Evaluation of column-averaged methane in models and TCCON with a focus on the stratosphere

Andreas Ostler1, Ralf Sussmann1, Prabir K. Patra2, Sander Houweling3,4, Marko De Bruine3, Gabriele P. Stiller5, Florian J. Haenel1, Johannes Plieninger5, Philippe Bouqueta6,7, Yi Yin6,7, Marielle Saunois6,7, Kaley A. Walker8, Nicholas M. Deutscher9,10, David W. T. Griffith9, Thomas Blumenstock6, Frank Hase6, Thorsten Warneke10, Zhiting Wang10, Rigel Kivi11, and John Robinson12

1Karlsruhe Institute of Technology, IMK-IFU, 82467 Garmisch-Partenkirchen, Germany
2Research Institute for Global Change, JAMSTEC, Yokohama, 236-0001, Japan
3Institute for Marine and Atmospheric Research Utrecht, Utrecht University, 3584 CC Utrecht, the Netherlands
4SRON Netherlands Institute for Space Research, 3584 CA Utrecht, the Netherlands
5Karlsruhe Institute of Technology, IMK-ASF, 76021 Karlsruhe, Germany
6Laboratoire des Sciences du Climat et de l’Environnement, IPSL-LSCE, CEA-CNRS-UVSQ, UMR8212, 91191 Gif-sur-Yvette, France
7Université de Versailles Saint Quentin en Yvelines, 78000 Versailles, France
8Department of Physics, University of Toronto, Toronto, Ontario M5S 1A7, Canada
9School of Chemistry, University of Wollongong, Wollongong, NSW 2522, Australia
10Institute of Environmental Physics, University of Bremen, 28334 Bremen, Germany
11Finnish Meteorological Institute, Arctic Research Center, 96600 Sodankylä, Finland
12Department of Atmospheric Research, National Institute of Water and Atmospheric Research (NIWA) Ltd, Wellington 6021, New Zealand

Correspondence to: Ralf Sussmann (ralf.sussmann@kit.edu)

Received: 14 March 2016 – Published in Atmos. Meas. Tech. Discuss.: 11 May 2016
Revised: 12 September 2016 – Accepted: 15 September 2016 – Published: 28 September 2016

Abstract. The distribution of methane (CH4) in the stratosphere can be a major driver of spatial variability in the dry-air column-averaged CH4 mixing ratio (XCH4), which is being measured increasingly for the assessment of CH4 surface emissions. Chemistry-transport models (CTMs) therefore need to simulate the tropospheric and stratospheric fractional columns of XCH4 accurately for estimating surface emissions from CH4. Simulations from three CTMs are tested against XCH4 observations from the Total Carbon Column Network (TCCON). We analyze how the model–TCCON agreement in XCH4 depends on the model representation of stratospheric CH4 distributions. Model equivalents of TCCON XCH4 are computed with stratospheric CH4 fields from both the model simulations and from satellite-based CH4 distributions from MIPAS (Michelson Interferometer for Passive Atmospheric Sounding) and MIPAS CH4 fields adjusted to ACE-FTS (Atmospheric Chemistry Experiment Fourier Transform Spectrometer) observations. Using MIPAS-based stratospheric CH4 fields in place of model simulations improves the model–TCCON XCH4 agreement for all models. For the Atmospheric Chemistry Transport Model (ACTM) the average XCH4 bias is significantly reduced from 38.1 to 13.7 ppb, whereas small improvements are found for the models TM5 (Transport Model, version 5; from 8.7 to 4.3 ppb) and LMDz (Laboratoire de Météorologie Dynamique model with zooming capability; from 6.8 to 4.3 ppb). Replacing model simulations with MIPAS stratospheric CH4 fields adjusted to ACE-FTS reduces the average XCH4 bias for ACTM (3.3 ppb), but increases the average XCH4 bias for TM5 (10.8 ppb) and LMDz (20.0 ppb). These findings imply that model errors in simulating stratospheric CH4 contribute to model biases. Current satellite instruments cannot definitively measure stratospheric CH4 to sufficient accuracy to eliminate these biases. Applying transport diag-

Published by Copernicus Publications on behalf of the European Geosciences Union.
nometrics to the models indicates that model-to-model differences in the simulation of stratospheric transport, notably the age of stratospheric air, can largely explain the inter-model spread in stratospheric CH₄ and, hence, its contribution to XCH₄. Therefore, it would be worthwhile to analyze how individual model components (e.g., physical parameterization, meteorological data sets, model horizontal/vertical resolution) impact the simulation of stratospheric CH₄ and XCH₄.

1 Introduction

The column-averaged dry-air mixing ratio of methane (CH₄), denoted as XCH₄, is an integrated measure of CH₄ with contributions from the troposphere and the stratosphere. Observations of XCH₄ contain source/sink information on a global to regional scale. They are provided by the ground-based networks NDACC (Network for the Detection of Atmospheric Composition Change, http://www.ndacc.org/; Sussmann et al., 2011, 2012, 2013) and TCCON (Total Carbon Column Observing Network, http://www.tccon.caltech.edu/; Wunch et al., 2011), and also by satellite-based observation platforms like SCIAMACHY (Scanning Imaging Absorption Spectrometer for Atmospheric Cartography; Burrows et al., 1995; Frankenberg et al., 2011) and GOSAT (Greenhouse Gases Observing Satellite; Kuze et al., 2009; Yokota et al., 2009). Satellite-inferred XCH₄ observations are increasingly used in atmospheric inverse modeling because of their beneficial spatial-temporal data coverage (Bergamaschi et al., 2013; Fraser et al., 2013, 2014; Monteil et al., 2013; Houweling et al., 2014; Wecht et al., 2014; Cressot et al., 2014; Alexe et al., 2015; Turner et al., 2015; Locatelli et al., 2015). Given the high accuracy of ground-based XCH₄ TCCON retrievals, these observations are typically used for the evaluation of both chemistry-transport model (CTM) simulations (Saito et al., 2012; Belikov et al., 2013; Monteil et al., 2013; Fraser et al., 2014; Alexe et al., 2015; Turner et al., 2015) and satellite-retrieved XCH₄ (Parker et al., 2011, 2015; Schepers et al., 2012; Dils et al., 2014; Houweling et al., 2014; Parker et al., 2015; Kulawik et al., 2016; Pandey et al., 2016; Inoue et al., 2016).

