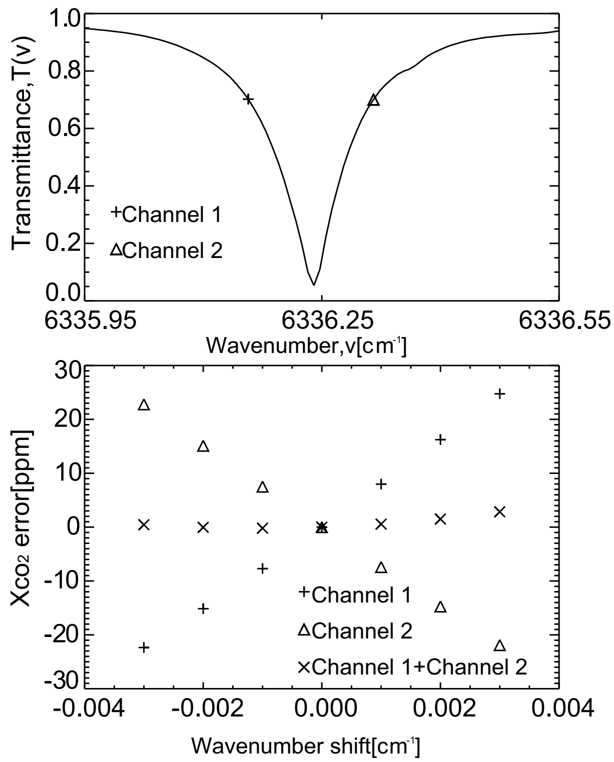

Fig. 6. The strong CO_2 absorption channels distributed on different sides of an absorption line (upper panel) and the inversion errors (lower panel) caused by frequency shift when channel 1 (plus symbol), channel 2 (triangle symbol), and the average of channel 1 and channel 2 (cross symbol) are used as the strong absorption channel in the super channel-pair.

coefficients with the surface pressure of *in-situ* measurements collected by automatic meteorological stations and reanalysis/forecasting atmospheric profiles. The profiles can be fixed for multiple measurements within some specific time period because only channels that are independent of temperature and water vapor are used in our retrieval algorithm. To illustrate the feasibility of fixed profiles, several inversions calculated by the different coefficients *a* and *b* at 0000, 0600 and 1200 UTC are shown in Fig. 1. All the errors are less than 0.15 ppm. The DOAS-like algorithm of Eq. (7) only has one unknown parameter. Hence, no iteration is needed.

2.2. *Forward model*

The Atmospheric Environment Research (AER) Line By Line Radiative Transfer Model (LBLRTM) is used to calculate the gases' absorption coefficients. The line parameters database, aer_v_3.2, is also from AER and a Voigt line shape is assumed. The extra-terrestrial solar spectrum is provided by Kurucz (Kurucz, 1995). The atmosphere is divided into 70 layers in 1 km intervals. The surface pressure and a priori VMR profiles at TCCON stations are from TCCON auxiliary data. In the forward model, LBLRTM is used to calculate the gas absorption optical depth with a step of 0.001 cm^{-1}, and these absorptions along with the Rayleigh scattering are used to calculate the fine structure of direct radiance. Finally, the fine spectra are convolved with a resolution and sample rate of 0.02 cm^{-1} and 7.5 kHz for measurements in Tsukuba, and 0.014 cm^{-1} and 7.5 kHz for measurements in Bremen (Wunch et al., 2011a).

2.3. *Channel selection*

The DOAS-like method could reduce computational cost, but the super channel-pair must be carefully selected to reduce the impacts of H_2O absorption, the solar Fraunhofer lines, and other factors such as instrument noise, temperature, pressure, frequency shift etc. In our channel-pair, the mean of 430 channels with very weak CO_2 absorption is regarded as the weak absorption channel in the super channel-pair, which is applied to the following analyses, and the mean of some strong CO_2 absorption channels is regarded as the strong absorption channel in the super channel-pair, as shown in Fig. 2. The selection of the strong absorption channel in the super channel-pair is presented in the following paragraphs.

