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Renewed methane increase for five years (2007–2011) observed by solar FTIR spectrometry

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Abstract. Trends of column-averaged methane for the time period (1996, Sep 2011) are derived from the mid-infrared (mid-IR) solar FTIR time series at the Zugspitze (47.42° N, 10.98° E, 2964 m a.s.l.) and Garmisch (47.48° N, 11.06° E, 743 m a.s.l.). Trend analysis comprises a fit to the deseasonalized time series along with bootstrap resampling for quantifying trend uncertainties. We find a positive trend during [1996, 1998] of 9.0 [3.2, 14.7] ppb yr^{-1} for Zugspitze (95 % confidence interval), an insignificant growth during [1999, mid 2006] of 0.8 [−0.1, 1.7] ppb yr^{-1} (Zugspitze), and a significant renewed increase during [mid 2006, Sep 2011] of 5.1 [4.2, 6.0] ppb yr^{-1} for Garmisch, which is in agreement with 4.8 [3.8, 5.9] ppb yr^{-1} for Zugspitze.

The agreement of methane trends at the two closely neighboring FTIR sites Zugspitze and Garmisch within the uncertainties indicates a good station-to-station consistency as a basis for future trend analyses by the ground-based mid-IR FTIR network on the global scale. Furthermore, the Zugspitze FTIR trend for the time interval [Jul 2006, Jun 2009] is found to agree with the trend derived from SCIA-MACHY (WFM-DOAS v2.0.2) data within the 95 % confidence intervals. In case a 1000-km pixel selection radius around the Zugspitze is used, the confidence interval is narrower for the FTIR trend (6.9 [4.2, 9.5] ppb yr^{-1}) compared to SCIAMACHY (7.1 [5.1, 8.6] ppb yr^{-1}). If, however, a loosened pixel selection is used (≈1000-km half-width latitudinal band), the SCIAMACHY trend significance interval is narrower (6.8 [5.1, 8.6] ppb yr^{-1}) compared to Zugspitze FTIR (5.7 [3.0, 8.3] ppb yr^{-1}).

While earlier studies using surface network data revealed changes of 8.0 ± 0.6 ppb in 2007, 6.4 ± 0.6 ppb in 2008, and 4.7 ± 0.6 ppb in 2009 (Dlugokencky et al., 2011), our updated result proves that the renewed methane increase meanwhile has been persisting for >5 years [mid 2006, Sep 2011]. This is either the longest and largest positive trend anomaly since the beginning of systematic observations more than 25 years ago or the onset of a new period of strongly increasing CH$_4$ levels in the atmosphere. Several scenarios have been developed to explain the persistent increase observed, mainly invoking an increase in emissions from natural wetlands, an increase in fossil fuel-related emissions or a decrease in OH concentrations. However, more work is needed to fully attribute this increase to a particular source or sink.

1   Introduction

The molecular symmetry of methane (CH$_4$) allows for highly active vibration-rotation excitation by infrared absorption. Carbon dioxide (CO$_2$) is also infrared-active, however, its strongest absorption lines are located outside the atmospheric window. These are basic reasons why methane is the second important anthropogenic greenhouse gas in spite of its still relatively small abundance in the atmosphere.

Methane concentrations in the atmosphere have more than doubled since the beginning of industrialization (Forster et al., 2007). After a period of near-zero growth at the beginning of this century (Dlugokencky et al., 2003; Bousquet et al., 2006), the growth rate of atmospheric methane started to increase strongly again after 2006 (Rigby et al., 2008; Dlugokencky et al., 2009). The attribution of this increase to a particular source or sink is still debated in the scientific

community (Rigby et al., 2008; Dlugokencky et al., 2009; EDGAR, 2012; Bloom et al., 2010; Ringeval et al., 2010; Aydin et al., 2011; Bousquet et al., 2011; Spahni et al., 2011).