Because of the various influences on XCH₄, however, the interpretation of residual XCH₄ differences with TCCON may be difficult. For example, a good agreement between XCH₄ simulations and observations may suggest that a CTM is able to represent atmospheric conditions in a realistic way. However, it could also be the case that systematic model and satellite data errors in the troposphere and the stratosphere compensate each other. For this reason, it is necessary to extend model validations with additional atmospheric CH₄ observations that are complementary to XCH₄ observations, like surface or airborne in situ measurements, or balloon-based vertical profiles (Karion et al., 2010). In the context of a refined model comparison, it is also possible to separate ground-based XCH₄ observations into tropospheric and stratospheric partial columns (Washenfelder et al., 2003; Sepúlveda et al., 2012, 2014; Wang et al., 2014; Saad et al., 2014).

Model–measurement XCH₄ residuals are minimized by atmospheric inversions in order to constrain CH₄ emission fluxes. Inversion models are also able to make use of in situ measurements and XCH₄ observations at the same time in order to adjust prior emission fluxes. Nevertheless, such inverse models still have to deal with ill-defined XCH₄ biases, which, in contrast to well-quantified biases, can only be attributed to errors in the model or the observations with an ambiguous assignment (Houweling et al., 2014). Currently, there are various approaches to optimize bias correction functions within the inverse model or to construct bias corrections as ad hoc functions of latitude or air mass. Ad hoc bias corrections, like removing a latitudinal background pattern in XCH₄ model–observation differences, are common, even though they bear the risk of obscuring real signals from emissions on the Earth’s surface. Given the fact that the stratospheric contribution relative to the CH₄ total column increases from ∼5% at the tropics up to ∼25% at midlatitudes and high latitudes, model errors in the representation of stratospheric CH₄ mixing ratios are expected to give rise to a latitudinal varying bias (Turner et al., 2015). Although it is known that CTMs differ by up to ∼50% in the simulation of lower stratospheric CH₄ distributions (Patra et al., 2011), an atmospheric region with a steep methane gradient of ∼−50 ppb km⁻¹, the impact of model errors in stratospheric CH₄ on XCH₄ has not been rigorously quantified up to now. In this context, the goal of this study is to better understand the sensitivity of XCH₄ model–observation differences to the model representation of stratospheric CH₄.

Our XCH₄ model–observation analysis is based on optimized model simulations from three well-established CTMs on the one side and accurate XCH₄ observations from TCCON on the other. The impact of model stratospheric CH₄ distributions on XCH₄ is estimated by replacing modeled stratospheric CH₄ fields with monthly mean CH₄ distributions observed by MIPAS (Michelson Interferometer for Passive Atmospheric Sounding) and by ACE-FTS (Atmospheric Chemistry Experiment Fourier Transform Spectrometer). In addition to this, we briefly evaluate the model characteristics of stratospheric transport in order to understand differences between simulated and observed CH₄ distributions. The paper has the following structure: After introducing the models (Sect. 2) and the observations (Sect. 3), we present both a direct model–TCCON comparison and a comparison with refined model data using satellite data products of stratospheric CH₄ in Sect. 4. The transport characteristics of the models are discussed in Sect. 5, followed by a summary and conclusions in Sect. 6.
The focus of this study is the assessment of the impact of stratospheric CH$_4$ on XCH$_4$. Therefore, we try to ensure that model simulations represent tropospheric CH$_4$ mixing ratios as well as possible. For this purpose, we use optimized CH$_4$ model simulations that have been constrained by surface observations. Our model analysis comprises simulations from three well-established CTMs that have already been part of the chemistry-transport model intercomparison experiment TransCom-CH$_4$ (Patra et al., 2011) and used in inverse modeling of CH$_4$ emissions. Furthermore, we use model simulations of stratospheric mean age for an evaluation of model transport characteristics in Sect. 5. Basic model features are given in Table 1.

### 2 Model simulations

#### 2.1 ACTM

The Atmospheric Chemistry Transport Model (ACTM) model (Patra et al., 2009a) is an atmospheric general circulation model (AGCM)-based CTM from the Center for Climate System Research/National Institute for Environmental Studies/Frontier Research Center for Global Change (CCSR/NIES/FRCCG). Here, we use optimized ACTM simulations presented in Patra et al. (2016) as inversion case 2 (CH$_4$ags). The ACTM horizontal resolution is $\sim 2.8 \times 2.8^\circ$ (T42 spectral truncations) with 67 sigma-pressure vertical levels. The meteorological fields of ACTM are nudged with reanalysis data from the Japan Meteorological Agency, version JRA-25 (Onogi et al., 2007). ACTM uses an optimized OH field (Patra et al., 2014) based on a scaled version of the seasonally varying OH field from Spivakovsky et al. (2000). The concentration fields that are relevant for stratospheric CH$_4$ loss – OH, O($^1$D), and chlorine (Cl) radicals – are based on simulations by the ACTM’s stratospheric model run (Takigawa et al., 1999). ACTM mean age is derived from the simulation of an idealized transport tracer with uniform surface fluxes, linearly increasing trend, and no loss in the atmosphere (Patra et al., 2009b). The ACTM simulates the observed CH$_4$ interhemispheric gradient in the troposphere and individual in situ measurements generally within 10 ppb (Patra et al., 2016).