The effects of random noise are also analyzed in the strong absorption channel selection to avoid large errors. Figure 3 illustrates the errors due to instrument noise in differently positioned strong CO_2 absorption channels if only one strong CO_2 absorption channel is used in X_{CO_2} retrieval. It is clearly shown that when the strong CO_2 absorption channel located at the far wing is used, large errors could be introduced due to the reduced information content of CO_2 (Fig. 3a); while in the line center, low signal-to-noise ratio (SNR) results in large uncertainty (Fig. 3c).

The line strength and absorption coefficients depend on pressure and temperature. For the ultra-high spectral resolution measurements, an inaccurate pressure and temperature

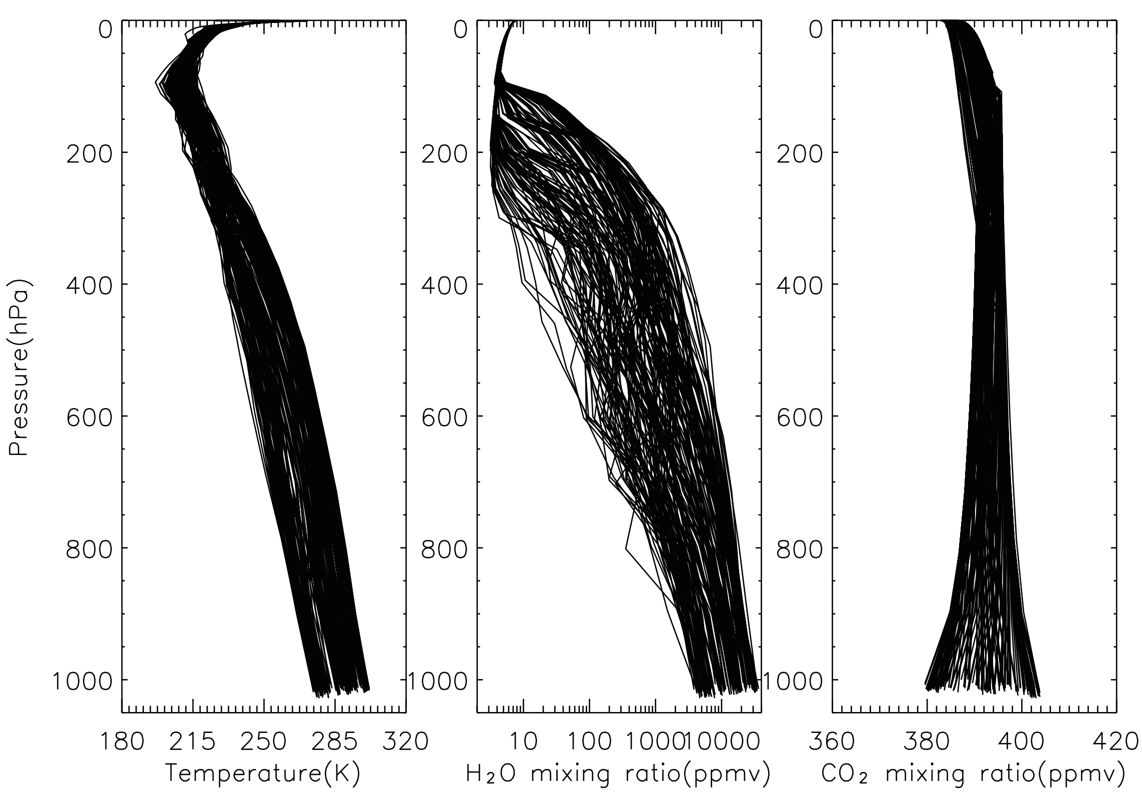

Fig. 7. The prior atmosphere profiles of temperature (left panel), water vapor (middle panel) and CO_2 (right panel) used in the sensitivity tests.

