



## Abstract

The practical usefulness of a desktop optical spectrum analyzer (OSA) for measuring atmospheric CO<sub>2</sub> and CH<sub>4</sub> column densities at surface sites was examined in two separate measurement campaigns. The first involved a long term measurement in parallel with a high resolution Fourier transform spectroscopy (FTS) studies at the University of Wollongong in Australia. Scale factors of the OSA were assigned for the column average volume mixing ratios of xCO<sub>2</sub> and xCH<sub>4</sub> by comparing with the well-studied FTS. The second method is a calibration against aircraft CO<sub>2</sub> profiles in situ over Tsukuba in Japan obtained during a GOSAT validation campaign carried out from 28 January to 7 February 2011. The xCO<sub>2</sub> values in the campaign, deduced by use of a derived OSA scale factor, were in excellent agreement with the integrated aircraft profiles.

## 1 Introduction

Carbon dioxide and methane have the highest and the second highest contributions of ~64 and ~18 %, respectively, to overall global radiative forcing from major greenhouse gases (WMO, 2011). The growth rate of atmospheric CO<sub>2</sub> averaged 2.9 PgCyr<sup>-1</sup> or 1.37 ppmyr<sup>-1</sup> in 1959–2006 (Canadell et al., 2007) and has increased to the annual mean growth rate of 2.38 ppmyr<sup>-1</sup> in 2010 (NOAA). The concentration of CO<sub>2</sub> in the marine surface layer has increased by 50 ppm in the last 30 yr (NOAA) and the global temperature has increased by about 0.6 °C (Brohan et al., 2006). Estimation of source and sink strengths is required to better manage CO<sub>2</sub> gas emission. The Greenhouse gases Observing Satellite (GOSAT: IBUKI) of Japan was launched on 23 January 2009 and data acquisition for the CO<sub>2</sub> and CH<sub>4</sub> column densities has progressed by using an onboard Fourier transform spectrometer, FTS (Kuze et al., 2009), with good precision from space. The Total Carbon Column Observing Network (TC-CON: <http://www.tcon.caltech.edu/>) is a network of ground-based FTS instruments with high-resolution providing precise column densities of CO<sub>2</sub>, O<sub>2</sub>, CH<sub>4</sub>, H<sub>2</sub>O, HDO,

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HF, CO, N<sub>2</sub>O, etc. (Wunch et al., 2011). TCCON is composed of twenty-one sites at this moment in time, and fifteen FTS instruments are operational in Canada, USA, New Zealand, Australia, Japan and some European countries. The TCCON data are highly reliable and are used for validations of GOSAT and other satellites (Washenfelder et al., 2006; Duetscher et al., 2010; Wunch et al., 2010; Morino et al., 2011). However surface monitoring sites of total column densities of greenhouse gases are lacking in many developing countries in the Asian, African and South American continents and on oceans. This is probably due to the high setup cost of an FTS which also requires a highly educated operator. In addition FTS is unsuitable for portable use under severe climate conditions.

In a previous paper we have proposed a desktop Optical Spectrum Analyzer (OSA) for measuring atmospheric CO<sub>2</sub> and CH<sub>4</sub> column densities at surface monitoring sites (Kobayashi et al., 2010: hereafter we denote Part 1). The grating-based OSA resolves rotational lines of CO<sub>2</sub> and CH<sub>4</sub> in the near infrared (NIR) region. The OSA instrument is compact, portable, low cost, rugged and basically maintenance free.

In the present paper the practical usefulness of the OSA system was examined for measuring atmospheric CO<sub>2</sub> and CH<sub>4</sub> column densities using two methods: One of them is a parallel measurement with a TCCON FTS at the University of Wollongong (UoW) for 14 months, in which scale factors for the column averaged volume mixing ratios of xCO<sub>2</sub> and xCH<sub>4</sub> were determined. The second method uses aircraft CO<sub>2</sub> profiles obtained in a GOSAT validation carried out at Tsukuba in Japan, a TCCON site, on 28 January–7 February 2011, which provided the opportunity to compare the scale factor against the aircraft profiles measured in situ. In this campaign the xCO<sub>2</sub> and xCH<sub>4</sub> from the OSA were also compared with those obtained with the FTS operated by the Japan Aerospace Exploration Agency (JAXA) measured under the same conditions.

