Atmos. Meas. Tech. Discuss., 5, 1355–1379, 2012
www.atmos-meas-tech-discuss.net/5/1355/2012/
doi:10.5194/amtd-5-1355-2012

Atmospheric
Measurement
Techniques
Discussions

First intercalibration of column-averaged methane from the Total Carbon Column Observing Network and the Network for the Detection of Atmospheric Composition Change

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Received: 15 January 2012 – Accepted: 7 February 2012 – Published: 13 February 2012

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

Abstract

We present the intercalibration of dry-air column-averaged mole fractions of methane (XCH_4) retrieved from solar FTIR measurements of the Network for the Detection of Atmospheric Composition Change (NDACC) in the mid-infrared (MIR) versus near-infrared (NIR) soundings from the Total Carbon Column Observing Network (TCCON). The study uses multi-annual quasi-coincident MIR and NIR measurements from the stations Garmisch, Germany (47.48° N, 11.06° E, 743 m a.s.l.) and Wollongong, Australia (34.41° S, 150.88° E, 30 m a.s.l.).

Direct comparison of the retrieved MIR and NIR time series shows a phase shift in XCH_4 seasonality, i.e. a significant time-dependent bias leading to a standard deviation (stdv) of the difference time series (NIR-MIR) of 8.4 ppb. After eliminating differences in a prioris by using ACTM-simulated profiles as a common prior, the seasonalities of the (corrected) MIR and NIR time series agree within the noise (stdv = 5.2 ppb for the difference time series). The difference time series (NIR-MIR) do not show a significant trend. Therefore it is possible to use a simple scaling factor for the intercalibration without a time-dependent linear or seasonal component. Using the Garmisch and Wollongong data together, we obtain an overall calibration factor MIR/NIR = 0.9926(18). The individual calibration factors per station are 0.9940(14) for Garmisch and 0.9893(40) for Wollongong. They agree within their error bars with the overall calibration factor which can therefore be used for both stations.

Our results suggest that after applying the proposed intercalibration concept to all stations performing both NIR and MIR measurements, it should be possible to obtain one refined overall intercalibration factor for the two networks. This would allow to set up a harmonized NDACC and TCCON XCH_4 data set which can be exploited for joint trend studies, satellite validation, or the inverse modeling of sources and sinks.

1 Introduction

Atmospheric methane(CH_4) has become one of the so-called Kyoto gases since it causes a considerable contribution (0.48 Wm^{-2}) to the total anthropogenic radiative forcing of 2.43 Wm^{-2} (Forster et al., 2007). In addition, CH_4 has an indirect greenhouse effect of 0.13 Wm^{-2} by forming tropospheric ozone, stratospheric water vapor, and other infrared-active trace gases (Lelieveld et al., 1998). The main methane sources are natural wetlands, biomass burning and anthropogenic activities like livestock breeding, rice cultivation, or usage of fossil fuels. Global emissions are about 515 Tg per year (Patra et al., 2011), whereof 60–70 % are anthropogenic (Denman et al., 2007). The major sink of methane is the destruction by hydroxyl radicals (OH), which contributes to about 90 % to the methane loss in the atmosphere. Other sinks are the uptake of methane by soils or the reaction with chlorine radicals (Denman et al., 2007).

Since the beginning of the industrialization, methane concentrations in the atmosphere have more than doubled (e.g. Etheridge et al., 1998). However, there was a period of a near-zero growth at the beginning of this century (Dlugokencky et al., 2003; Bousquet et al., 2006), and after 2006 the atmospheric methane concentration started to increase again (Rigby et al., 2008; Dlugokencky et al., 2009). The increase for the years 2007–2008 has been quantified, and possible causes discussed (e.g. Bousquet et al., 2011; Frankenberg et al., 2011). More recently, it has been shown via ground-based FTIR methane column measurements that the renewed increase after 2006 has been ongoing for about 5 years until today (end of 2011) with a rate of ≈5 ppb yr^{-1} above northern mid-latitudes (Sussmann et al., 2011a).