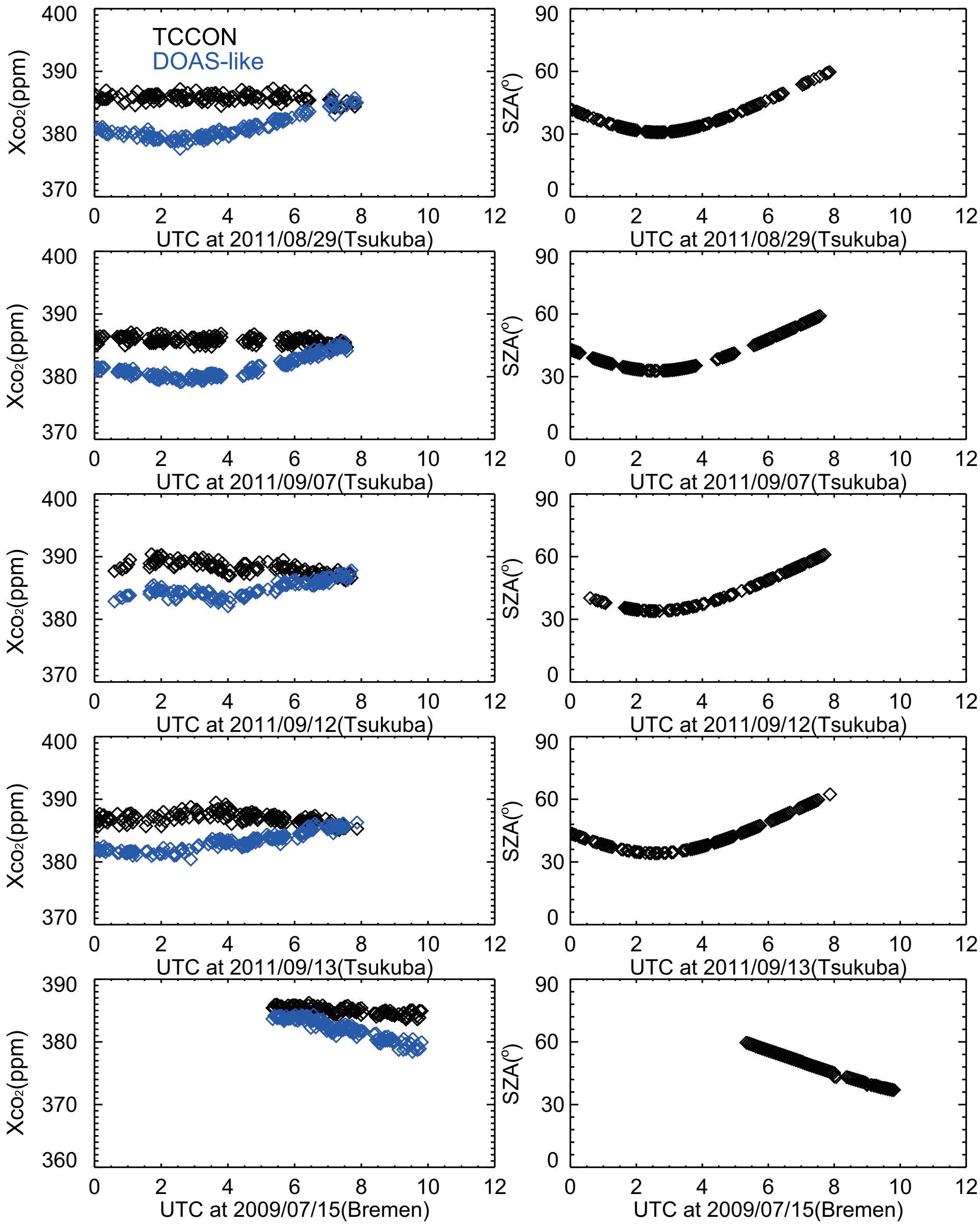

Fig. 8. Left panels: inversion X_{CO_2} of TCCON (black) and the DOAS-like (blue) method at Tsukuba and Bremen. Right panels: the corresponding SZA time series.