## 2 Instrumental

The system for measuring atmospheric CO<sub>2</sub> and CH<sub>4</sub> column densities is composed of a desktop OSA (Yokogawa AQ6370 series) and a portable sun tracker as described in detail in Part 1. A block diagram of the data acquisition is shown in Fig. 1. The OSA (AQ6370-custom) disperses radiation from 600 to 1800 nm, and its dimensions are 43(*W*) × 22(*H*) × 46(*D*) cm with a weight of 19 kg. The wavelength resolution is 0.08 cm<sup>-1</sup> at 1600 nm (6250 cm<sup>-1</sup>) but this depends on the core size of optical fibre employed. The sun tracker was equipped with a GPS and a small telescope for concentrating the sunlight onto an optical fibre. A long-pass filter (HOYA RM100, λ > 1000 nm) was attached at the front of the object lens of the telescope to cut off the second order stray light. Geophysical data of latitude, longitude, a.s.l. and UTC from the GPS and meteorological information of pressure, temperature and relative humidity on the measuring surface site are accumulated using a laptop computer and a data logger. Solar absorption spectra measured by the OSA were stored in the laptop computer. Reference solar intensity in the region of 1000–1700 nm monitored by an InGaAs detector was coincidentally measured with the spectrum signal and used to compensate for fluctuations in the sunlight intensity.

## 3 Performances for practical usability of the OSA

### 3.1 OSA measurements in parallel with FTS

Solar absorption spectra in the regions of 1569–1575 and 1673–1679 nm were measured for the CO<sub>2</sub> and CH<sub>4</sub> rotational lines, respectively, from July 2010 to August 2011 at UoW in Australia. Typical spectra from the OSA are shown in Fig. 2 where the black and red curves are the observed and fitted spectra, respectively. The residual is shown in the top panel. The full widths at half maximum of the peaks were found to be 0.2090 and 0.1475 cm<sup>-1</sup> for CO<sub>2</sub> and CH<sub>4</sub>, respectively, deduced as a fitting parameter under

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standard operational conditions of the OSA. The absorption spectra of CO<sub>2</sub> and CH<sub>4</sub> were also measured by a Bruker IFS 125HR FTS under the same conditions following TCCON standards. The OSA and FTS systems were operated in parallel about 2 m apart fed by the same solar beam. The spectra obtained by the FTS and the OSA were independently retrieved, respectively, by means of the GFIT algorithm (Toon et al., 1992; Wunch et al., 2011) and the software given in Part 1 where a constant volume mixing is assumed and the HITRAN 2008 database (Rothman et al., 2009) is adopted.

**3.1.1 Carbon dioxide**

The column average volume mixing ratio  $x\text{CO}_2$  is defined as the ratio of the column density of CO<sub>2</sub> to the total column of dry air (Washenfelder et al., 2006):

$$x\text{CO}_2 = [\text{column of CO}_2] / [\text{total column of dry air}] \quad (1)$$

where the total column of dry air is given by

$$[\text{total column of dry air}] = [\text{total column of air}] - [\text{column of H}_2\text{O}] \quad (2)$$

The total column of air is calculated by use of the pressure measured at the surface site, the gravitational acceleration, the molecular weight of air and Avogadro's constant. The profiles of temperature, pressure and relative humidity against altitude are available from the database of National Centers for Environmental Prediction/National Center for Atmospheric Research (NCEP/NCAR) to calculate the column of H<sub>2</sub>O.

An example of daily analyses is shown in Fig. 3 for 3 July 2010 where the solid and the open circles represent the OSA and the UoW FTS measurements, respectively. In the present OSA analyses, a discrimination level of 3% was employed for the fractional solar variation monitored as the NIR intensity shown in Fig. 1. The scale factor for  $x\text{CO}_2$  from the OSA is given just below. In spite of several times larger standard deviations of the OSA in the third and sixth columns in Table 1, the averages of the column density and  $x\text{CO}_2$  from the OSA (the second and fifth columns) are very close to those from the

FTS. Sources of the errors for the TCCON FTS measurements have been described in detail (Wunch et al., 2011).

