

## Seasonal and latitudinal variation of atmospheric methane: A ground-based and ship-borne solar IR spectroscopic study

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[1] Column-averaged volume mixing ratios of CH<sub>4</sub> were retrieved with a precision of better than 0.5% from infrared solar absorption spectra obtained at Ny-Alesund (Spitsbergen, 79°N) between 1997 and 2004 and during two ship cruises (54°N–34°S) on the Atlantic in 2003. The retrieval has been performed in a spectral region available to all operational FTIR (Fourier Transform InfraRed) spectrometers performing solar absorption measurements. The seasonality and the long-term increase of the tropospheric volume-mixing ratio, derived from the infrared measurements agree well with data from surface sampling at this site. The latitudinal variation of ship-borne measurements between 54°N and 34°S is in agreement with inverse model simulations which are optimized vs. the global NOAA/ESRL measurements. **Citation:** Warneke, T., J. F. Meirink, P. Bergamaschi, J.-U. Grooß, J. Notholt, G. C. Toon, V. Velazco, A. P. H. Goede, and O. Schrems (2006), Seasonal and latitudinal variation of atmospheric methane: A ground-based and ship-borne solar IR spectroscopic study, *Geophys. Res. Lett.*, **33**, L14812, doi:10.1029/2006GL025874.

### 1. Introduction

[2] Methane (CH<sub>4</sub>) belongs to the most important greenhouse gases in the atmosphere and is one of the target gases in the Kyoto protocol. Its atmospheric concentration has more than doubled since pre-industrial times and is currently at about 1780 ppb. Sources and sinks of atmospheric methane are not well quantified. In particular, in the tropics with no ground based methane measuring stations a large uncertainty in the methane budget exists.

[3] The first spaceborne measurements with sufficient precision revealed unexpected features of the global distribution of atmospheric methane [Frankenberg *et al.*, 2005]. Especially puzzling were the enhancement of methane of up to 70 ppb over tropical forests compared to model simulations which use current emission inventories. Recently, terrestrial plants were identified as a hitherto unknown source of methane, possibly contributing 10–30% to the

total annual methane emissions into the atmosphere [Keppler *et al.*, 2006]. These plant emissions may help to explain the methane enhancements seen from space. However, a confirmation of the satellite observations by other measurements is still missing.

[4] In situ measurements from surface monitoring sites cannot be directly used for the satellite validation because they are probing the earth surface, while the satellites average over the entire atmospheric column. In particular gases with a non-uniform height profile like CH<sub>4</sub> must be validated by ground based remote sensing measurements. A suitable method for the satellite validation is ground based solar absorption spectrometry. Washenfelder *et al.* [2003] showed that the CH<sub>4</sub> column as well as the average tropospheric CH<sub>4</sub> vmr can be retrieved from ground-based solar absorption measurements in the near-IR with a precision of better than 0.5% using O<sub>2</sub> as a reference gas. However, most FTIR-spectrometers within the NDSC network do not measure the near-IR spectral region used by Washenfelder *et al.* [2003] and therefore cannot use O<sub>2</sub>. This may significantly contribute to their relatively low precision, estimated at 3% by Dils *et al.* [2005].

[5] In this paper we present ground-based FTIR observations from the high Arctic at Ny-Alesund (79°N) and for two ship cruises on the Atlantic (54°N to 34°S). We show that a precision of better than 0.5% for the CH<sub>4</sub> column are not only achievable in the near-IR, but also in a spectral region which is available to all operational FTIR spectrometers performing solar absorption measurements. At Ny-Alesund the tropospheric vmr derived from the FTIR observations is compared with surface sampling data performed by the NOAA ESRL Global monitoring division (NOAA-ESRL). Furthermore, these data and the data of the two ship cruises are compared with inverse model simulations which are optimized vs. the global NOAA/ESRL measurements (P. Bergamaschi *et al.*, Satellite cartography of atmospheric methane from SCIAMACHY onboard ENVISAT: 2. Evaluation based on inverse model simulations, submitted to *Journal of Geophysical Research*, 2006, hereinafter referred to as Bergamaschi *et al.*, submitted manuscript, 2006).