profile will introduce extra errors in the retrieval of CO_2. Figure 4 shows the X_{CO_2} errors of each channel for a +1 hPa bias of surface pressure, which is calculated by comparing inversions with and without a 1 hPa change, when the channel is regarded as the only strong CO_2 absorption channel. In the error calculations, the coefficients a and b are calculated under the surface pressure, while the "measurements" are given under the +1 hPa bias of the surface pressure. Similarly, Fig. 5 shows the X_{CO_2} errors for a +1 K shift of the temperature profile. As shown in Fig. 4, inversion errors caused by the +1 hPa pressure bias of most channels are positive, except for some channels in the weak absorption area. To reduce the

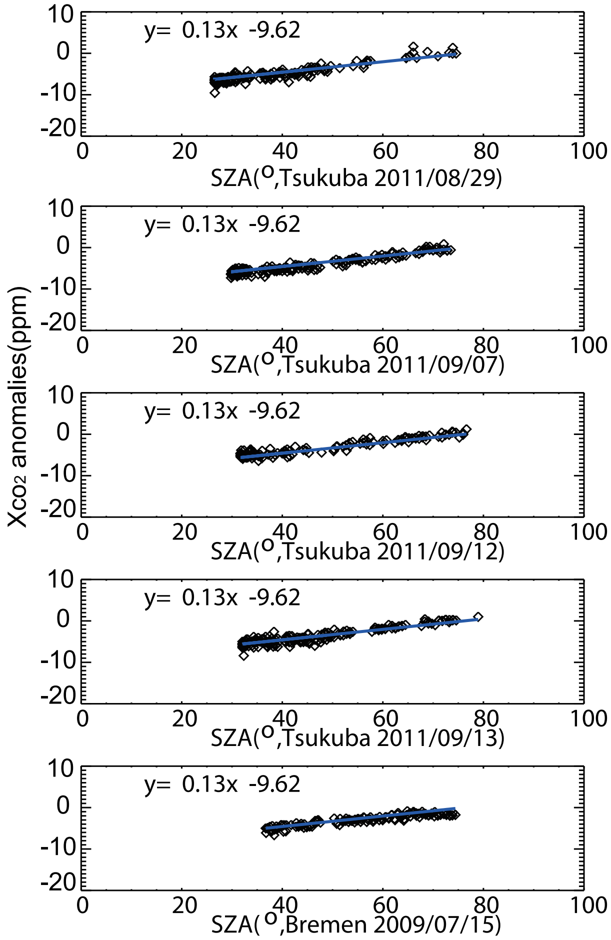

Fig. 9. The variation of X_{CO_2} anomalies between the DOAS-like method and TCCON results with SZA at Tsukuba and Bremen.

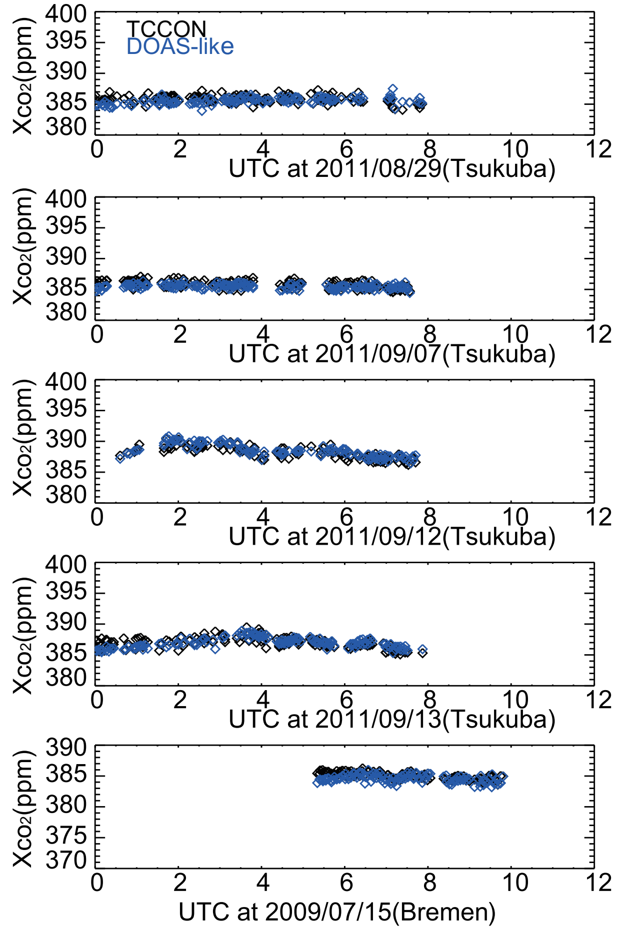

Fig. 10. The inversion X_{CO_2} of TCCON (black) and the DOAS-like method (blue) in Tsukuba and Bremen after correction.