Time series of 14 months of the column density and  $x\text{CO}_2$  for the OSA are shown in Fig. 4 where scaled data from the FTS are superimposed over the period July–October 2010 for the column density and July 2010–June 2011 for  $x\text{CO}_2$ . The averages between 10:00–14:00 LT, while the solar intensity is stable, were plotted with the standard deviations. Averages of the OSA and FTS column densities measured during July–October were  $(8.369 \pm 0.087) \times 10^{21}$  and  $(8.413 \pm 0.056) \times 10^{21}$  molecules  $\text{cm}^{-2}$ , respectively, a ratio of 0.995 and thus in good agreement. The straight line in the upper panel has a slope of the global  $\text{CO}_2$  growth rate of  $2.38 \text{ ppm yr}^{-1}$  in 2010 (NOAA).

The scale factor of 0.989 has been applied to the  $x\text{CO}_2$  from the FTS instruments in the TCCON network derived from aircraft profiles measured over the TCCON sites in order to place them on the World Meteorological Organization (WMO) standard reference scales (Wunch et al., 2010). The  $\text{CO}_2$  scale factors for the present OSA are given in the fourth column in Table 2: they were determined to be the ratio of the mean  $\text{CO}_2$  concentration over two months (the second column) derived by the fitting algorithm described in Part 1 relative to  $x\text{CO}_2$  in the same period from the FTS (the third column). The average of  $1.008 \pm 0.002$  was assigned to the scale factor for  $x\text{CO}_2$  from the OSA. The standard deviations in the  $x\text{CO}_2$  from the OSA are less than  $\pm 0.6\%$  (the second column) but still larger than those from the FTS (the third column) by 2–4 times. The numerals in the last column indicate the total number of days used in evaluating the scale factors.

### 3.1.2 Methane

A typical spectrum of methane along with a model fit and difference obtained with the OSA is shown in Fig. 2b. The two peaks at 1674.4 and 1677.6 nm have been assigned to the absorption of  $\text{CH}_4$  while the other features are water or Fraunhofer lines. The scale factor of  $1.035 \pm 0.004$  for  $x\text{CH}_4$  in Table 2 was obtained as an adjusting parameter between the OSA and FTS  $x\text{CH}_4$  normalized by the TCCON scale factor of 0.978

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(Wunch et al., 2010). The standard deviations in the mean  $x\text{CH}_4$  from the OSA (the fifth column in Table 2) are similarly larger to the case of  $x\text{CO}_2$ .

Time series of the column density and  $x\text{CH}_4$  are shown in Fig. 5 where the solid and the open circles are from the OSA and FTS, respectively. The averages of the column density and  $x\text{CH}_4$  from the OSA over a period of 14 months were  $(3.886 \pm 0.071) \times 10^{19}$  molecules  $\text{cm}^{-2}$  and  $1.759 \pm 0.030$  ppm, respectively. The latter agrees with the value of  $1.766 \pm 0.008$  ppm at Baring Head in New Zealand ( $41.41^\circ$  S,  $174.87^\circ$  E) measured by means of gas chromatography from June to December 2010 (WMO WDCGG). The seasonal cycle observed in the flask samplings at Baring Head, however, was not clear in the OSA data as it may be buried in the measurement noise.

### 3.2 Substantiation with in situ aircraft profiles

We measured atmospheric  $\text{CO}_2$  and  $\text{CH}_4$  column densities at Tsukuba in Japan using an OSA and a ground-based Bruker IFS 125HR FTS operated by JAXA from 25 January to 13 March in 2011. During this period a campaign for the validation of the GOSAT measurements took place in collaboration with JAXA and the National Institute for Environmental Studies of Japan on 28, 31 January, 3 and 7 February in 2011 by flying an aircraft over Tsukuba. We examine here the usability of the OSA instrument by comparing with the JAXA FTS retrieved by using the GFIT algorithm and aircraft  $\text{CO}_2$  profiles measured in situ. Meteorological data against altitude were obtained from the surface up to 20 km for pressure, temperature, relative humidity, wind direction and wind speed by launching sondes equipped with GPS.

#### 3.2.1 Carbon dioxide

Figure 6 shows the results obtained by the OSA (solid circles), the JAXA FTS (open circles) and the aircraft (open squares), where the scale factor for the OSA as determined in Sect 3.1.1 was employed. The mean value of the column density obtained by the OSA from 27 January to 13 March was  $(8.514 \pm 0.059) \times 10^{21}$  molecules  $\text{cm}^{-2}$