#### 2.2 TM5

The global chemistry Tracer Model, version 5 (TM5) has been described in Krol et al. (2005) and used as an atmospheric inversion model for CH$_4$ emissions (Bergamaschi et al., 2005; Meirink et al., 2008; Houweling et al., 2014). Here, we use TM5 simulations of CH$_4$ optimized with surface measurements only (Pandey et al., 2016). TM5 is run with a horizontal resolution of $6^\circ \times 4^\circ$ and a vertical grid of 25 layers. TM5 meteorology is driven by the reanalysis data set ERA-Interim (Dee et al., 2011) from the European Centre for Medium Range Weather Forecasts (ECMWF). The simulation of the chemical CH$_4$ sink uses OH fields from Spivakovsky et al. (2000), which have been scaled to match methyl chloroform measurements. In addition to that, stratospheric CH$_4$ loss via Cl and O($^1$D) radicals is simulated using their concentration fields based on the 2-D photochemical Max Planck Institute (MPI) model (Brühl and Crutzen, 1993). Known deficiencies in the TM5 simulation of interhemispheric mixing have been corrected by extending the model with a horizontal diffusion parameterization that is adjusted to match SF$_6$ simulations with SF$_6$ measurements (Monteil et al., 2013).

TM5 simulations of sulfur hexafluoride (SF$_6$) were used to derive stratospheric mean age data. SF$_6$ mixing ratios are monotonically increasing with time, showing higher mixing ratios in the troposphere than in the stratosphere, given the transport time from SF$_6$ surface sources to higher altitudes. This implies that tropospheric and stratospheric SF$_6$ mixing ratios of equal size are separated from each other by a time lag, which is commonly defined as mean age of air. In order to derive mean age from SF$_6$ model simulations, the same tropospheric SF$_6$ reference time series was used as for the derivation of MIPAS mean age data (see Stiller et al., 2012).

#### 2.3 LMDz

The LMDz (Laboratoire de Météorologie Dynamique model with zooming capability) is a general circulation model (Hourdin et al., 2006), which has been used to investigate the impact of transport model errors on inverted CH$_4$ emissions (Locatelli et al., 2013). Here, we use optimized LMDz simulations of CH$_4$, recently presented as LMDz-SP constrained

---

**Table 1. Overview of CTMs used for model–TCCON comparison.**

<table>
<thead>
<tr>
<th>Model name</th>
<th>Institution</th>
<th>Horizontal$^a$</th>
<th>Vertical$^b$</th>
<th>Output CH$_4$</th>
<th>Mean age derived from</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>ACTM</td>
<td>JAMSTEC</td>
<td>$\sim 2.8 \times 2.8^\circ$</td>
<td>67$^\sigma$</td>
<td>1-hourly, monthly</td>
<td>idealized transport tracer simulations</td>
<td>Patra et al. (2016)</td>
</tr>
<tr>
<td>TM5</td>
<td>SRON</td>
<td>$\sim 6 \times 4^\circ$</td>
<td>25$^\eta$</td>
<td>daily</td>
<td>SF$_6$ simulations</td>
<td>Pandey et al. (2016)</td>
</tr>
<tr>
<td>LMDz</td>
<td>LSCE</td>
<td>$\sim 3.75 \times 1.875^\circ$</td>
<td>39$^\eta$</td>
<td>monthly</td>
<td>SF$_6$ simulations</td>
<td>Locatelli et al. (2015)</td>
</tr>
</tbody>
</table>

$^a$ Longitude $\times$ latitude. $^b$ Vertical coordinates in sigma-pressure $\sigma$ (pressure divided by surface pressure) and hybrid sigma-pressure $\eta$. 

www.atmos-meas-tech.net/9/4843/2016/
by surface measurements from background sites (Locatelli et al., 2015). These model simulations are nudged with the ERA-Interim reanalysis data set for horizontal winds \((u, v)\). LMDz has a horizontal resolution of \(3.75^\circ \times 1.875^\circ\), and 39 hybrid sigma-pressure layers. The chemical destruction of \(\text{CH}_4\) by \(\text{OH}\) and \(\text{O}^{(1D)}\) is based on prescribed concentration fields simulated by the chemistry–climate model LMDz-INCA (Szopa et al., 2013). No Cl-based \(\text{CH}_4\) destruction is prescribed in this version of the model. Besides \(\text{CH}_4\), LMDz simulations of \(\text{SF}_6\) were used to derive mean age data similarly to the method used for TMS.

3 Intercomparison strategy and observations

3.1 Intercomparison strategy

We want to quantify the dependence of the \(X\text{CH}_3\) on observations, intercomparison strategy

3.2 TCCON observations of column-averaged methane

Solar absorption measurements in the near-infrared are performed via ground-based Fourier transform spectrometers (FTSs) at TCCON sites across the globe. TCCON-type measurements are analyzed with the GGG software package, including the spectral fitting code GFIT to derive total column abundances of several trace gases (Wunch et al., 2011). The \(\text{CH}_4\) total column is inverted from the spectra in three different spectral windows centered at \(5938, 6002,\) and \(6076\ \text{cm}^{-1}\). The spectral fitting method is based on iteratively scaling a priori profiles to provide the best fit to the measured spectrum. The general shape of the a priori profiles has been inferred from aircraft, balloon and satellite profiles (ACE-FTS profiles measured in the 30–40\(^\circ\) N latitude range from 2003 to 2007). In addition, the shape of the daily a priori profile is vertically squeezed/stretched depending on tropopause altitude and the latitude of the measurement site. This means that the tropopause altitude is used as a proxy for stratospheric ascent/descent to represent the origin of the air mass in the a priori profile. \(X\text{CH}_4\) is calculated by dividing the \(\text{CH}_4\) number density by the simultaneously measured \(\text{O}_2\) number density (a proxy for the dry-air pressure column).