### 2. Measurements and Data Analysis

[6] Solar absorption Fourier-transform (FT) measurements have been performed at the primary NDSC (Network for the Detection of Stratospheric Change) station at Ny-Alesund (Spitsbergen, 79°N, 12°E, 20 m asl) and aboard the research vessel Polarstern during meridional transects on the Atlantic. The first ship cruise started in Cape Town (33.9°S, 18.4°E) on January 24, 2003 and ended in Bremerhaven (53.5°N, 8.6°E) on February 17, 2003. The second cruise

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**Table 1.** Microwindows and Fitted Interfering Gases Used in the Retrieval

Target Gas	Spectral Windows and Interfering Gases	
	Ny Alesund	Ship Cruise
CH <sub>4</sub>	4209–4220 cm <sup>-1</sup> (H <sub>2</sub> O, CO, HDO)	6090–6120 cm <sup>-1</sup> (H <sub>2</sub> O, CO <sub>2</sub> )
HF	4038.78–4039.10 cm <sup>-1</sup> (H <sub>2</sub> O, CH <sub>4</sub> , HDO)	-
H <sub>2</sub> O	4110–4117 cm <sup>-1</sup> (CH <sub>4</sub> , CO, HDO)	6090–6120 cm <sup>-1</sup> (CH <sub>4</sub> , CO <sub>2</sub> )

started in Bremerhaven on October 22, 2003 and ended in Cape Town on November 15, 2003. The general experimental set-up used in these studies is described elsewhere [Notholt *et al.*, 1997, 2000]. The observations in the near-infrared (NIR) cover the spectral range between 5800 cm<sup>-1</sup> and 9000 cm<sup>-1</sup> and were carried out with a CaF<sub>2</sub> beamsplitter and an InGaAs photodiode.

[7] The spectra were analysed using the line-by-line code GFIT, developed at NASA/JPL [e.g., Toon *et al.*, 1992]. Profiles of pressure, temperature and relative humidity up to 30 km are taken from sondes that were launched daily at Ny-Alesund and on the ship. The initial vmr-profiles are based on balloon observations [Toon *et al.*, 1999] and were modified as explained by Notholt *et al.* [1997, 2000]. The spectral line parameters were taken from an updated version (July2004) of the ATMOS database [Brown *et al.*, 1996]. For the retrieval of methane two wavelength regions were employed (see Table 1 for details): Region 1 at 6105 cm<sup>-1</sup> (1.638 μm) and region 2 at 4215 cm<sup>-1</sup> (2.372 μm).

### 3. Results

#### 3.1. Tropospheric CH<sub>4</sub> and Precision of the FTIR Measurements

[8] Washenfelder *et al.* [2003] showed that the CH<sub>4</sub> column as well as the average tropospheric CH<sub>4</sub> vmr can be retrieved from ground-based solar absorption measurements in the near-IR with a precision of better than 0.5% using O<sub>2</sub> as a reference gas. However, most FTIR-spectrometers within the NDSC do not measure the near-IR spectral region used by Washenfelder *et al.* [2003] and therefore cannot use O<sub>2</sub>. It would be desirable that the existing NDSC instruments also measure CH<sub>4</sub> with such a high precision. For the analysis of the Ny-Alesund spectra the 4209–4220 cm<sup>-1</sup> region was chosen, which can be measured by the spectrometers dedicated to near-IR measurements, but also by the spectrometers operated within the NDSC. Instead of the O<sub>2</sub> reference taken by Washenfelder *et al.* [2003] we use the atmospheric surface pressure at the location of the instrument and the water vapor retrieved from the same spectra to calculate the dry air column. Dividing the CH<sub>4</sub> column by the dry air column yields the column averaged volume-mixing ratio of CH<sub>4</sub>. The calculation of the tropospheric vmr of CH<sub>4</sub> is similar to the analysis by Washenfelder *et al.* [2003]. Following Washenfelder *et al.* [2003] the varying contribution of stratospheric CH<sub>4</sub> to the total column is inferred from an assumed stratospheric “slope equilibrium” relationship between CH<sub>4</sub> and HF, which are retrieved from the same spectra. Here a CH<sub>4</sub>-HF slope of -870 is used for the calculation of the tropospheric volume-mixing ratio. This slope is the mean stratospheric CH<sub>4</sub>-HF slope at Ny-Alesund based on a stratospheric climatology for HF and CH<sub>4</sub> derived from HALOE measurements [Grooß and Russell, 2005].