Ground-based column measurements of methane are complementary to in situ measurements. They are representative of a much wider area while in situ measurements can represent a specific location or biome. In situ measurements are more directly traceable to calibration standards while ground-based column measurements provide the same quantity as satellites measure and are therefore preferred for satellite validation. Measured methane columns are impacted by the varying stratospheric contribution while the interpretation of surface measurements to infer sources and sinks requires additional (model) information on vertical boundary layer transport. There are two established global networks performing ground-based measurements of column-integrated methane. Within the Network for the Detection of Atmospheric Composition Change (NDACC, http://www.ndacc.org) solar FTIR measurements in the mid infrared (MIR) have been performed for about two decades (currently 22 stations). Retrievals of methane from NDACC-MIR spectra have been used for trend studies (Angelbratt et al., 2011; Sussmann et al., 2011a) and satellite validation (e.g. Sussmann et al., 2005).

Since 2004 the NDACC has been complemented by the Total Carbon Column Observing Network (TCCON, http://www.tccon.caltech.edu/), which is dedicated to high-precision retrievals of climate gases (CO_2, CH_4, N_2O) from solar absorption spectra in the near-infrared (NIR) spectral region (Wunch et al., 2011a). Column-averaged dry-air mole fractions are retrieved by scaling an a priori profile to provide the best fit to the measured spectra, and, finally, by dividing these columns by the simultaneously-retrieved O_2 column. The TCCON has been used for the validation of models (Houweling et al., 2010) and satellite measurements of methane (e.g. Morino et al., 2011; Schneising et al., 2012), but also for deriving information on sources and sinks of greenhouse gases (e.g. Wunch et al., 2009; Chevallier et al., 2011). The TCCON measurements are calibrated against the World Meteorological Organization (WMO) in situ trace gas measurement scales, using profiles obtained by aircraft in-situ measurements flown over TCCON sites (Deutscher et al., 2010; Wunch et al., 2010; Messerschmidt et al., 2011; Geibel et al., 2011). Currently, there are 15 operational TCCON stations, most of which have been established during the last couple of years.

Accurate knowledge of the intercalibration relation between these two networks is important as it provides the possibility of combining data from both networks and thereby attaining a wider spatial and a longer temporal coverage than they would provide independently. This is not only an advantage for satellite validation but also provides the opportunity for trend analysis dating back 15 years before TCCON operations began. It is, therefore, the goal of this study to establish the NDACC-TCCON intercalibration for XCH_4. An important question in this context is whether or not one overall intercalibration factor for all stations can be found and quantified, or whether a time-dependent intercalibration parameterization, with a significant linear and/or seasonal component, is necessary.

Our paper is structured as follows: After introducing the participating FTIR sites and their measurement settings in Sect. 2 along with the MIR and NIR retrieval strategies, we describe our intercalibration method (Sect. 3). The results of the intercalibration are shown in Sect. 4, and Sect. 5 gives the conclusions and an outlook.

2 Ground-based sounding of columnar methane in the MIR and NIR

2.1 Garmisch FTIR soundings

The Garmisch solar FTIR system (47.48° N, 11.06° E, 743 m a.s.l.) is operated by the group “Variability and Trends” at the Institute for Meteorology and Climate Research, Karlsruhe Institute of Technology, Germany. It was set up in 2004 as part of the TCCON network and is based on a Bruker IFS125HR interferometer and took part in the European IMECC aircraft calibration campaign (Messerschmidt et al., 2011; Geibel et al., 2011). Column-averaged methane is retrieved from single-scan measurements in the NIR (see Table 1 for the spectral micro-windows) recorded with an InGaAs diode using a maximum optical path difference of 45 cm. The FTIR system also performs NDACC-type measurements in the MIR (Table 1) in alternating mode with the NIR measurements. The interferograms for the MIR methane retrievals were recorded with an InSb detector using an optical path difference of typically 175 cm. Six scans were averaged with an integration time of approximately seven minutes. Data obtained with the Garmisch FTIR have been used for satellite validation (de Laat et al., 2010; Morino et al., 2011; Wunch et al., 2011b), carbon cycle research (Chevallier et al., 2011), and studies of atmospheric variability and trends (e.g. Borsdorff and Sussmann, 2009; Sussmann et al., 2011b). The intercalibration uses the Garmisch time series between

July 2007–November 2010 which comprises 2302 MIR spectra and 24 417 NIR spectra.