These \(X\text{CH}_4\) retrievals are corrected a posteriori for known air-mass-dependent biases and calibrated to account for air-mass-independent biases, which can, among other errors, arise from spectroscopic uncertainties (Wunch et al., 2011). The air-mass-independent calibration factor, which is determined by comparisons with coincident airborne or balloon-borne in situ measurements over TCCON sites (Wunch et al., 2010; Messerschmidt et al., 2011; Geibel et al., 2012), allows for a calibration of TCCON \(X\text{CH}_4\) retrievals to in situ measurements on the WMO scale. Furthermore, the quality of the retrievals is continuously improved by correcting the influence of systematic instrumental changes over time. As a result of these improvements there are different versions of the GGG software package. In this study we use TCCON retrievals performed with version GGG2014 (for details see https://tccon-wiki.caltech.edu/). The TCCON measurement precision \((2\sigma)\) for \(X\text{CH}_4\) is \(< 0.3\% \ (< 5\ \text{ppb})\) for single measurements. For the year 2010, \(X\text{CH}_4\) observations are available from 11 TCCON sites, listed in Table 2. Knowing that TCCON \(X\text{CH}_4\) accuracy can be affected by a strong polar vortex (Ostler et al., 2014), we exclude high-latitude observations at Sodankylä within the early spring period (March, April, May) from the analysis. TCCON data were obtained from the TCCON Data Archive, hosted by the Carbon Dioxide Information Analysis Center (CDIAC: http://cdiac.ornl.gov/). The individual data sets of the TCCON sites used in this study are available from this database.

3.3 Satellite-based data sets of stratospheric methane

In order to correct modeled stratospheric \(\text{CH}_4\) fields, we use satellite-borne MIPAS measurements covering the stratosphere. As a Fourier-Transform Infrared Spectrometer aboard the Environmental Satellite (Envisat), MIPAS detected atmospheric emission spectra in the mid-infrared region via limb sounding (Fischer et al., 2008). Profiles of various atmospheric trace gas concentrations are derived by the research processor developed by the Karlsruhe Institute of
Technology, Institute of Meteorology and Climate Research (KIT IMK) and the Instituto de Astrofísica de Andalucía (CSIC) (von Clarmann et al., 2003). The MIPAS CH$_4$ data set comprises zonal monthly means with a horizontal grid resolution of 5° latitude. In the vertical, the resolution of the MIPAS CH$_4$ fields range from 2.5 to 7 km; see Plieninger et al. (2015) for more details. As an additional quality criterion, we only select MIPAS data points that are averaged over more than 300 profile measurements. As a result, our MIPAS CH$_4$ data set typically covers altitudes higher than ~10 km at midlatitudes and heights above ~15 km in the tropics. This implies that we do not use a thermal or chemical tropopause definition, but use the MIPAS data where they are available. Therefore, we cannot exclude that our MIPAS-based CH$_4$ fields contain some upper tropospheric MIPAS values; i.e., our definition of stratospheric CH$_4$ is not strict from a meteorological point of view.

The corrected model CH$_4$ profiles rely on original model CH$_4$ fields that are merged with MIPAS-based zonal CH$_4$ fields (monthly means) interpolated to the model grid. Merging original model CH$_4$ fields/profiles with zonal monthly means implies that we lose some spatial and temporal variability in the corrected model CH$_4$ fields. For example, vertical shifts of the tropopause can cause significant variations in XCH$_4$ of ~25 ppb even within a day (Ostler et al., 2014). As these XCH$_4$ changes can be positive but also negative (tropopause shifted upwards and downwards), we expect that dynamically induced XCH$_4$ variations should be negligible from a statistical point of view as used in this study. For our aim – investigating the overall impact of model stratospheric CH$_4$ fields on the quantity XCH$_4$ – a monthly mean representation of stratospheric CH$_4$ in the corrected model fields is sufficient.

In our study we use the strongly revised MIPAS CH$_4$ data product for the MIPAS reduced-resolution period from January 2005 to April 2012. This new data set (version V5R_CH4_224/V5R_CH4_225) was recently introduced by Plieninger et al. (2015) with an emphasis on retrieval characteristics. Plieninger et al. (2015) showed that CH$_4$ mixing ratios are reduced in the lowermost stratosphere when using the new retrieval settings. This finding implies that the high bias of the older CH$_4$ data version in the lowermost stratosphere, which was determined by Laeng et al. (2015), has been partially alleviated. Nevertheless, a recent comparison study by Plieninger et al. (2016) suggests a remaining positive bias (100–200 ppb) relative to other satellite measurements such as ACE-FTS observations.

For this reason, a second satellite CH$_4$ data set was constructed by adjusting MIPAS stratospheric CH$_4$ mixing ratios to ACE-FTS (Boone et al., 2013) measurements of CH$_4$. Given the sparse data coverage of ACE-FTS observations for the year 2010, we did not use ACE-FTS measurements directly. Instead, the MIPAS CH$_4$ fields were adjusted by offsets relative to ACE shown in Fig. 1, yielding the second satellite-based CH$_4$ data set abbreviated by MIPAS_ACE. We used collocated pairs of CH$_4$ profiles from MIPAS and ACE-FTS to derive a CH$_4$ offset as a function of altitude and latitude for the year 2010. The collocation criteria are based on a maximum radius of 500 km and a maximum temporal deviation of 5 h, which is identical to Plieninger et al. (2016). Furthermore, the MIPAS averaging kernels were applied to ACE-FTS CH$_4$ profiles. ACE-FTS operates in solar occultation mode (Bernath et al., 2005) and also provides retrievals of several trace gases including CH$_4$. Here, we use ACE-FTS data from a research version of the 3.5 retrieval described in Buzan et al. (2016).