[9] Scaling factors are applied on the remote sensing data to account for small systematic differences between the in situ and the remote sensing data. These differences arise mainly from errors in the spectral database used for the retrieval of the remote sensing data. The in situ measurements of CH<sub>4</sub> by NOAA-ESRL are performed by gas chromatography, a relative measurement, which uses standards for calibration. Recently the NOAA-ESRL data have been adjusted to the new NOAA04 scale [Dlugokencky *et al.*, 2005], which we use throughout this work. We determine a scaling factor of 1.0124 for the window at 4215 cm<sup>-1</sup> from a comparison of the NOAA-ESRL data with the average tropospheric CH<sub>4</sub> derived from the solar absorption measurements at Ny Alesund (Table 2). For the two cruises on the Atlantic only a few spectra were available at 4215 cm<sup>-1</sup>, but observations in the near-IR region (5800–9000 cm<sup>-1</sup>) were a major focus during these cruises. For this reason for the cruises a spectral window at 6105 cm<sup>-1</sup> was selected for the methane retrieval. To determine the scaling factor for the spectral window at 6105 cm<sup>-1</sup>, methane was also retrieved from the window at 4215 cm<sup>-1</sup> from all available cruise spectra in this spectral region. Minimising the bias between methane retrieved from 4215 cm<sup>-1</sup> and the methane retrieved from the spectral window at 6105 cm<sup>-1</sup> yields a scaling factor of 0.965 for the 6105 cm<sup>-1</sup> window (Table 2).

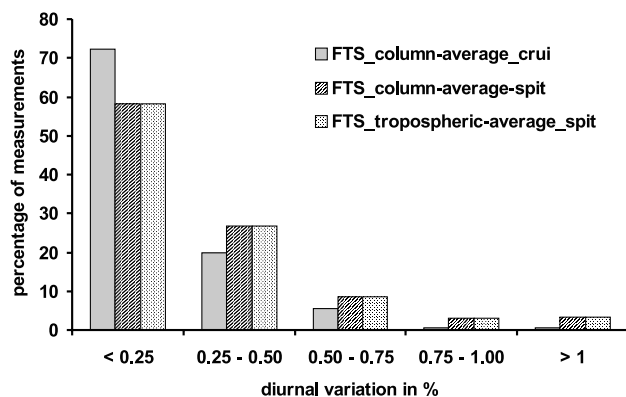
[10] The near-IR spectra recorded during the cruise also allow to retrieve O<sub>2</sub>. O<sub>2</sub> can be used as a standard because its vmr is, to the degree required, constant in the atmosphere. The O<sub>2</sub> could be used to calculate the column-averaged vmr of CH<sub>4</sub> from the CH<sub>4</sub>/O<sub>2</sub> ratio. However, in the case of the ship-borne data, the quality of the O<sub>2</sub> retrieval would limit the precision of the column averaged CH<sub>4</sub>. Therefore we used the O<sub>2</sub> only to identify spectra which are affected by clouds. Only spectra with a O<sub>2</sub> vmr within 1% of the mean retrieved vmr of O<sub>2</sub> were used for the analysis.

[11] The precision of the column-averaged and tropospheric vmrs is estimated from its diurnal variation. Part of the diurnal variation will be caused by real variations in CH<sub>4</sub> over the day, therefore this method gives an upper limit for the precision. Figure 1 shows the distribution of the achieved precisions. For the cruises 95% of the measurements show a diurnal variation of smaller than 0.5%. The

**Table 2.** Scaling Factors Between the Calibration Standards CMDL83 and NOAA04 and the FTS Measurements<sup>a</sup>

	CMDL83 ×	FTS 4200 ×	FTS 6000 ×
CMDL83 =	1	1	0.9532
NOAA04 =	1.0124	1.0124	0.965

<sup>a</sup>Standards are from Dlugokencky *et al.* [2005]. The symbol × denotes multiplication.



**Figure 1.** Diurnal variations of the column averaged and tropospheric vmr of CH<sub>4</sub>. The diurnal variation is defined as the absolute value of the deviation of one measurement from the daily mean, hence as the absolute value of  $100 \times \left( \frac{x}{\langle x \rangle} - 1 \right)$  where  $x$  is one measurement and  $\langle x \rangle$  the mean of the day.

smaller diurnal variations during the cruises compared to Spitsbergen (Figure 1) is most likely due to the sorting of the cruise spectra according to the criterion that the O<sub>2</sub> vmr is within 1% of the mean retrieved vmr of O<sub>2</sub>. At Spitsbergen about two thirds of the measurements with diurnal O<sub>2</sub> variations higher than 1% have diurnal CH<sub>4</sub> variations of more than 2%. Hence these measurements can be identified as outliers, most likely resulting from the impact of clouds. Disregarding the outliers, more than 95% of the measurements show diurnal CH<sub>4</sub> variations of less than 0.5% in agreement with the cruises. Based on these results we conclude that CH<sub>4</sub> can be retrieved with a precision of better than 0.5% in the near-IR as well as from a spectral region available to all operational FTIR spectrometers performing solar absorption measurements.