The finding of an atmospheric increase since 2007 is based on highly precise measurements ($\approx 0.2\%$) of the methane mixing ratio by atmospheric networks at the surface and in the planetary boundary layer (WMO, 2012). Satellite retrievals (Crevoisier et al., 2009; Frankenberg et al., 2011; Schneising et al., 2011, 2012) complement these surface observations with a higher spatial coverage, but at the cost of lower precisions ($\approx 2\%$). Several studies have focused on how column observations can complement surface networks and help further reduce uncertainties associated with sources and sinks in atmospheric inversions (e.g., Olsen and Randerson, 2004 or Bergamaschi et al., 2009, and references therein). Columnar methane retrieved from ground-based Fourier-transform-infrared (FTIR) spectrometry in the mid-infrared (mid-IR) provides a column view of atmospheric methane at a high precision close to that attainable by surface networks ($\approx 0.3\%$, Sussmann et al., 2011). Hence such ground-based observations are very useful to document atmospheric methane changes. In this paper, we investigate the persistence of the 2007–2008 anomaly by an updated trend analysis including FTIR data until today (fall 2011) and report possible causes of the increase observed.

After describing the mid-IR sounding technique (Sect. 2), methane time series and trends will be presented in Sect. 3.1, and the trends from the neighboring sites of Zugspitze and Garmisch will be compared to each other as well as to SCIAMACHY trends derived with the same analysis method (Sect. 3.2). Section 4 will provide a discussion of the trend behavior derived for the 2007–2011 time period along with some conclusions.

2   Sounding technique

Time series of column-averaged dry-air mole fractions of methane (XCH$_4$) were retrieved from long-term FTIR solar absorption measurements at two northern mid-latitude sites, namely, the Zugspitze high-altitude site ($47.42°$ N, $10.98°$ E, 2964 m a.s.l.) and Garmisch ($47.48°$ N, $11.06°$ E, 743 m a.s.l.). The two sites are located only a few kilometers apart in horizontal distance. However, the levels of integrated water vapor differ strongly (min/max $= 0.2$ mm/12.7 mm for Zugspitze, 1.9 mm/34.9 mm for Garmisch) because of the altitude difference.

The Zugspitze FTIR system has been conducting continuous measurements since 1995 as part of the Network for the Detection of Atmospheric Composition Change (NDACC, http://www.ndacc.org). It is operated by the Group "Variability and Trends" of IMK-IFU[1], Karlsruhe Institute of Tech-

nology, at the Zugspitze[2]. The FTIR system is based on a Bruker IFS125HR interferometer; details can be found in Sussmann and Schäfer (1997). The interferograms for the methane retrievals were recorded with an InSb detector using an optical path difference of typically 175 cm. 6 scans were averaged (≈ 7-min integration time). The pressure-temperature profiles necessary for the inversion were taken from the National Center for Environmental Prediction (NCEP) automailer. The Garmisch solar FTIR system was set up in 2004 at the Garmisch site[3] and is part of the Total Carbon Column Observing Network (TCCON, http://www.tccon.caltech.edu/) operating in the near-infrared for high-precision retrieval of column-averaged mixing ratios of carbon dioxide and methane. The system performs mid-IR NDACC-type measurements in parallel (in alternating mode on the time scale of several minutes). The latter are utilized for this study. The measurement settings for the Garmisch mid-IR methane measurements are the same as for the Zugspitze.

Retrievals of columnar methane from ground-based FTIR spectrometry in the mid-IR were used previously for trend studies (Zander et al., 1989; Angelbratt et al., 2011) and satellite validation (e.g., Sussmann et al., 2005). However, it became obvious only recently that the methane-column retrievals at high-humidity (low-altitude) sites may be dominated by water vapor (H_2O/HDO)-CH_4 interference errors of up to 5 % (Sussmann et al., 2011). Consequently, artifacts dominated the seasonal cycle of methane retrieved with the standard NDACC retrieval strategy. Dry (high-altitude) sites were not affected. Note that water vapor interference also perturbed methane retrievals from SCIAMACHY (Frankenberg et al., 2008). A general formulation of the (water vapor) interference problem was given before (Sussmann and Borsdorff, 2007) and water vapor can be quantified by the FTIR technique itself (e.g., Sussmann et al., 2009). To improve the standard methane retrieval strategy used within NDACC, we recently developed a new retrieval strategy (MIR-GBM v1.0) using systematically selected spectral micro windows and spectroscopic parameters (Sussmann et al., 2011). MIR-GBM v1.0 was shown to eliminate H_2O/HDO-CH_4 interference errors down to the $\approx 0.1\%$ level (i.e., 0.14 % for the wettest test site and 0.10 % and 0.02 %, respectively, for the two dryer test sites). For the first time, a good agreement of the retrieved northern-hemisphere seasonal cycle with SCIAMACHY results was achieved (Sussmann et al., 2011).