Figure 1 shows the CH$_4$ offset functions computed as mean differences between MIPAS and ACE-FTS for 30° latitudinal bands. Figure 1 confirms the findings by Plieninger et al. (2016) that MIPAS is biased positive by ~150 ppb relative to ACE-FTS within the lowermost stratosphere. For higher altitudes (>25 km), mean differences between MIPAS and ACE-FTS are larger for the tropical domain (up to 100 ppb) compared to higher latitudes (up to 50 ppb).
3.4 MIPAS-observed mean age

Besides MIPAS CH$_4$ observations, we also use MIPAS data sets of stratospheric mean age inferred from SF$_6$ measurements. Here, we use the new MIPAS mean age data set presented by Haenel et al. (2015). This new mean age data set contains several improvements compared to the previous version introduced by Stiller et al. (2012). For MIPAS, the mean age is calculated as the average transport time from the tropical troposphere to a certain location in the stratosphere using NOAA (National Oceanic and Atmospheric Administration) observations as reference. The mean age of stratospheric air is of special interest for climate research because the distributions of greenhouse gases like ozone critically depend on possible changes in the stratospheric transport pathways (Engel et al., 2009). Mean age can be inferred from observations of clock tracers (concentrations monotonically increasing with time) like SF$_6$ or CO$_2$, and can also be simulated by models. For this reason, it is a well-known diagnostic for stratospheric transport and is very suitable for the evaluation of model transport characteristics (Waugh and Hall, 2002). The combined MIPAS data set of stratospheric CH$_4$ and mean age is used for the evaluation of model transport characteristics in Sect. 5.1.

Figure 1. Mean CH$_4$ differences between collocated MIPAS and ACE-FTS CH$_4$ profiles measured in the year 2010. Mean CH$_4$ differences in parts per billion (ppb) are derived for 30° latitudinal bands indicated by different colors.

Figure 2. Site-specific model XCH$_4$ biases with respect to TCCON observations in parts per billion (ppb) for the year 2010. Different colors indicate different stratospheric CH$_4$ fields used for the calculation of model XCH$_4$.

4 Model–TCCON comparison of column-averaged methane

Figure 2 shows model biases in XCH$_4$ with respect to TCCON observations, where each TCCON site is represented by its geographical latitude. For each CTM a triplet of model CH$_4$ fields (uncorrected, MIPAS and MIPAS_ACE corrected) yields a triplet of model XCH$_4$ biases. All site-specific XCH$_4$ model biases are individually listed in Table 3. In addition, Table 4 provides an average XCH$_4$ bias for each model data set, computed as the mean of absolute site-specific biases.

The original XCH$_4$ bias for ACTM lies between 18.8 and 51.3 ppb (see Fig. 2a and Table 3). This high bias is significantly reduced when ACTM stratospheric CH$_4$ fields are replaced by satellite-based CH$_4$ fields. The model correction with MIPAS CH$_4$ reduces the average ACTM XCH$_4$ bias from 38.1 to 13.7 ppb (see Table 4). Site-specific XCH$_4$ biases are ranging from 4.8 to 19.9 ppb (see Table 3). The
Table 3. Site-specific model XCH₄ biases with respect to TCCON observations in 2010. The model–TCCON agreement in XCH₄ is evaluated with different stratospheric CH₄ model fields: the original model distribution (orig), the MIPAS-based stratospheric CH₄ (MIPAS), and the MIPAS-based stratospheric CH₄ adjusted to ACE-FTS observations (MIPAS_ACE). XCH₄ biases and corresponding 2σ standard errors (in brackets) are in parts per billion (ppb).

<table>
<thead>
<tr>
<th>Site</th>
<th>ACTM Orig</th>
<th>MIPAS</th>
<th>MIPAS_ACE</th>
<th>TM5 Orig</th>
<th>MIPAS</th>
<th>MIPAS_ACE</th>
<th>LMDz Orig</th>
<th>MIPAS</th>
<th>MIPAS_ACE</th>
</tr>
</thead>
<tbody>
<tr>
<td>SOD</td>
<td>51.3 ±2.7</td>
<td>19.9 ±2.9</td>
<td>1.8 ±2.8</td>
<td>-3.7 ±1.7</td>
<td>8.1 ±2.6</td>
<td>-8.0 ±2.5</td>
<td>13.0 ±3.0</td>
<td>9.1 ±3.2</td>
<td>-15.0 ±6.6</td>
</tr>
<tr>
<td>BIA</td>
<td>43.9 ±1.7</td>
<td>12.8 ±1.7</td>
<td>-5.0 ±1.9</td>
<td>-10.5 ±1.3</td>
<td>1.4 ±1.6</td>
<td>-14.1 ±1.6</td>
<td>4.0 ±1.7</td>
<td>1.2 ±1.8</td>
<td>-20.9 ±2.1</td>
</tr>
<tr>
<td>KAR</td>
<td>47.0 ±2.0</td>
<td>19.7 ±1.8</td>
<td>3.5 ±1.9</td>
<td>-4.0 ±1.4</td>
<td>5.7 ±1.5</td>
<td>-7.7 ±1.6</td>
<td>9.8 ±2.0</td>
<td>8.8 ±2.1</td>
<td>-13.8 ±2.2</td>
</tr>
<tr>
<td>ORL</td>
<td>47.2 ±1.7</td>
<td>19.8 ±2.2</td>
<td>3.5 ±2.3</td>
<td>-7.0 ±1.5</td>
<td>4.8 ±1.6</td>
<td>-9.2 ±1.7</td>
<td>5.4 ±2.1</td>
<td>5.3 ±2.0</td>
<td>-15.7 ±2.1</td>
</tr>
<tr>
<td>GAR</td>
<td>45.6 ±1.8</td>
<td>15.4 ±1.8</td>
<td>-0.9 ±2.0</td>
<td>-6.1 ±1.3</td>
<td>4.7 ±1.5</td>
<td>-8.1 ±1.5</td>
<td>6.1 ±1.8</td>
<td>7.3 ±1.8</td>
<td>-15.7 ±1.8</td>
</tr>
<tr>
<td>PAR</td>
<td>39.2 ±1.5</td>
<td>13.5 ±1.6</td>
<td>-1.3 ±1.6</td>
<td>-9.7 ±1.2</td>
<td>1.2 ±1.2</td>
<td>-11.0 ±1.2</td>
<td>4.4 ±1.4</td>
<td>5.9 ±1.6</td>
<td>-16.0 ±1.6</td>
</tr>
<tr>
<td>LAM</td>
<td>31.1 ±1.3</td>
<td>11.8 ±1.2</td>
<td>1.8 ±1.1</td>
<td>-4.4 ±0.8</td>
<td>2.6 ±0.9</td>
<td>-3.7 ±0.8</td>
<td>-2.0 ±1.1</td>
<td>1.7 ±1.1</td>
<td>-20.4 ±1.2</td>
</tr>
<tr>
<td>IZA</td>
<td>34.6 ±2.0</td>
<td>12.6 ±2.2</td>
<td>-1.6 ±2.2</td>
<td>-11.4 ±1.5</td>
<td>5.0 ±1.5</td>
<td>-12.6 ±1.5</td>
<td>-4.8 ±1.9</td>
<td>1.9 ±2.2</td>
<td>-31.1 ±2.2</td>
</tr>
<tr>
<td>DAR</td>
<td>18.8 ±1.6</td>
<td>8.9 ±1.7</td>
<td>0.1 ±1.8</td>
<td>-8.1 ±1.0</td>
<td>-3.1 ±1.1</td>
<td>-8.8 ±1.1</td>
<td>-9.2 ±1.6</td>
<td>-2.9 ±2.6</td>
<td>-15.0 ±1.4</td>
</tr>
<tr>
<td>WOL</td>
<td>25.8 ±1.5</td>
<td>4.8 ±1.6</td>
<td>-6.6 ±1.6</td>
<td>-17.6 ±1.4</td>
<td>-11.1 ±1.4</td>
<td>-17.9 ±1.3</td>
<td>-11.9 ±1.8</td>
<td>0.4 ±1.7</td>
<td>-29.6 ±1.9</td>
</tr>
<tr>
<td>LAU</td>
<td>34.8 ±1.0</td>
<td>11.4 ±1.2</td>
<td>-9.9 ±1.3</td>
<td>-12.7 ±1.2</td>
<td>0.0 ±1.3</td>
<td>-18.3 ±1.3</td>
<td>-4.0 ±1.4</td>
<td>3.2 ±1.4</td>
<td>-26.6 ±1.6</td>
</tr>
</tbody>
</table>