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at JAXA in Tsukuba (36.066° N, 140.229° E: 25 m a.s.l.). In general the column density is sensitive to the height above sea level at the measuring surface site. The column density of  $(8.339 \pm 0.061) \times 10^{21}$  molecules  $\text{cm}^{-2}$  measured by the OSA at UoW (34.406° S, 150.879° E: 30 m a.s.l.) in the same period is thus directly comparable with that at JAXA in Tsukuba: the column density at JAXA in the Northern Hemisphere is higher than that at UoW in the Southern Hemisphere by 2.1 %. The average  $x\text{CO}_2$  from the OSA and FTS over the 7 weeks were  $392.74 \pm 1.18$  and  $391.85 \pm 0.79$  ppm, respectively, in agreement with a ratio of 0.998. The weather in the latter half of February was cloudy and probably contributed to the large standard deviation in the OSA data. The maximum and minimum differences in the  $x\text{CO}_2$  between the OSA and FTS were 2.45 ppm on 31 January and  $-0.51$  ppm on 8 March, respectively. The average  $x\text{CO}_2$  from the OSA at UoW during 31 January–13 March was  $387.23 \pm 1.57$  ppm, being lower than that at JAXA in Tsukuba by 5.5 ppm.

The vertical profiles of temperature over Tsukuba measured by the sondes are shown in Fig. 7a where the first tropopauses lie at 8.7, 7.5, 8.3 and 10.2 km on 28 January (black), 31 January (red), 3 February (blue) and 7 February (olive), respectively, indicated by arrows while a common second tropopause exists around 16 km. Tropospheric  $\text{CO}_2$  concentrations were measured in situ by means of a continuous measuring instrument installed in the aircraft with a height profile ranging from about 400 m to 7 km in altitude. The flight time of the aircraft overlapped with the duration of the sonde. The vertical  $\text{CO}_2$  profiles over Tsukuba were depicted by a previously reported estimation (Araki et al., 2010): the  $\text{CO}_2$  concentration at the lowest observable point was assumed to continue down to the surface while that at the highest point at around 7 km extends up to the first tropopause (Fig. 7b). The  $\text{CO}_2$  concentration at the first tropopause was assumed to decrease linearly to the value at 20 km in latitude (Araki et al., 2010). The stratospheric  $\text{CO}_2$  concentration above 20 km is considered to be constant, the value of which lags about five years behind the global mean  $\text{CO}_2$  in the troposphere (Aoki et al., 2003). Thus the annual mean  $\text{CO}_2$  of 380.91 ppm in 2006 (NOAA) was adopted in the present work as the concentration above 20 km in latitude.



Table 3 summarizes the integrated aircraft  $x\text{CO}_2$ , OSA and FTS for the 4 days of the comparison. The aircraft  $x\text{CO}_2$  derived from the vertical profile mentioned above is given in the second column. The volume mixing ratios from the OSA and FTS relative to that from the aircraft (the fourth and last columns) give the averages of  $0.999 \pm 0.003$  and  $0.996 \pm 0.001$ , respectively, indicating that the three independent measurements are in good agreement. This also shows that the scale factor of 1.008 for the OSA deduced in Sect 3.1.1 is consistent with both FTS and aircraft results. The difference in  $x\text{CO}_2$  between the aircraft and the OSA or FTS on 7 February is larger than usual,  $-1.7$  or  $-1.9$  ppm, respectively. The reason for the deviation is not clear but the NIR intensity reference signal (Fig. 1) was 10% weaker than the other 3 days, most likely due to the presence of thin clouds and this may therefore result in the less reliable value for  $x\text{CO}_2$ .

### 3.2.2 Methane

Time series of  $x\text{CH}_4$  and column densities for methane are shown in Fig. 8, where the averages of  $x\text{CH}_4$  from the OSA (solid circles) and FTS (open circles) were  $1.778 \pm 0.019$  and  $1.790 \pm 0.007$  ppm, respectively, for the 7 weeks, which are in good agreement. The mean  $x\text{CH}_4$  at UoW in the same period was  $1.764 \pm 0.026$  ppm, being lower than those at JAXA in Tsukuba by 14–26 ppb. Concentrations of  $\text{CH}_4$  in the air analyzed by precision instruments are lower in the Southern Hemisphere than in the Northern Hemisphere (WMO, 2006) and thus the present observation of  $x\text{CH}_4$  is consistent with the global trend of  $\text{CH}_4$ . However, when we take into account the standard deviations in  $x\text{CH}_4$ , the present difference is only qualitative. The mean column density was  $(3.948 \pm 0.048) \times 10^{19}$  molecules  $\text{cm}^{-2}$  at JAXA while that at UoW during this period was  $(3.899 \pm 0.060) \times 10^{19}$  molecules  $\text{cm}^{-2}$ , being a little low but still within the measurement noise.