impact of pressure, only channels with an inversion error of less than 1 ppm are selected to be the component of the strong absorption channels in the super channel-pair. Different from that of pressure, the error due to the +1 K offset of the temperature profile could be either positive or negative, and it could be minimized by careful channel selection in real retrievals.

Inaccurate wavelength registration is another source of error in retrievals. As shown in Fig. 6, the errors in X_{CO_2} retrieval due to a frequency shift of 0.003 cm^{-1} could be up to 25 ppm if only one strong CO_2 absorption channel located on one side of the line center is used. But if strong CO_2 absorption channels located on both sides of the line center are used, the errors due to the frequency shift tend to be very small, or even zero.

Based on the above sensitivity studies, and the additional removal of the channels with strong H_2O absorption and Fraunhofer lines, the final 588 strong CO_2 absorption channels are used in our retrieval. In order to evaluate the dependence of the DOAS-like method on the atmospheric state uncertainty, one-year prior profiles in Tsukuba, as shown in Fig. 7, are used in simulated inversions. The results for both the DOAS-like and spectral fitting methods are listed in Table 1, in which errors of 1 K for the temperature profile, +5% for water vapor, +1 hPa for surface pressure, and 0.001 cm^{-1} for frequency offset are assumed for solar zenith angles (SZAs) at 20° and 70°. For specific atmospheric parameter analysis, both the spectral fitting and DOAS-like method have one unknown state vector. Relatively, the DOAS-like retrievals are less sensitive to the temperature and H_2O uncertainties, especially for large SZAs and high H_2O amounts. The effects of surface pressure and frequency shift to being slightly better in the spectral fitting method.

3. Case studies and comparisons

To validate the DOAS-like algorithm, TCCON data in Tsukuba, Japan (36.0513°N, 140.1215°E) and Bremen, Germany (53.10°N, 8.85°E) are used. The spectra at both stations are measured with an FTS (IFS 125HR, Bruker Optics GmbH, Germany). The absorption spectrum is calculated by a Fourier transform of the interferogram, which is formed by beams reflected from a moving mirror and a static mirror. The resolution and sample rate of the FTS are determined by

Table 1. The X_{CO_2} errors due to temperature, water vapor, surface pressure and spectral shift.

	20° SZA				70° SZA			
	DOAS-like		Spectral fitting		DOAS-like		Spectral fitting	
Error (ppm)	mean	std	mean	std	mean	std	mean	std
Temperature profile (+1 K)	−0.11	0.05	−0.19	0.02	−0.1	0.05	−0.39	0.03
H_2O (+5%)	0.14	0.11	−0.19	0.17	0.16	0.12	−0.55	0.37
Surface pressure (+1 hPa)	0.5	0.02	0.39	0.03	0.54	0.02	0.39	0.04
Spectral shift (+0.001 cm^{-1})	0.07	0.02	0.0	0.02	−0.02	0.02	0.01	0.04