### 3.2. Seasonal and Latitudinal Variation

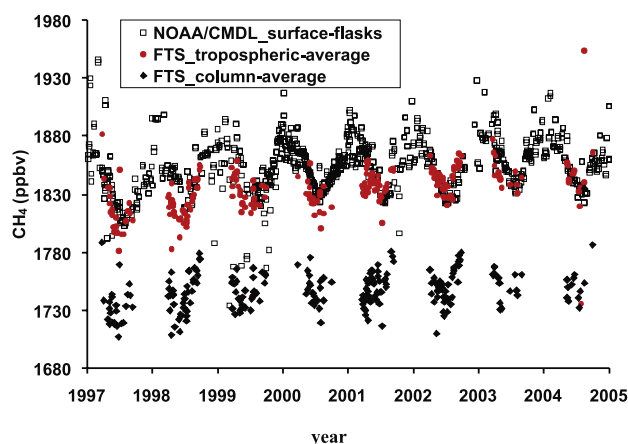
[12] At Ny-Alesund surface CH<sub>4</sub> data are available either from (i) flask measurements by the NOAA ESRL Global monitoring division (NOAA-ESRL) (Carbon Cycle Greenhouse Gases Group, NOAA Climate Monitoring and Diagnostics Laboratory, Boulder, Colorado, 2003, available at <ftp://ftp.cmdl.noaa.gov/ccg/>) and (ii) continuous in-situ measurements by the Norwegian Institute for Air Research (NILU) [Pedersen *et al.*, 2005]. Both, flask sampling and continuous in-situ measurements take place at Zeppelin Mountain (475 m asl) only about 2 km away from the NDSC station with the FTIR spectrometer. A comparison of the averaged tropospheric CH<sub>4</sub> derived from solar absorption measurements with NOAA-ESRL in situ data at Ny Alesund is shown in Figure 2. The seasonal variability as well as the long-term trend of the tropospheric CH<sub>4</sub> derived from the solar absorption measurements and the NOAA data agree well. This also indicates that the shape of the CH<sub>4</sub> profile in the troposphere did not change throughout the time of the observations. The only exception is the year 1998, where the in situ data are higher than the solar absorption data. During this year a significant enhancement

in CO resulting from El-Nino related biomass burning was detected in the Arctic [Yurganov *et al.*, 2004]. The higher CH<sub>4</sub> concentration seen in the in situ data compared to the remote sensing data could be due to pollution from biomass burning predominantly present in the lower troposphere. The column averaged vmr, retrieved from solar absorption measurements without applying the correction with HF, is lower than the tropospheric vmr due to the strong decrease of CH<sub>4</sub> in the stratosphere.

[13] Recently, global maps of atmospheric methane concentrations became available from SCIAMACHY observations [Frankenberg *et al.*, 2005, 2006; Buchwitz *et al.*, 2005]. These data show an unexpected enhancement of methane over tropical forests if compared with model data. Up to now no ground truth exists for these observations. Solar absorption FTIR-spectrometry is the only ground based remote sensing technique that allows to retrieve column averaged vmrs of CH<sub>4</sub> with precisions of better than 0.5% and is therefore well suited for the validation of the satellite measurements. Due to the low reflectivity and the resulting low signal-to-noise ratio no satellite measurements are available over water and the ship-borne solar absorption measurements cannot directly be compared with the satellite data.