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## 4 Summary

Two field campaigns presented in this paper have shown that an optical spectrum analyzer (OSA) is a promising technology for measuring atmospheric CO<sub>2</sub> and CH<sub>4</sub> column densities at surface sites. The OSA due to its smaller physical size and lower cost is expected to provide a supplemental measuring system to the existing FTS network. The standard deviation of the retrieved OSA column density at present is 2–4 times higher than that of the collocated FTS. The data quality will be improved in future through improvements in the analysis procedures, the shortening of the data acquisition intervals and the application of more stringent data quality criteria.

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**Table 1.** Column densities and  $x\text{CO}_2$  for the OSA and FTS on 3 July 2010 at the University of Wollongong in Australia.

Instrument	Column density <sup>a</sup>	Standard deviation	Error estimated	$x\text{CO}_2$ ppm	Standard deviation	Error estimated
OSA	8.384	0.031	nd <sup>b</sup>	383.88	1.39	nd
FTS (UoW)	8.429	0.009	0.10	384.48	0.32	1.0

<sup>a</sup> Units in  $10^{21}$  molecules $\text{cm}^{-2}$ .<sup>b</sup> Not determined.

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**Table 2.** Scale factors for  $x\text{CO}_2$  and  $x\text{CH}_4$  from the OSA.

Year/ Month	Mean $x\text{CO}_2$ in 2 months		SF <sup>a</sup> CO <sub>2</sub>	Mean $x\text{CH}_4$ in 2 months		SF CH <sub>4</sub>	Total days
	OSA <sup>b</sup>	FTS (UoW)		OSA	FTS (UoW)		
2010/7–8	389.67 ± 1.82	385.83 ± 0.85	1.010	1.792 ± 0.030	1.737 ± 0.011	1.032	20
9–10	390.31 ± 2.35	386.77 ± 0.70	1.009	1.812 ± 0.017	1.741 ± 0.009	1.041	15
11–12	390.11 ± 1.17	387.04 ± 0.47	1.008	1.814 ± 0.029	1.745 ± 0.009	1.039	18
2011/1–2	390.22 ± 2.71	388.41 ± 0.47	1.005	1.825 ± 0.024	1.772 ± 0.007	1.030	12
3–4	390.90 ± 2.01	388.20 ± 0.50	1.007	1.824 ± 0.030	1.765 ± 0.006	1.033	16
5–6	392.19 ± 1.09	389.26 ± 0.61	1.007	1.827 ± 0.020	1.772 ± 0.004	1.031	21

<sup>a</sup> SF denotes scale factor. Average =  $1.008 \pm 0.002$  and  $1.035 \pm 0.004$  for  $x\text{CO}_2$  and  $x\text{CH}_4$ , respectively.

<sup>b</sup> Units in ppm.

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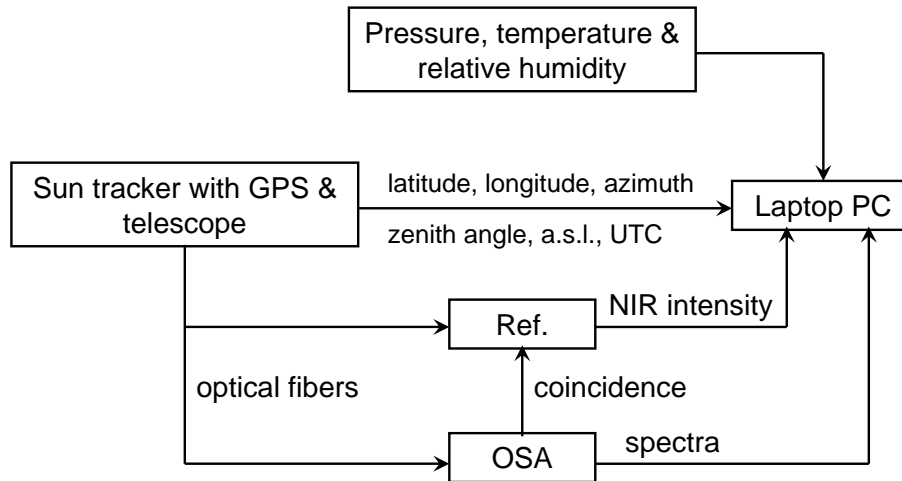
**Table 3.** Volume mixing ratios by aircraft, OSA and FTS at Tsukuba in Japan.