| Range | 32.5 | 15.1 | 13.4 | 13.9 | 19.2 | 14.6 | 24.9 | 12.0 | 17.3 |

Table 4. Average model XCH₄ bias with respect to TCCON observations in 2010 computed as mean of absolute site-specific biases (see Table 3). Average XCH₄ biases in ppb are derived for different model stratospheric CH₄ fields.

<table>
<thead>
<tr>
<th>Model stratospheric CH₄ field</th>
<th>ACTM</th>
<th>TM5</th>
<th>LMDz</th>
</tr>
</thead>
<tbody>
<tr>
<td>Original model</td>
<td>38.1</td>
<td>8.7</td>
<td>6.8</td>
</tr>
<tr>
<td>MIPAS</td>
<td>13.7</td>
<td>4.3</td>
<td>4.3</td>
</tr>
<tr>
<td>MIPAS_ACE</td>
<td>3.3</td>
<td>10.8</td>
<td>20.0</td>
</tr>
</tbody>
</table>

The model correction with MIPAS_ACE reduces the average ACTM XCH₄ bias further from 38.1 to 3.3 ppb (see Table 4), with values in an interval between −9.9 and 3.5 ppb (see Table 3); values similar to that were expected from the comparison with ACTM simulations with tropospheric measurements (Patra et al., 2016).

For the original TM5 we detect negative site-specific XCH₄ biases with values between −17.6 and −3.7 ppb (see Fig. 2b and Table 3). When TM5 CH₄ fields are corrected with MIPAS observations, this negative XCH₄ bias is reduced from −8.7 to −4.3 ppb on average (see Table 3). The corresponding site-specific XCH₄ biases are then between −11.1 and 8.1 ppb (Table 3). If the MIPAS_ACE is applied to TM5 then the site-specific TM5 XCH₄ biases are shifted further to the negative direction with values between −18.3 and −3.7 ppb. In this case the average XCH₄ bias increased from 8.7 to 10.8 ppb (Table 4).

With respect to TCCON observations LMDz produces both negative and positive XCH₄ biases ranging from −11.9 ppb (Wollongong) to 13.0 ppb (Sodankylä); see Fig. 2c and Table 3. The average LMDz XCH₄ bias is slightly reduced from 6.8 to 4.3 ppb if LMDz is corrected with MIPAS_ACE. After this correction, site-specific LMDz XCH₄ biases lie between −2.9 and 9.1 ppb. Using MIPAS_ACE CH₄ fields for the LMDz model correction produces LMDz XCH₄ biases between −13.8 and −31.1 ppb. At the same time, the average LMDz XCH₄ bias is increased from 6.8 to 20.0 ppb (Table 4).

Figure 3. Model–MIPAS differences of stratospheric CH₄ volume mixing ratios (vmr) in parts per billion (ppb). Zonally averaged CH₄ vmr differences are annual means for the year 2010.