the maximum optical path differences (MOPDs) and speed of the moving mirror. The MOPDs of the FTS in Tsukuba and Germany are 45.01 and 64.29 cm, respectively. The retrieved X_{CO_2} using the DOAS-like method are illustrated in Fig. 8 (left panels), and the results of the official TCCON algorithm are also included for comparison. At first sight, the X_{CO_2} of the DOAS-like method is smaller than that of the official TCCON algorithm. After comparing the difference between the TCCON and DOAS-like retrievals with the SZA (Fig. 8, right panels), we find that the difference is linearly dependent on the SZA (Fig. 9). Moreover, the linear relationship does not vary with time and place. For the TCCON results, a post-retrieval algorithm is used to correct an airmass-dependent bias based on the assumption that any symmetric variability within a day should be an artifact (Deutscher et al., 2010; Wunch et al., 2011a). Through a simple correction process in which the linear dependency on the SZA is removed, the DOAS-like and TCCON results agree well with each other, as shown in Fig. 10. The standard deviation of the difference between the TCCON and DOAS-like methods is less than 0.8 ppm, both in Tsukuba and Bremen (Fig. 11). This suggests that the DOAS-like algorithm provides retrievals with similar precision to TCCON. However, the temporal variability of the atmospheric state in Fig. 12 limits the possibility of a higher inversion accuracy. Certainly, there could be many other factors for the low values of DOAS-like retrievals. For example, the solar lines provided by Kurucz used in our algorithm are not so good (Yoshida et al., 2013), and the FTS only focuses on the center of the solar disk due to its very small FOV. This inaccurate extra-terrestrial solar spectrum may be a factor for our lower value of X_{CO_2}.

4. Conclusions and future directions

A new algorithm using a channel-pair ratio to derive X_{CO_2} is presented in this paper. The algorithm is similar to that of the DOAS method. For the purpose of channel selection, the effects of solar lines, water vapor, air temperature, pressure, instrument noise and wavelength registration shift on the retrieval error are analyzed through a series of sensitivity tests. One super channel-pair is used in the retrieval algorithm. FTS measurements at the TCCON stations in Tsukuba and Bremen are used to validate the new algorithm by comparing our results with the official TCCON product. Our X_{CO_2} results are lower than those of TCCON with airmass correction. Taking the TCCON data as a reference, our results are further corrected using an SZA-dependent method. After the correction, our corrected results agree well with those of the TCCON products, suggesting that this new algorithm is useful. However, due to insufficient ground measurements, the new retrieval method is validated by observations at only two stations. Clearly, a thorough validation with extensive

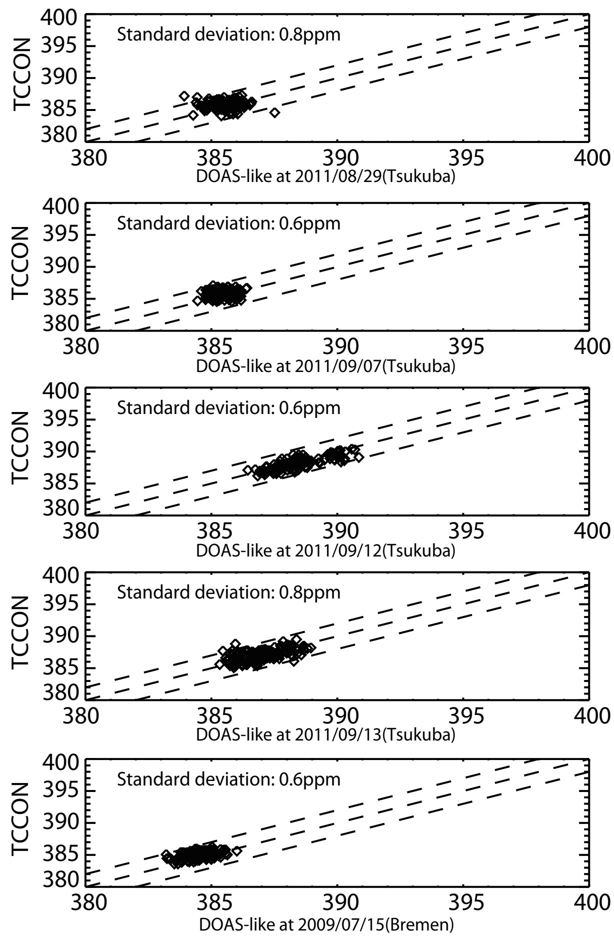

Fig. 11. Comparison between the results of TCCON and the DOAS-like method after correction at Tsukuba and Bremen. The standard deviations are shown in each panel.