Date in 2011	Aircraft <sup>a</sup>	OSA	Ratio <sup>b</sup>	FTS (JAXA)	Ratio
28 January	392.03	392.19 ± 0.71	1.000	390.53 ± 0.68	0.996
31 January	392.19	393.67 ± 0.89	1.004	391.22 ± 0.77	0.998
3 February	392.68	391.76 ± 0.79	0.998	391.12 ± 0.58	0.996
7 February	394.05	392.34 ± 0.77	0.996	392.17 ± 0.77	0.995

<sup>a</sup> Units in ppm.<sup>b</sup> Ratio denotes the relative  $x\text{CO}_2$  from the OSA or FTS (JAXA) to that from the aircraft. Average =  $0.999 \pm 0.003$  and  $0.996 \pm 0.001$  for OSA and FTS (JAXA), respectively.

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**Fig. 1.** Block diagram of the data acquisition system.

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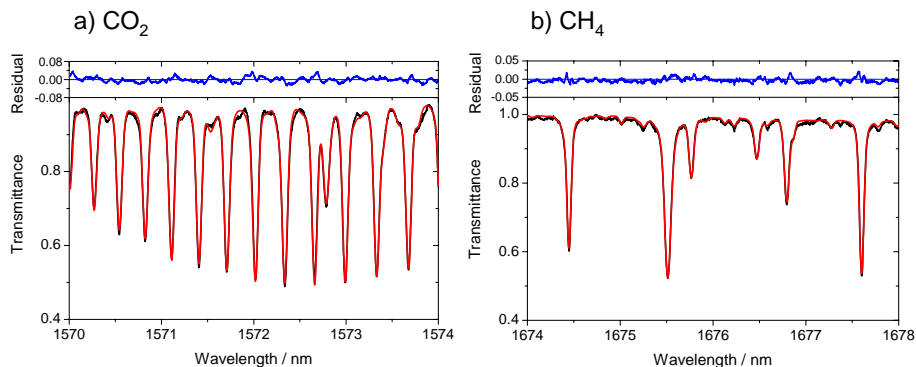
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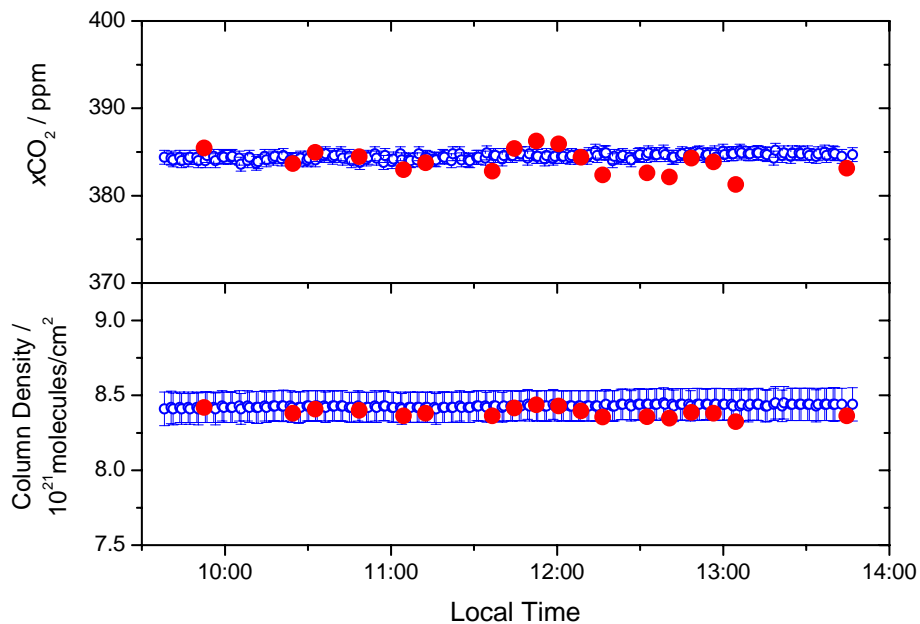


**Fig. 2.** Absorption spectra of CO<sub>2</sub> and CH<sub>4</sub>. Black and red curves denote the observed and fitted spectra, respectively. The residual is shown in the top panel.