www.atmos-meas-tech.net/9/4843/2016/
Overall, our results confirm that the model–TCCON agreement in XCH$_4$ depends very much on the model representation of stratospheric CH$_4$. It is obvious that the XCH$_4$ offset between ACTM and TCCON is significantly reduced with stratospheric CH$_4$ fields based on satellite data. In contrast, for TM5 and LMDz, the impact of the model correction on the model–TCCON agreement is ambiguous, in that the model–TCCON agreement can be improved (with MIPAS), but can also be reduced (with MIPAS_ACE). In order to understand this inter-model spread we look at the differences between modeled and satellite-retrieved CH$_4$ fields. Figure 3 shows zonal and annual averaged CH$_4$ mixing ratio differences between MIPAS and each CTM. Figure 3a illustrates that stratospheric CH$_4$ mixing ratios are generally much higher in ACTM than in MIPAS. The ACTM–MIPAS differences in CH$_4$ are increasing from negligible values within the lowermost stratosphere up to 450 ppb in the upper stratosphere. Furthermore, the ACTM–MIPAS difference in CH$_4$ also shows a latitudinal dependence, with middle and upper stratospheric values increasing towards higher latitudes. The positive bias in stratospheric ACTM CH$_4$ mixing ratios causes a positive ACTM bias in XCH$_4$. In contrast to that, we find negative model–MIPAS differences in stratospheric CH$_4$ mixing ratios for TM5 (Fig. 3b), resulting in a small negative XCH$_4$ bias. We identify two altitude regions, where TM5 modeled CH$_4$ mixing ratios are smaller than MIPAS CH$_4$ mixing ratios: the lower stratosphere with differences in CH$_4$ mixing ratios of up to $-100$ ppb, and the upper stratosphere (> 30 hPa) with maximum CH$_4$ differences of $\sim -150$ ppb. Figure 3c shows the CH$_4$ mixing ratio differences between LMDz and MIPAS with noticeable negative CH$_4$ differences of up to $-200$ ppb within the tropical upper stratosphere. Negative CH$_4$ differences ($\sim -100$ ppb) are also visible in the upper stratosphere of the midlatitude and high-latitude region. In contrast to this, we identify positive CH$_4$ differences of up to 100 ppb within the middle stratosphere ($\sim 50$ hPa) of the midlatitudes and high latitudes. The negative and positive CH$_4$ differences partially cancel out in XCH$_4$. Similarly to Fig. 3, the CH$_4$ differences between model and MIPAS_ACE fields are illustrated in Fig. 4. Given the offset adjustment of MIPAS to ACE-FTS (see Fig. 1), the MIPAS_ACE CH$_4$ fields comprise lower CH$_4$ mixing ratios compared to MIPAS, mostly in the lower stratosphere. Hence, the ACTM–satellite CH$_4$ difference is larger for MIPAS_ACE fields than for MIPAS fields. For TM5 and LMDz, model–satellite CH$_4$ differences are shifted into the positive direction (Fig. 4b and c). In other words, modeled stratospheric CH$_4$ mixing ratios appear to be too high when compared to MIPAS and too low in comparison to MIPAS_ACE.

The zonal difference fields between model and satellite-based CH$_4$ data sets have also been converted to XCH$_4$ differences and are shown in Fig. 5. Two main features can be found in Fig. 5: (i) the XCH$_4$ difference range between the two satellite-based data sets MIPAS (dark red) and MIPAS_ACE (light red), which is $\sim 27$ ppb (1$\sigma$ standard deviation (SD) = 4 ppb) on annual mean basis; and (ii) the model–satellite XCH$_4$ differences, which indicate the latitudinal dependence of ACTM (Fig. 1a) and LMDz (Fig. 1c). For example, ACTM–satellite XCH$_4$ differences are clearly increasing toward higher latitudes. In contrast to this, the TM5–satellite XCH$_4$ difference does not show a latitudinal dependence. These findings on the latitudinal dependence of model–satellite XCH$_4$ differences are supported by Table 5, which provides some statistical results. For example, the SDs and the minimum–maximum ranges of model–satellite XCH$_4$ differences are much smaller for TM5 compared to the other models. Besides that, Fig. 5 also shows that the model–satellite XCH$_4$ differences for the year 2010 only slightly depend on season. A noticeable seasonal variation in the model–satellite XCH$_4$ differences can be found in the tropical/subtropical region of the Northern Hemisphere. However, in order to analyze seasonal variations, a more thorough analysis is needed, including model and satellite-based CH$_4$ data sets with a larger time period than used in this study. Furthermore, in the context of seasonality the role of TCCON station elevation needs to be considered in more detail. Since we only apply 1 year of model and satellite data, the focus of this study is not on the seasonal agreement between model and satellite-based XCH$_4$ data sets.

Modeled stratospheric CH$_4$ fields have been directly replaced by satellite data sets. As a result, there can be discontinuities in the merged CH$_4$ fields around the tropopause,
Table 5. Average XCH$_4$ differences between model simulations and model CH$_4$ fields with satellite-based stratospheric CH$_4$ fields. Annual mean differences as XCH$_4$ bias (with 1σ SD) and minimum–maximum range of zonal XCH$_4$ differences are in ppb.

<table>
<thead>
<tr>
<th>Satellite data</th>
<th>ACTM</th>
<th>TM5</th>
<th>LMDz</th>
</tr>
</thead>
<tbody>
<tr>
<td>MIPAS</td>
<td>22.3 (±14.1)</td>
<td>45.2</td>
<td>-13.9 (±3.4)</td>
</tr>
<tr>
<td>MIPAS_ACE</td>
<td>48.7 (±11.0)</td>
<td>35.4</td>
<td>13.6 (±3.5)</td>
</tr>
</tbody>
</table>

Figure 5. Zonal XCH$_4$ differences resulting from model–satellite differences of stratospheric CH$_4$ volume mixing ratios. Mean XCH$_4$ differences are shown as solid lines for the summer period (June, July, and August) and as dashed lines for the winter period (December, January, and February).

where the lowest satellite-based CH$_4$ mixing ratios strongly deviate from the original modeled CH$_4$ mixing ratios. In order to quantify the impact of these discontinuities on the XCH$_4$ data sets, we have also performed a smoother replacement method. For this purpose we defined a vertical transition range of 75 hPa, starting at the lowest vertical MIPAS data grid point. From this position the model vertical profile of CH$_4$ mixing ratios was linearly interpolated to the satellite-based CH$_4$ mixing ratio profile, starting at the upper boundary of the transition range. This method was applied to each latitudinal MIPAS grid point corresponding to a vertical profile of CH$_4$ mixing ratios. The method was not used if the model–satellite difference of CH$_4$ mixing ratios was smaller than 30 ppb at the lower boundary of the transition range. Consequently, we also computed XCH$_4$ differences between the original model and the smoothed satellite-based data sets. Figure 6 then shows model–satellite XCH$_4$ differences resulting from the force replacement (solid lines) and from the smoothly interpolated replacement (dashed lines).