In this paper we apply MIR-GBM v1.0 to a trend analysis. The basic features of this retrieval strategy are given in Table 1, more details can be found in Sussmann et al. (2011).

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[2] Zugspitze site details can be found at http://www.imk-ifu.kit.edu/311.php

[3] Garmisch site details can be found at http://www.imk-ifu.kit.edu/315.php

Table 1. Characteristics of the strategy for retrieval of column-averaged methane from mid-IR solar spectra (MIR-GBM v1.0). For more details, see Sussmann et al. (2011).

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)
line list	HITRAN 2000, including 2001 update release
retrieval constraint	Tikhonov L_1
regularization strength α	optimized via L-curve/minimum diurnal variation (dofs[a] ≈ 2)
altitude dependency of reg. strength	altitude constant on per-cent-vmr[b] scale
a priori vmr profiles	WACCM[c]
background fit	linear slope
retrieval quality selection	threshold (0.15 %) for rms-noise/dofs
calculation of column-averaged dry-air mole fractions	use 4-times-daily-NCEP[d] PTU profiles, interpolate to FTIR measurement time, calculate air column, subtract water vapor column
precision (1-σ diurnal variation)[e]	< 0.3 %
seasonal bias (H_2O/HDO-CH_4 interferenceerror)	< 0.14 %

[a] dofs – *d*egrees *o*f *f*reedom *f*or *s*ignal
[b] vmr – *v*olume *m*ixing *r*atio
[c] WACCM – *W*hole *A*tmosphere *C*hemistry *Cl*imate *M*odel
[d] NCEP – *N*ational *C*enter for *E*nvironmental *P*rediction
[e] for 7-min integration

3 Trend results

3.1 Time series and trend analysis

Figure 1a shows the time series of monthly-mean column-averaged methane above Zugspitze and Garmisch. Typically, $n = 40$–60 individual column measurements per month are recorded on 6–8 clear sky days. Shaded bars in Fig.1a indicate the statistical uncertainties of the monthly means calculated from the individual measurements ($\pm 3\sigma/sqrt(n)$). These suggest that statistically stable monthly means are obtained for the trend analysis.

The de-seasonalized time series (Fig. 1b) shows a significant increase during the 3-years' time interval [1996, 1998], followed by a period with no significant growth [1999, mid 2006]. Afterwards, a renewed strong increase can be observed, which has been persisting for 5 years now [mid 2006, Sep 2011]. The division into 3 time periods is in agreement with dedicated trend change points defined and used for methane in earlier work (Dlugokencky et al., 2003, 2009; Angelbratt et al., 2011). Our trend analysis is based on the approach described in Gardiner et al. (2008). See Table 2 for the resulting figures. Briefly, the approach augments a basic linear trend model applied to the indicated parts of the time series after subtraction of a fitted intra-annual function and uses least squares regression in conjunction with a boot-

strap resampling of the residuals in order to determine confidence limits associated with the trend estimates. For the intra-annual model, a 3rd-order Fourier series is used.

As a main result from Table 2, the original anomaly of the period [mid 2006, 2008] with a Zugspitze trend of 6.6 [3.5, 9.8] ppb yr^{-1} and a Garmisch trend of 5.1 [2.0, 8.3] ppby yr^{-1}) is found to persist also over the extended period [mid 2006, Sep 2011] investigated now (Zugspitze trend 4.8 [3.8, 5.9] ppb yr^{-1}, Garmisch trend 5.1 [4.2, 6.0] ppb yr^{-1}).