2.2 Wollongong FTIR soundings

The Wollongong solar FTIR system (34.41° S, 150.88° E, 30 m a.s.l.) was set up in 1995 as part of the NDACC network. It is operated by the Center for Atmospheric Chemistry at the University of Wollongong, Australia. From 1995 to 2007 a Bomem DA8 FTIR system was operated (Griffith et al., 1998). It was replaced in 2007 with a Bruker IFS 125HR instrument to render measurements in both the MIR and the NIR spectral ranges possible (Jones et al., 2012; Wunch et al., 2011a). For this study only the Bruker data were used. Spectra in the MIR range are recorded with an InSb detector, using an optical path difference of 257 cm and averaging two successive scans with an integration time of approximately four minutes. The settings for the NIR measurements are identical to those at Garmisch. The intercalibration uses the Wollongong time series between June 2008–April 2010 which comprises 862 MIR spectra and 10 732 NIR spectra.

2.3 MIR and NIR retrieval strategies

For the retrieval of XCH_4 from NDACC-type MIR measurements the retrieval strategy MIR-GBM v1.0 (Sussmann et al., 2011b) is used in this study along with the spectral fitting software SFIT2 ver. 3.94 (Pougatchev et al., 1995). The basic features of MIR-GBM v1.0 are given in Table 1. TCCON-type NIR measurements are analyzed with the spectral fitting software GFIT ver. 4.4.10 (release ggg_20091107). The basic features of GFIT are given in Table 1, more details can be found in Wunch et al. (2010).

3 Intercomparison method

3.1 Eliminating a priori impact

Any quantitative comparison of two different remote sounders is complicated because in general they contain a differing a priori impact (differing averaging kernels and a priori profiles) influencing the retrieved trace gas column amounts. According to Rodgers (2000) this can be taken into account by an adjustment of the soundings for a common a priori profile which eliminates the differences relating to smoothing and a priori information. To obtain this common basis we use modeled mole-fraction profiles x_{mod} and obtain corrected column-averaged mole fractions c_{cor} for the MIR or NIR soundings which can be directly compared:

$$c_{\mathrm{cor}} = \hat{c} + \frac{1}{p_0} \sum_{l} \left(1 - A^l\right) \left(x^l_{\mathrm{mod}} - x^l_a\right) \Delta p^l . \tag{1}$$

Here c represents the column-averaged mole fraction of methane retrieved from MIR or NIR spectra. For every model layer l the difference between 1 (i.e. the ideal averaging kernel) and the total column averaging kernel A^l in this layer is multiplied with the difference between the common a priori mole fraction x^l_{mod} and the FTIR (MIR or NIR) a priori mole fraction x^l_a as well as with the pressure difference between the upper and lower boundaries of layer l; p_0 denotes the surface pressure.

Obviously, this correction can be neglected in cases of the averaging kernel being close to ideal or the a priori profile x_a being close to the modeled profile x_{mod}. This approach has been applied recently for the comparison of carbon dioxide and methane columns measured by SCIAMACHY to ground-based FTIR measurements and to model results (Reuter et al., 2011; Schneising et al., 2012).

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3.2 Description of the a priori model

The model used for eliminating the a priori contribution according to Eq. (1) is the CCSR/NIES/FRCGC AGCM-based chemistry-transport model (i.e. ACTM), which has been developed for simulating the major long-lived greenhouse gases (Patra et al., 2009). The ACTM simulations are conducted at T42 spectral truncations in the horizontal (≈2.8 × ≈ 2.8° latitude-longitude) and 67 vertical levels covering the height range from the earth's surface to the mesosphere ($\approx 1.3 \times 10^{-5}\,\sigma$ pressure or ≈80 km). The emissions and loss of methane in ACTM are adopted from the TransCom-CH_4 simulation protocol (Patra et al., 2011). Comparisons showed that forward ACTM simulations of annual-mean methane are in close agreement (within 1 ppb) with measurements from surface sites as to inter-hemispheric gradients (Patra et al., 2011). ACTM-simulated vertical profiles of dry-air mole fractions on the native model vertical grid and nearest horizontal grid of the FTIR sites are sampled at 3-hourly intervals for use as a priori in this study.

4 MIR-NIR intercalibration

4.1 Qualitative effect of correcting for a priori contribution

Figure 1 shows the time series of column-averaged methane above the Garmisch FTIR site retrieved by MIR and NIR spectrometry. In addition the figure shows both time series after applying the correction to a common a priori as described in Sect. 3.1. It can be seen that the correction affects the MIR measurements in a different way than the NIR measurements. This is because of the differing averaging kernels. The MIR averaging kernels are plotted in Fig. A1 of Sussmann et al. (2011b) and the NIR averaging kernels are displayed in Wunch et al. (2010, Fig. 3 therein).