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**Fig. 3.** Time series of column density and  $x\text{CO}_2$  on 3 July 2010 at UoW in Australia. OSA: solid circles; FTS: open circles with estimated errors.

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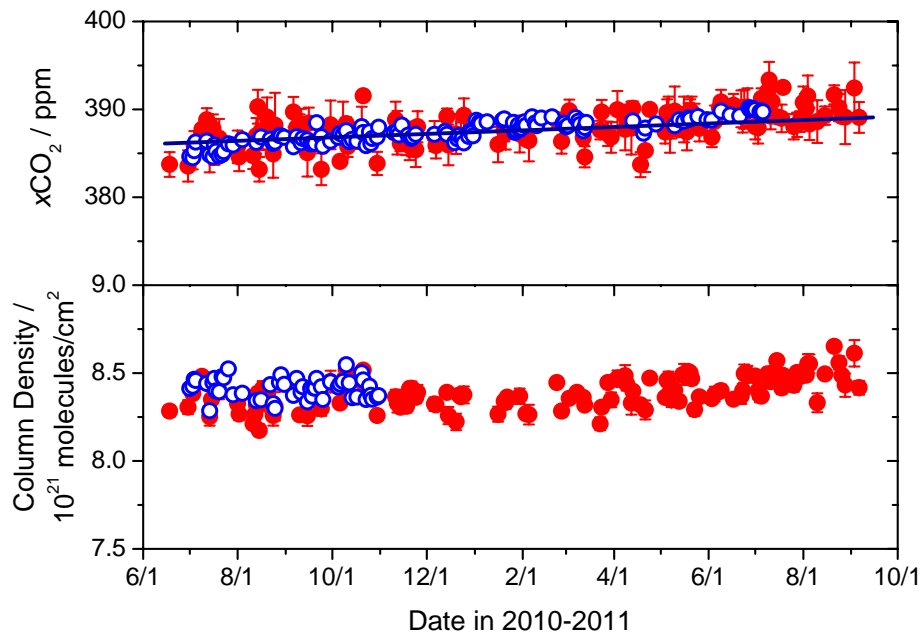
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**Fig. 4.** Time series of column density and  $x\text{CO}_2$  with the standard deviations for 14 months at UoW in Australia. OSA: solid circles; FTS: open circles. The line in the top panel has a slope of the global growth rate  $2.38 \text{ ppm yr}^{-1}$  in 2010.