3.2 Station-to-station consistency and comparison to SCIAMACHY trends

The FTIR trends retrieved for the neighboring sites of Zugspitze and Garmisch (Table 2) are in very good agreement: For the [mid 2006–2008] period, a trend of 6.6 ppb yr^{-1} with a 95 % confidence interval of [3.5, 9.8] ppb yr^{-1} is found above Zugspitze, which agrees with 5.1 [2.0, 8.3] ppb yr^{-1} retrieved from Garmisch measurements for the same period. For the [mid 2006–Sep 2011] period, the trend of 4.8 [3.8, 5.9] ppb yr^{-1} above Zugspitze is close to the Garmisch trend of 5.1 [4.2, 6.0] ppb yr^{-1}. These numbers indicate a very good station-to-station consistency of the ground-based FTIR trend results.

Table 2. Existence and significance of trends of column-averaged mole fractions of CH_4.

		trend (ppb yr^{-1})	uncertainty interval [2.5th percentile, 97.5th percentile][a] (ppb yr^{-1})	significant non-zero trend? (95 % confidence)
Zugspitze FTIR				
	[1996, 1998]	**9.0**	**[3.2, 14.7]**	**yes**
	[1999, mid 2006]	0.8	[−0.1, 1.7]	no
	[mid 2006, 2008]	**6.6**	**[3.5, 9.8]**	**yes**
	[mid 2006, Sep 2011]	**4.8**	**[3.8, 5.9]**	**yes**
Garmisch FTIR				
	[mid 2006, 2008]	**5.1**	**[2.0, 8.3]**	**yes**
	[mid 2006, Sep 2011]	**5.1**	**[4.2, 6.0]**	**yes**
SCIAMACHY[b] (1000 km radius around Zugspitze)	[Jul 2006, Jun 2009], 23 months available (missing months Nov–Feb 2007, Nov–Feb 2008, Oct–Feb 2009)	**7.1**	**[3.7, 10.4]**	**yes**
Zugspitze FTIR	same 23 months as SCIAMACHY	**6.9**	**[4.2, 9.5]**	**yes**
SCIAMACHY (38.5° N–56.5° N)	[Jul 2006, Jun 2009], i.e., 36 months	**6.8**	**[5.1, 8.6)**	**yes**
Zugspitze FTIR	all 36 months as SCIAMACHY	**5.7**	**[3.0, 8.3]**	**yes**

[a] underlying uncertainty distributions based on 5000 bootstrap resamplings for each trend
[b] WFM-DOAS version 2.0.2

The trend of 6.6 [3.5, 9.8] ppb yr^{-1} derived from our Zugspitze time series for the period [mid 2006, 2008] agrees qualitatively with earlier studies using in-situ data for both hemispheres, according to which the growth rate of atmospheric methane started to increase after 2006 with changes of 8.0 ± 0.6 ppb in 2007, 6.4 ± 0.6 ppb in 2008, and 4.7 ± 0.6 ppb in 2009 (Dlugokencky et al., 2011).

For a quantitative validation of the FTIR trend results, a dedicated intercomparison with SCIAMACHY retrievals of column-averaged mole fractions using the WFM-DOAS algorithm version 2.02 (Schneising et al., 2011) was made, see Table 2. The trend intercomparison was performed for the time interval [Jul 2006, Jun 2009]. For a 1000-km pixel selection radius around the Zugspitze, the SCIAMACHY XCH$_4$ data available cover 23 months (typically, the fall-winter months November to February are missing). For these SCIAMACHY data, the trend and its 95 % confidence interval were analyzed with the same (bootstrap) technique as described for the FTIR trend analysis above. The resulting SCIAMACHY trend is 7.1 [3.7, 10.4] ppb yr^{-1}, see Table 2. For the same 23 months, an FTIR trend of 6.9 [4.2, 9.5] ppb yr^{-1} is obtained. Both trends agree within the error

bars. The FTIR uncertainty interval is narrower than that of SCIAMACHY.