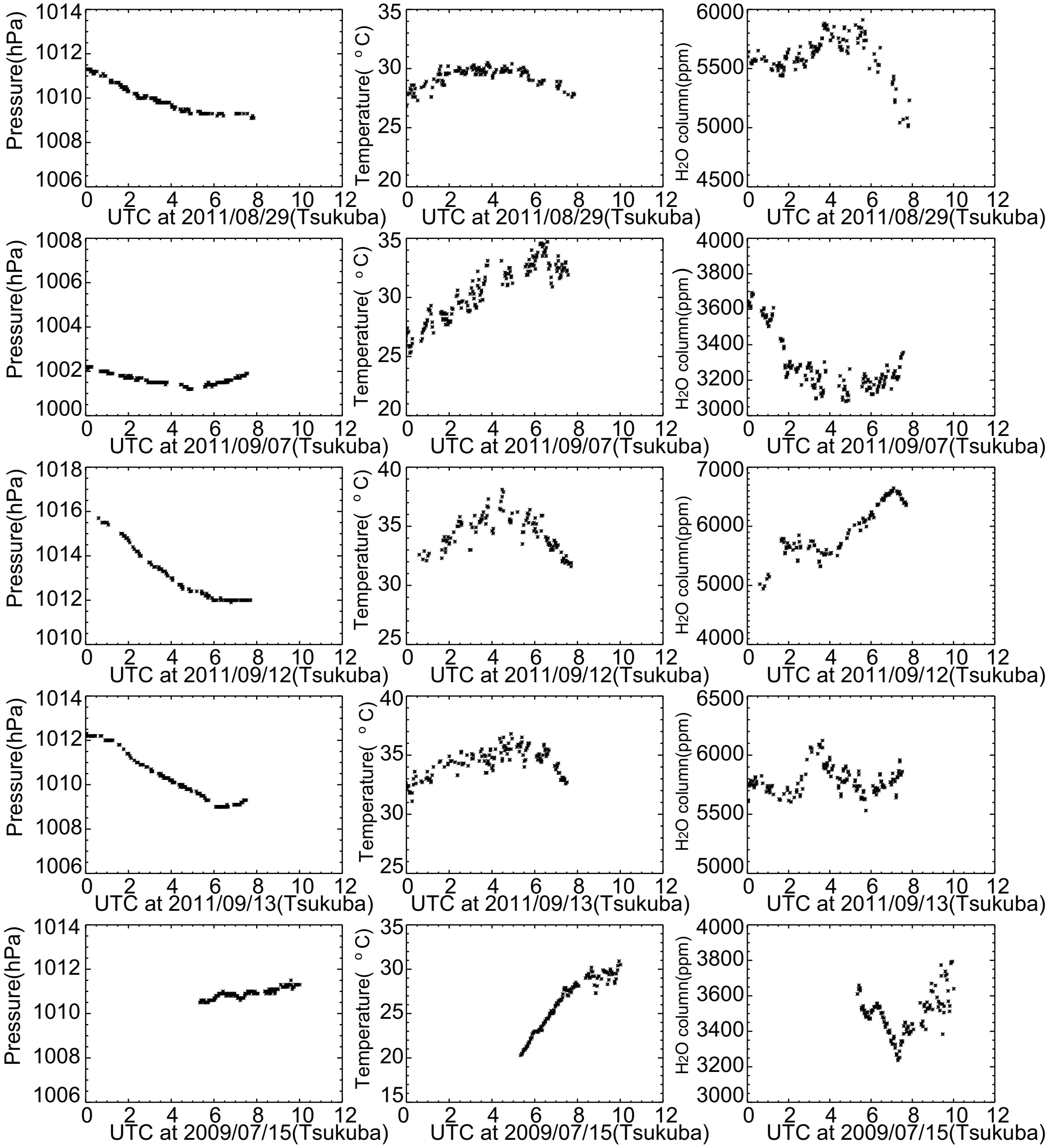

Fig. 12. The time series of surface pressure (left panels), surface temperature (middle panels), and H_2O column (right panels) for each day.

observation is warranted for our DOAS-like algorithm.

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