The effect of correcting for a priori impact is that the seasonalities of the MIR and NIR time series converge. This can be seen from comparing Fig. 2 with Fig. 3: for the uncorrected time series Fig. 2 shows a significant seasonal bias. The standard deviation (stdv) of the difference time series XCH_4(NIR) – XCH_4(MIR) is 8.4 ppb in this case. After the correction to a common a priori (Fig. 3) the difference between NIR and MIR is reduced mainly to one time constant bias, and the NIR and MIR seasonalities agree within the noise (stdv of difference time series reduced to 5.2 ppb).

Another finding from analyzing the difference time series XCH_4(NIR) – XCH_4(MIR) is that they do not show a significant trend. See Fig. 3 (upper trace) for the example of Garmisch data, and Table A1 for derived numbers on trend and uncertainty. The same result is found for Wollongong (Table A1).

The bias between the MIR and NIR data is in the order of 1 % (see Sect. 4.2 for details) and this can be attributed to the differing spectroscopy used in the NIR (Frankenberg et al., 2008; Rothmann et al., 2009) and MIR (Rothmann et al., 2003), respectively; i.e. differing integrated line strengths. Note, a typical column uncertainty of ≈1 % for TCCON measurements due to spectroscopy errors has been found in earlier work and this is the reason why TCCON measurements are calibrated against aircraft profile measurements which can be traced back to the WMO in-situ trace-gas measurement scale (Wunch et al., 2010).

4.2 Intercalibration

Since the TCCON-type NIR soundings of XCH_4 have been calibrated to the WMO scale using aircraft profiles (Wunch et al., 2010; Geibel et al., 2011), the NIR soundings are used as reference, and the MIR column-averaged mole fractions are scaled to fit the NIR.

In prolongation of the result from Sect. 4.1 we investigate the question of whether one common intercalibration factor can be applied for our two test stations. In Fig. 4 monthly means of the MIR time series for Wollongong and Garmisch (both corrected for a priori impact) are plotted versus the corresponding NIR monthly means (also corrected for a priori impact). Assuming a linear relationship between the MIR and NIR soundings of both stations together, a linear-least squares fit with a forced zero intercept has been be performed. The resulting intercalibration factor is 0.9926(18) (Table 2). Besides this common intercalibration shown in Fig. 4 we have also determined the intercalibration relations individually for Garmisch (0.9940(14)) and Wollongong (0.9893(40)). Since the individual intercalibration factors of the two stations agree within error bars we conjecture that the common intercalibration factor (0.9926(18)) can be used for both Garmisch and Wollongong.

For reference, Table 3 provides calibration factors derived in an analogous way from the time series as retrieved, i.e. without correcting to a common a priori before. Note, the calibration factors are different from Table 2. Also, the uncertainty on the calibration factor for Garmisch is significantly increased relative to Table 2. This is because the northern hemisphere site Garmisch shows a pronounced (minus sine type) seasonality which is retrieved differently by the NIR and MIR sounders because of the differing a priori contribution. Therefore, the series do not fit together well without the a priori correction (see Fig. 1).

5 Conclusions and outlook

In this study we introduced a concept for the intercalibration of soundings of column-averaged methane performed by the NDACC (MIR) and the TCCON (NIR) networks, respectively. The concept was demonstrated using multi-annual data from the stations Garmisch (47° N) and Wollongong (34° S), both performing quasi-coincident MIR and NIR measurements with the same instrument. As a basis for the determination of the intercalibration relation we showed that it is possible to correct the time series to a common a priori profile in order to eliminate differences resulting from differing a priori information. In particular the a priori impact is critical to retrieved seasonality, i.e. it turned out that the pronounced northern hemisphere seasonal cycle of XCH_4 is retrieved differently by NIR and MIR soundings. We presume that the same holds true for latitudinal artifacts due to a priori impact.

After elimination of prior impact the seasonalities of the MIR and NIR soundings of column-averaged methane agree within the noise. Also, the difference time series (NIR-MIR) do not show a significant trend. I.e. no time-dependent intercalibration with linear or seasonal components is required and it is possible to use one single scaling factor for the intercalibration. We found station-to station consistency, i.e. the intercalibration factors derived for the individual stations Garmisch and Wollongong agree within their error bars. Therefore, it is possible to use one common intercalibration factor for the multi-annual MIR and NIR soundings of the stations Garmisch and Wollongong, i.e. MIR/NIR = 0.9926(18).