Using a full latitudinal band of $\approx$1000 km half-width centered around the Zugspitze latitude (i.e., covering 38.5° N–56.5° N), we obtained a SCIAMACHY trend of 6.8 [5.1, 8.6] ppb yr^{-1} (Table 2). Note that due to the loosened spatial selection (full latitudinal band), SCIAMACHY retrievals become available for all 36 months of the 3-years' time interval. For the same 36 months, we derived an FTIR trend above the Zugspitze of 5.7 [3.0, 8.3] ppb yr^{-1} which again agrees to the SCIAMACHY trend. As more SCIAMACHY data are applied in the trend analysis in this case, the SCIAMACHY confidence interval is narrower than the FTIR confidence (Table 2). Overall, the XCH$_4$ trends derived from ground-based FTIR and SCIAMACHY are in good agreement.

4 Discussion and conclusion

The methane increase persisting for 5 years [mid 2006, Sep 2011] as documented in Fig. 1 and Table 2 is either the
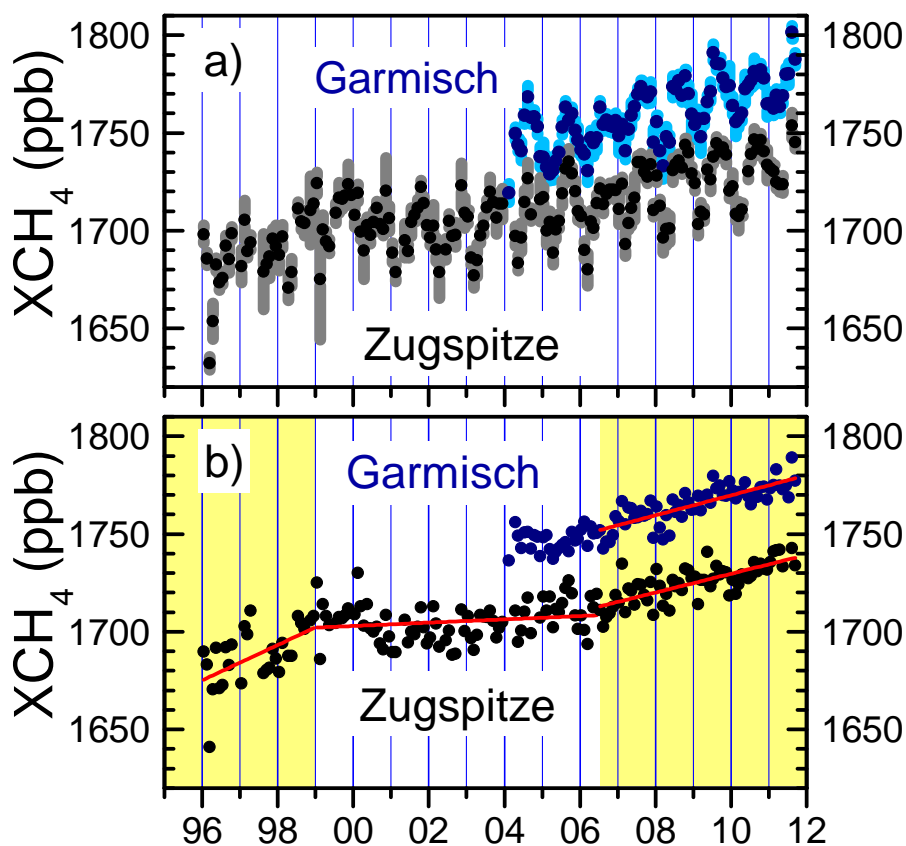
Fig. 1. (a) Time series of methane column-averaged mole fractions above Zugspitze and Garmisch (monthly means). Shaded bars indicate the statistical error of the monthly means calculated from the individual measurements ($\pm 3\ \sigma/sqrt(n)$), where n is the number of FTIR measurements per month. **(b)** De-seasonalized time series and linear trends (red lines). See Table 2 for trend magnitudes and significance.

longest and largest positive trend anomaly since the beginning of systematic observations more than 25 years ago or marks the onset of a new period of strongly increasing CH$_4$ levels in the atmosphere. Our study cannot identify the mechanisms behind this increase, but our findings shall be discussed briefly in relation to previous studies on this subject and most reasonable causes shall be proposed.