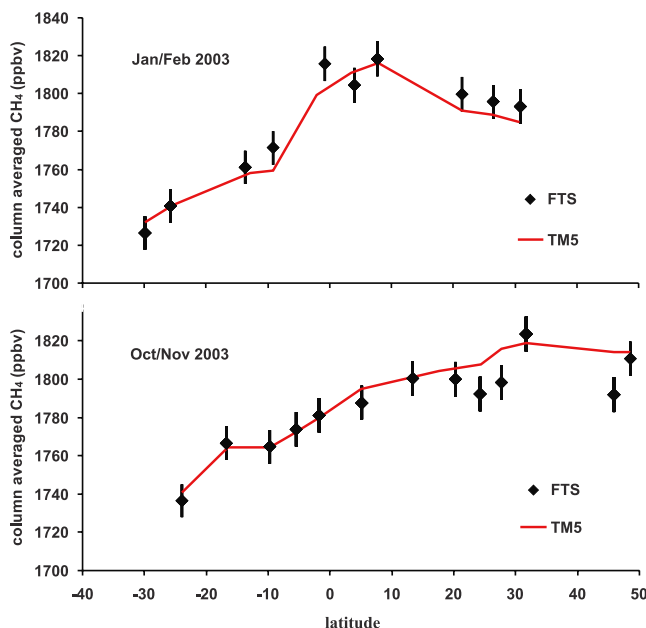
[14] Results from the two ship cruises are shown in Figure 3. We have compared our measurements with TM5 inverse model simulations based on the TM5 model [Krol *et al.*, 2005]. The inverse simulations are optimized vs. the global NOAA/ESRL measurements as described by Bergamaschi *et al.* (submitted manuscript, 2006) (here simulations of their scenario S1 are applied). It has been demonstrated that the optimized model fields have a high degree of consistency with the NOAA/ESRL background measurements and should therefore constitute a good reference for comparison in remote regions.

[15] A scaling factor of 1.0134 was derived from a least square fit between the model data and the FTS data. Since



**Figure 2.** Ground-based Fourier Transform Spectroscopic measurements of the column averaged vmr of CH<sub>4</sub> (solid black diamonds) and tropospheric vmr of CH<sub>4</sub> (red circles). NOAA-ESRL surface sampling (open black squares) is performed at Zeppelin Mountain (475masl) about 2 km away from the remote sensing observations. The FTS data are scaled by 1.0124 and the NOAA-ESRL data are expressed on the NOAA04 scale.





**Figure 3.** Daily average of column averaged vmr of CH<sub>4</sub> (black diamonds) measured by Fourier Transform Spectroscopy (FTS) during two ship cruises on the Atlantic compared with TM5 model data (red line). The error bars of the FTS data represent the precision of 0.5% derived from the diurnal variation (Figure 1). The model data is the data for the day of the measurement. The FTS data are scaled by 0.965 (see text for details). The model is scaled by 1.0134. The model scaling factor for the model was derived from a least square fit between the model data and the FTS data.

the comparison of the latitudinal variation is most important the model data were scaled with the factor 1.0134 to match the measurements (Figure 3). A possible reason for the offset between the model and the measurements could be a wrong CH<sub>4</sub>-HF slope, derived from the HALOE data. A wrong CH<sub>4</sub>-HF slope would directly impact the scaling factor applied to the FTIR-data. Instrumental and retrieval biases are not expected to be significant.

[16] After scaling, the latitudinal variation of model- and solar absorption data agrees well and no evidence of a tropical methane enhancement is seen in the data. The reason could be that the ship-borne measurements are performed too far away from the potential sources to detect their signal. The only disagreement between model and measurements are three datapoints in the Northern hemisphere during the cruise in Oct/Nov 2003, which show lower values than the model. During the cruise in Jan/Feb 2003 concurrent solar absorption measurements of CO by our spectrometer reveal a strong impact by biomass burning in the region between the equator and 10°N [Velazco et al., 2005]. In this region no discrepancy between model and measurements can be detected, which shows that the impact of biomass burning is well represented in the model.

#### 4. Conclusions

[17] Column averaged volume-mixing ratios as well as average tropospheric volume-mixing ratios of CH<sub>4</sub> can be

retrieved with a precision of better than 0.5% from the near-IR as well as from the IR, a spectral region available to all operational FTIR spectrometers of the NDSC network.

[18] Using a scaling factor of 1.0124, the tropospheric CH<sub>4</sub> derived from solar absorption measurements at Ny-Alesund shows the same seasonal behavior and long-term increase of methane between 1997 and 2004 as the surface flask data.

[19] The latitudinal variation of the column-averaged vmr of CH<sub>4</sub> derived from ship-based solar absorption measurements agrees well with model simulations for the two cruises in 2003 and is therefore consistent with surface observations.

[20] The good agreement of FTIR measurements with in situ measurements and with inverse model simulations demonstrate the high precision of the FTIR measurements and their suitability for validation of satellite data.

[21] **Acknowledgments.** We are grateful to the NOAA ESRL Global monitoring division for providing free access to their surface data (<http://www.cmdl.noaa.gov/ccgg/iadv/index.php>). We would like to acknowledge financial support by the German Ministry for Research and Education (BMBF) that funded this work via the DLR-Bonn (grant 50EE0013), the European Commission that funded the UFTIR project (EVK2-CT-2002-00159) and the Helmholtz Association within the framework of the virtual institute PEP.

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