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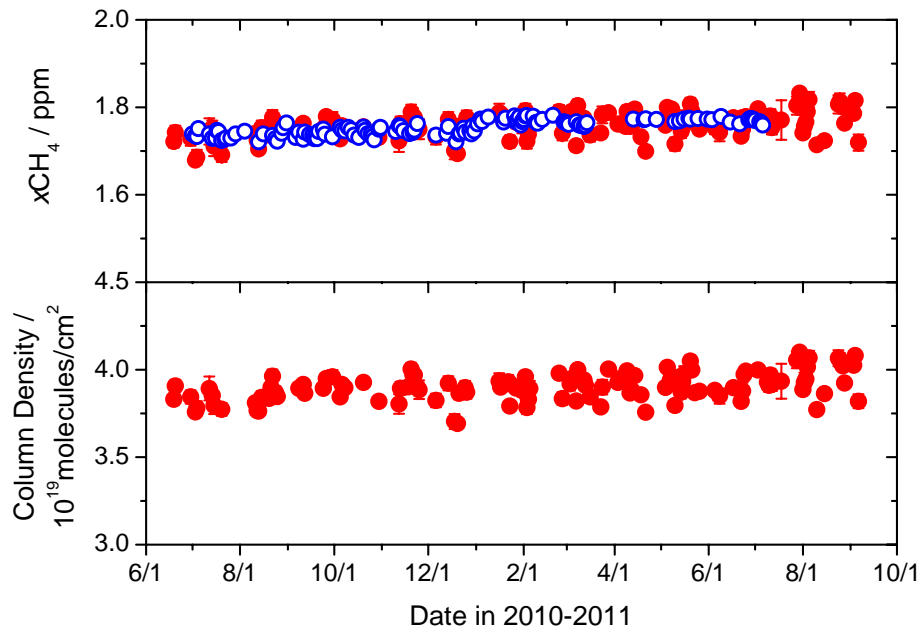
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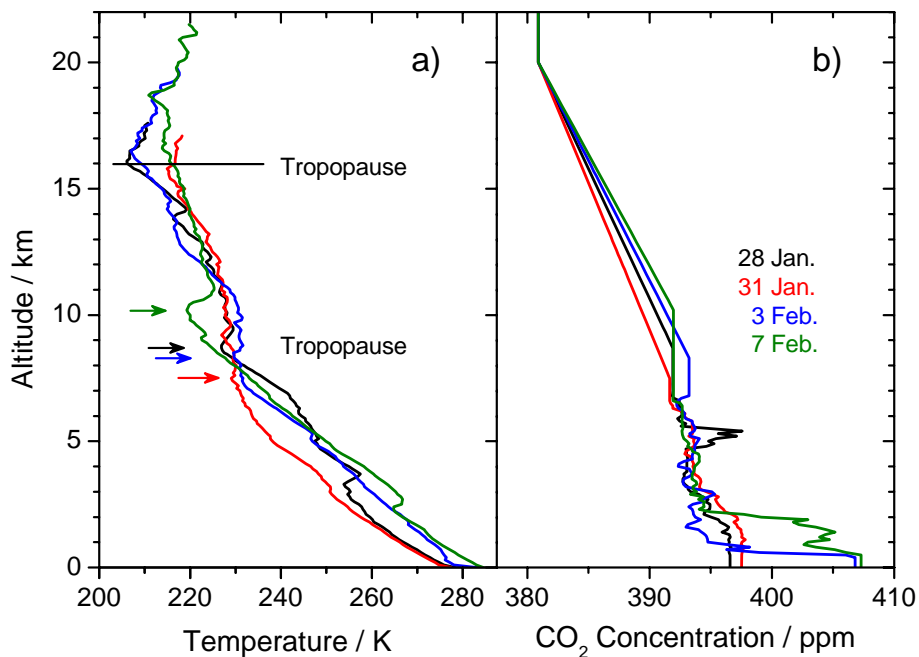
**Fig. 5.** Time series of column density and  $x\text{CH}_4$  with the standard deviations for 14 months at UoW in Australia. OSA: solid circles; FTS: open circles.

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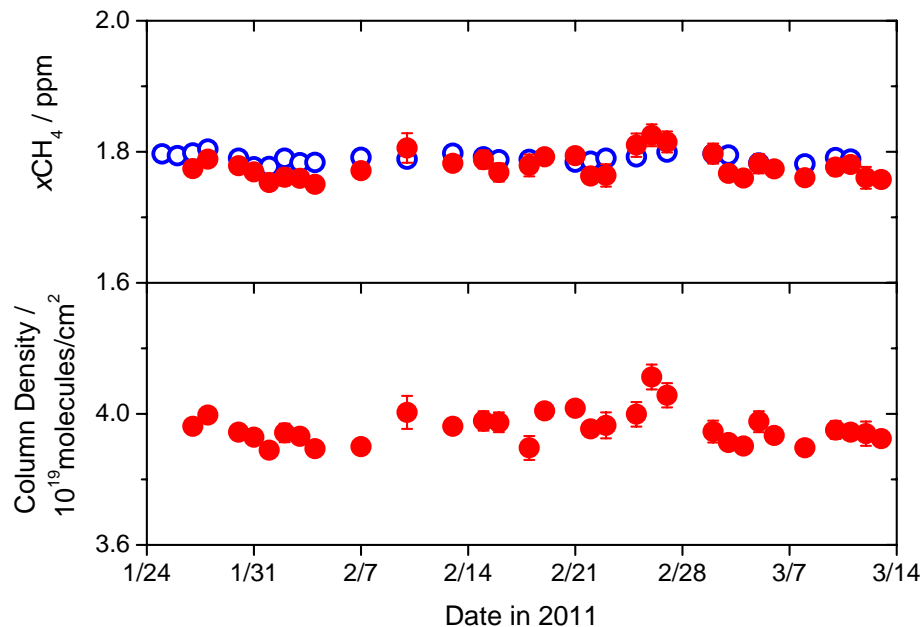


**Fig. 7.** Vertical profiles of temperature and CO<sub>2</sub> concentration over Tsukuba in Japan on 28 (black), 31 January (red), 3 (blue) and 7 February 2011 (olive). Details are given in the text.

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**Fig. 8.** Time series of column density and  $x\text{CH}_4$  with the standard deviations at JAXA in Tsukuba, Japan. OSA: solid circles; FTS: open circles.

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