Previous studies discussed possible causes of the increase in the years 2007 and 2008 (Rigby et al., 2008; Duglokencky et al., 2009; Bloom et al., 2010; van der Werf et al., 2010; Bousquet et al., 2011; Frankenberg et al., 2011; Spahni et al., 2011; Montzka et al., 2011). On the source side, a positive anomaly of CH$_4$ emissions from natural wetlands in 2007–2008 is suggested to be the cause by several authors (Duglokencky et al., 2009; Bloom et al., 2010; Bousquet et al., 2011), with the contribution from boreal regions in 2007 being estimated to amount about 25 % (Bousquet et al., 2011). Indeed, Siberia experienced high abnormally positive temperatures in autumn (+4 °C compared to the 1961–1990 mean, National Climatic Data Center, Dlugokencky et al., 2009). The remaining 75 % are mostly attributed to tropical natural wetlands. Tropical areas which include large wet-

land surface areas on a global scale experienced the 3rd-largest (2007) and the largest (2008) positive precipitation anomalies from 1986 to 2008 (Schneider et al., 2008; Dlugokencky et al., 2009).These findings are supported by the emission increase computed by two process-based models for natural wetlands (Ringeval et al., 2011; Spahni et al., 2011) because of tropical precipitation changes associated with the La Niña event in these years and because of boreal positive temperature anomalies in 2007. These findings are also consistent with processes relating to hydrology being the dominant driver of wetland CH$_4$ emissions in the tropics, whereas temperature is more important at high latitudes (Walter et al., 2001; Ringeval et al., 2010). According to economic statistics, the EDGAR inventory for anthropogenic emissions (EDGAR, 2012) reveals a large increase of fossil fuel emissions after 2000, mainly due to Asian regions, of +6–7 Tg yr^{-1}. This finding is supported neither by a recent atmospheric inversion (Bousquet et al., 2011) nor by a study based on ethane trends (Aydin et al., 2011). In addition, the biomass burning contribution is insignificant, because no large CO anomaly is observed in 2007–2008 (van der Werf et al., 2010).

On the sink side, Rigby et al. (2008) and Montzka et al. (2011) find a generally declining OH concentration between 2004 and 2007, which might have contributed to the enhanced CH_4 in the atmosphere. However, the large, although uncertain, $4 \pm 14\%$ drop between 2006 and 2007 inferred by Rigby is much larger than the small drop found by Montzka using several modeling approaches ($\sim 1\%$/yr). Small inter-annual variability of OH concentrations is in better agreement with the results of most climate chemistry models which largely buffer OH changes (Dentener et al., 2003). Having in mind that methane loss by OH radicals is proportional to the product of OH concentrations by the CH_4 mixing ratio, even a decrease of OH concentration does not guarantee a decreased loss of methane by OH radicals. CH_4 has increased by about 0.3 %/yr since 2007, which may, at least partly, compensate the impact of the potentially decreasing OH concentrations on CH_4 mixing ratios.

Attribution of the persistent increase to a particular source or sink after 2009 still is under investigation. On the one hand, a persisting natural wetland anomaly over more than a few years may be in conflict with the understanding that wetland emissions typically cause inter-annual variability (i.e., 1–2 yr time scale), except for long-term precipitation or temperature anomalies. On the other hand, more gradual changes on the time scale of several years would be explained more consistently by changing anthropogenic emissions.

Finally, no evidence of strongly increased emissions from stores of carbon in melting permafrost and from marine hydrates as a reaction to climate change can be found today from atmospheric observations. However, both will remain potential sources of severely increasing methane emissions in the future, which should be monitored closely by remote sensing at the surface or from space.

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