Finally, the simple linear intercalibration found in this work allows to obtain a harmonized NDACC (MIR) and TCCON (NIR) data set for column-averaged methane which can be exploited for joint trend studies, satellite validation or the inverse modeling of sources and sinks.

In future work we will apply the concept introduced in this study to all other existing stations that perform coincident MIR and NIR soundings of column-averaged methane. The goal is to further refine the intercalibration relation found in this work.

Acknowledgements. We thank H. P. Schmid (IMK-IFU) for his continual interest in this work. Provision of the GFIT code by G. Toon (JPL) as well as the SFIT software and WACCM profiles by J. Hannigan (NCAR) is gratefully acknowledged. Our work has been performed as part of the ESA GHG-cci project via subcontract with University of Bremen. In addition we acknowledge funding by the EC within the INGOS project. We thank for support by Deutsche Forschungsgemeinschaft and Open Access Publishing Fund of Karlsruhe Institute of Technology. The Wollongong work was funded through the Australian International Science Linkage grant CG130014 and the Australian Research Council, grants DP0879468 and DP110103118.

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Table 1. Strategies for retrieval of column-averaged methane from MIR and NIR solar spectra.

	MIR	NIR
micro windows (interfering species fitted)	2613.70–2615.40 (HDO, CO_2) 2835.50–2835.80 (HDO) 2921.00–2921.60 (HDO, H_2O, NO_2)	5880.00–5996.00 (CO_2, H_2O, HDO) 5996.45–6007.55 (CO_2, H_2O, HDO) 6007.00–6145.00 (CO_2, H_2O, HDO)
line list	HITRAN 2000 including 2001 update release	HITRAN 2008 including update by Frankenberg et al. (2008)
retrieval constraint	Tikhonov L_1, regularization strength α optimized via L-curve/minimum diurnal variation (≈2 degrees of freedom for signal); altitude constant on per-cent-vmr[1] scale	scaling of a methane a priori profile
a priori vmr profiles	WACCM[2] (1 fixed profile)	generated from MkIV FTS balloon profiles (1 fixed profile)
background fit	linear slope	linear slope
retrieval quality selection	threshold (0.15 %) for rms-noise/dofs[3]	fractional var. in solar intensity (0.0–20.0 %) XCH_4 (0.0–2.0e-6) XCH_4 error (0.0–1.0e-4) SZA[4] (0.0–85°)
calculation of column-averaged dry-air mole fractions	use 4-times-daily-NCEP[5] PTU profiles, calculate air column, subtract water vapor column	use simultaneously measured O_2 column
precision (1-σ diurnal variation)	<0.3 %	<0.3 %
seasonal bias (H_2O/HDO-CH_4 interference error[6])	<0.14 %	hitherto undetermined
references	Sussmann et al. (2011b)	Wunch et al. (2010)

[1] vmr – volume mixing ratio; [2] WACCM – Whole Atmosphere Chemistry Climate Model; [3] dofs – degrees of freedom for signal; [4] SZA – solar zenith angle; [5] NCEP – National Center for Environmental Prediction; [6] see Sussmann and Borsdorff (2007) for a definition.

Table 2. MIR-NIR intercalibration factors (slope MIR/NIR) if both data sets are corrected to a common a priori according to Eq. (1).

data set	calibration factor	error (3 σ)
Garmisch + Wollongong	0.9926	0.0018
Garmisch	0.9940	0.0014
Wollongong	0.9893	0.0040

Table 3. MIR-NIR intercalibration factors (slope MIR/NIR) if both data sets are used as retrieved, i.e. without correcting to a common a priori.

data set	calibration factor	error (3 σ)
Garmisch + Wollongong	0.9959	0.0019
Garmisch	0.9961	0.0023
Wollongong	0.9955	0.0034

Table A1. Trend analysis of the XCH_4 difference time series (NIR-MIR), obtained after the NIR and MIR time series have been corrected to a common a priori. See Fig. 3 (upper trace) for the underlying Garmisch data.

data set	time period	trend NIR-MIR (ppb yr^{-1})	2-σ trend uncertainty (ppb yr^{-1})	significant trend (95 % confidence)?
Garmisch	Jul 2007–Nov 2010	+1.45	±1.62	no
Wollongong	Jun 2008–Apr 2010	+1.04	±7.72	no

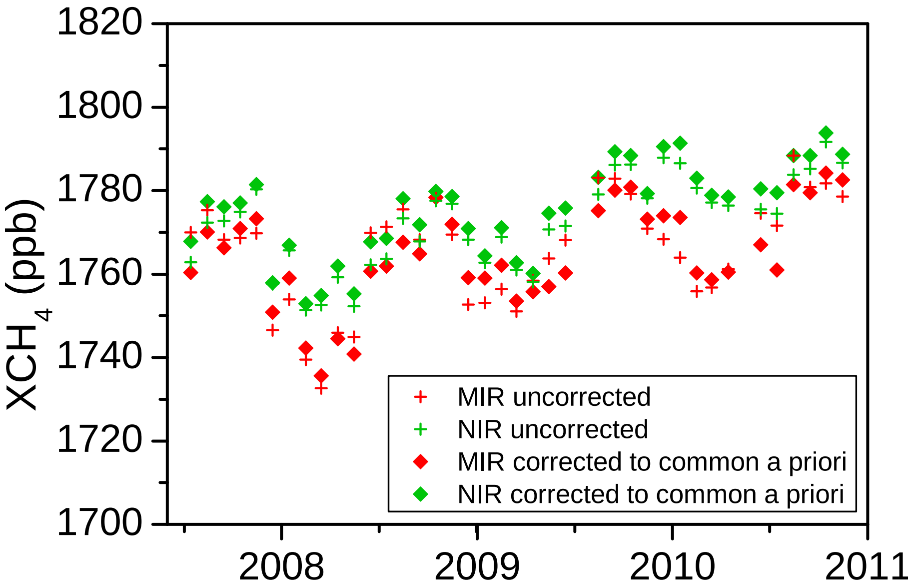

Fig. 1. Monthly-mean time series of column-averaged methane retrieved from MIR and NIR spectral measurements at Garmisch, before and after applying the correction to a common a priori.

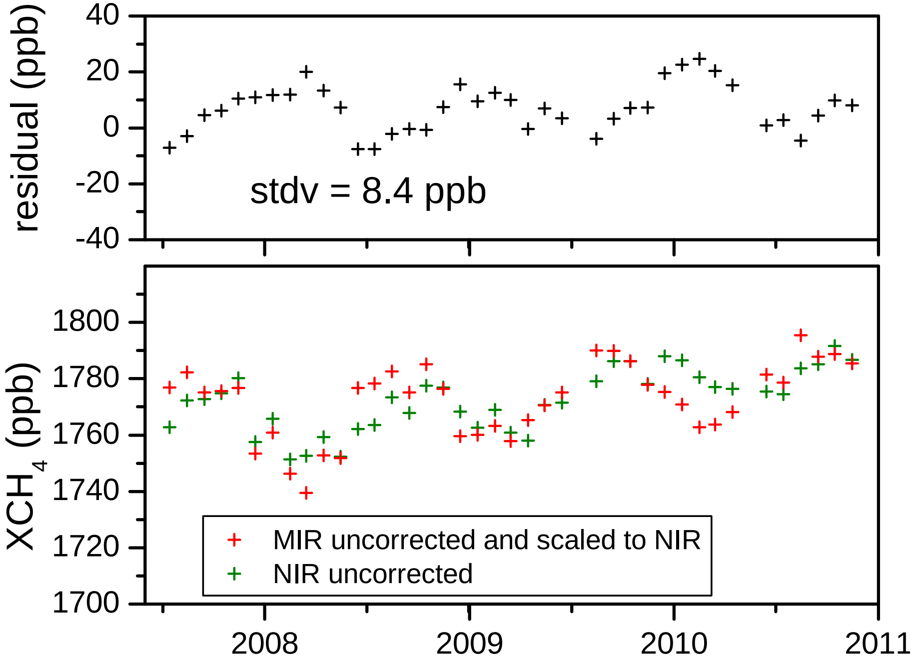

Fig. 2. Lower panel: monthly-mean MIR and NIR time series for Garmisch. Both column series are plotted as retrieved, i.e. no correction for a priori impact according to Eq. (1) has been performed. To visualize the disagreement of the seasonalities the MIR data have then been scaled by 1/0.9961 to match the average of the NIR data. Upper panel: residual time series, i.e. difference time series of the NIR and MIR data shown in the lower trace.

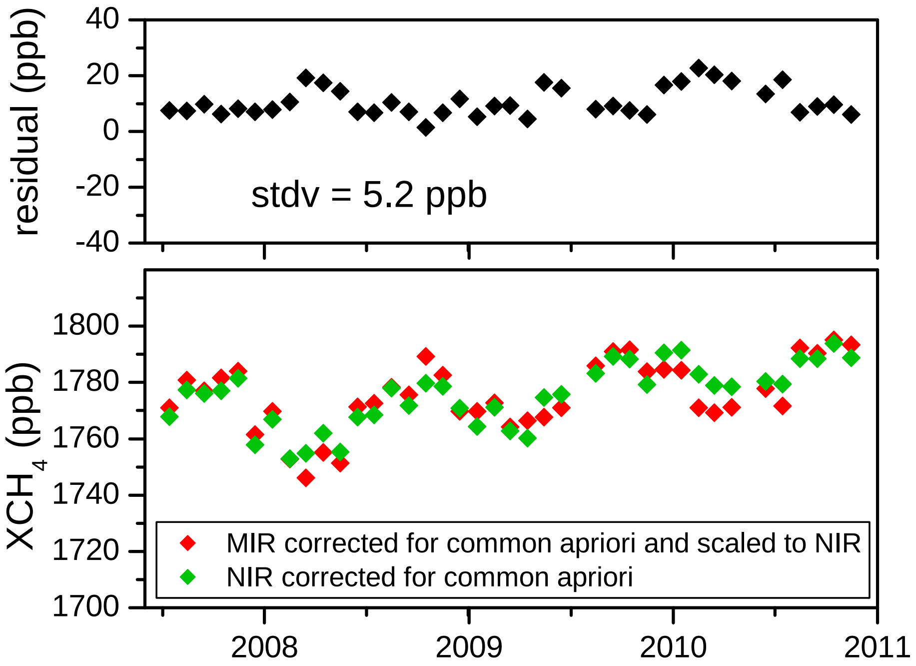

Fig. 3. Lower panel: monthly-mean MIR and NIR time series for Garmisch. Both column series have been corrected for a priori impact according to Eq. (1). To visualize the agreement of the seasonalities the MIR data have then been scaled by 1/0.9940 to match the average of the NIR data. Upper panel: Residual time series, i.e. difference time series of the NIR and MIR data shown in the lower trace.

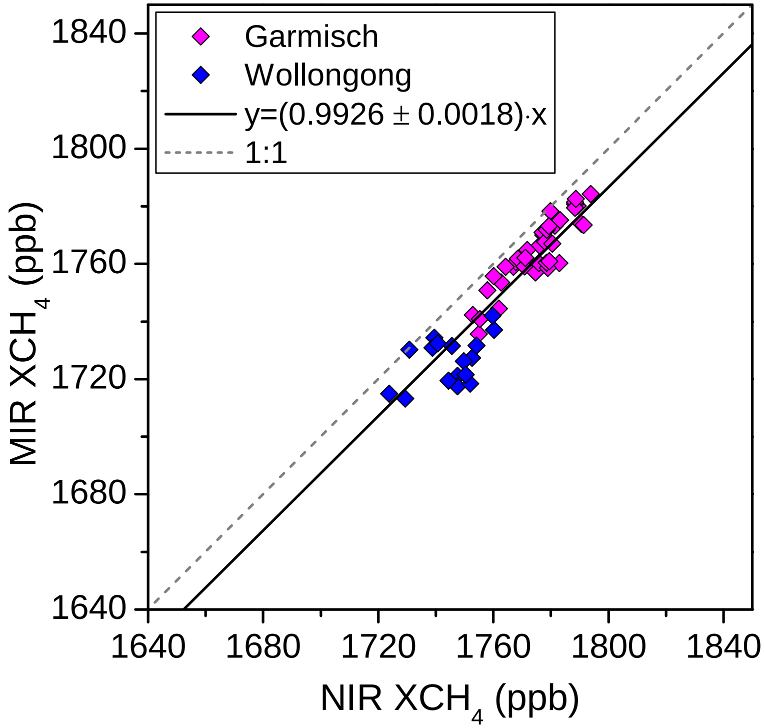

Fig. 4. MIR-NIR intercalibration. The black line is a linear fit to all monthly-mean data from both stations. The slope error is for 3-σ confidence. Only months with >5 measurements have been included.