Figure 6. Zonal XCH$_4$ differences as a result of model–satellite differences of stratospheric CH$_4$ volume mixing ratios. Solid lines refer to the merged model–satellite CH$_4$ fields, including discontinuities at the model–satellite transition zone around the tropopause. Dashed lines refer to merged model–satellite CH$_4$ fields that have been smoothly interpolated at the model–satellite transition zone.
From Fig. 6 it is obvious that the impact of the smoothly interpolated replacement on the model–satellite XCH$_4$ differences is small; i.e., differences between solid and dashed lines are typically smaller than 4 ppb. For this reason we expect that the impact of discontinuities in the merged model–satellite CH$_4$ fields on the results of the XCH$_4$ validation against TCCON is negligible.

5 Discussion

Our analysis shows that the model–TCCON agreement in XCH$_4$ critically depends on the model representation of stratospheric CH$_4$, which is diverse for the presented CTMs. In the following we discuss possible causes for the inter-model spread in stratospheric CH$_4$. In addition to that, we evaluate the findings of our XCH$_4$ model–TCCON comparison with respect to satellite data uncertainty.

5.1 Model transport characteristics as possible cause for inter-model spread in stratospheric methane

An inter-model spread in stratospheric CH$_4$ fields has already been detected by Patra et al. (2011) despite applying uniform fields of OH, Cl, and O$_3$D for all models. Their findings, therefore, suggested a predominant role of transport in the simulation of CH$_4$ vertical distributions. For this reason, here we tested whether differences in the modeling of stratospheric transport are noticeable. To do this, we follow the approach of Strahan et al. (2011) who sought to understand chemistry–climate model ozone simulations using transport diagnostics. This method is based on the compact relationship between a long-lived stratospheric tracer and mean age in the lower stratosphere. In their work, they compared simulations and air-borne observations of N$_2$O/mean age correlations, in order to evaluate the model transport characteristics. Here, we use the MIPAS data of CH$_4$ and mean age as a reference to identify model-to-model differences in the simulation of stratospheric transport. The MIPAS data are not used to evaluate whether modeled stratospheric circulations are realistic or not, given the uncertainties of MIPAS CH$_4$ and mean age data. For example, the MIPAS mean age range may be too large because MIPAS mean age can be up to 0.8 years too old due to the impact of mesospheric SF$_6$ loss (Stiller et al., 2012). This loss process was not included in the models used for this study. Moreover, the MIPAS CH$_4$ data significantly differ from ACE-FTS CH$_4$ data within the lower stratosphere (see Fig. 1).

In analogy to Strahan et al. (2011) the model transport diagnostics are focused on the tropical domain because tropical diagnostics quantities allow a better assessment of the individual transport processes’ ascent and mixing. Annual means of age for modeled as well as MIPAS-observed fields were calculated for the lower stratosphere (30–100 hPa) of the tropical domain (10$^\circ$ S–10$^\circ$ N), and of the northern hemisphere midlatitude region (35–50$^\circ$ N), respectively. Subsequently, vertical profiles of mean model–MIPAS differences were calculated to provide insight into the tropical transport characteristics. Figure 7 illustrates that the model–MIPAS difference of tropical mean age is almost identical for all models; i.e. the model simulations produce similar mean ages that are younger than MIPAS-observed mean ages. Knowing that mean age represents the combined effects of ascent and mixing, we separately look at the tropical ascent rate, which is assessed by the horizontal mean age gradient, calculated as the difference between midlatitude and tropical mean ages. The model–MIPAS difference of the tropical ascent rate is shown in Fig. 8, indicating that ACTM and LMDz simulate tropical ascent in a similar way. The TM5-modeled tropical ascent is faster compared to ACTM and LMDz. Finally, these model transport diagnostics indicate model-to-model differences in the simulation of tropical ascent, which are likely to cause an inter-model spread in model stratospheric CH$_4$ fields.

Indeed, model-to-model differences affecting the simulation of stratospheric transport are present in the vertical/horizontal resolution, sub-grid-scale physical parameterizations, advection schemes, and numerical methods, etc. Furthermore, the simulation of stratospheric transport depends on the reanalysis data used to drive the model meteorology; e.g., the ECMWF reanalysis data set ERA-Interim leads to an improved representation of the stratospheric circulation in comparison to the older ERA-40 reanalysis data (Monge-Sanz et al., 2007, 2013; Diallo et al., 2012). The ERA-Interim data are used by TM5 and LMDz, whereas ACTM applies 

![Tropical mean age grid](figure7.png)

Figure 7. Model–MIPAS differences of mean age for the tropical lower. Mean age data in years (yr) are calculated as annual means on the MIPAS pressure–latitude grid.
found a similar high bias for MIPAS CH$_4$ data in comparison to satellite-based CH$_4$ observations from SCIAMACHY or HALOE (HALogen Occultation Experiment). Furthermore, they showed that surface measurements provide CH$_4$ mixing ratios with slightly lower values than MIPAS-retrieved CH$_4$ mixing ratios of the upper troposphere, a finding that is against expectation. For these reasons, it is likely that our satellite data range is dominated by high biased lower stratospheric MIPAS CH$_4$ data. Thus, the model correction with ACE-FTS-based CH$_4$ fields seems more reliable. However, a definite assessment of the satellite data accuracies is not possible yet due to the lack of an extensive observational data set based on stratospheric in situ measurements.

6 Summary and conclusions

This study analyzed the importance of uncertainties in stratospheric CH$_4$ in comparisons of modeled and TCCON observed XCH$_4$. Modeled stratospheric CH$_4$ fields were substituted by satellite-retrieved CH$_4$ fields from MIPAS and ACE-FTS. Original and satellite-corrected model CH$_4$ fields were converted to XCH$_4$ and subsequently evaluated by comparison to TCCON XCH$_4$ observations from 11 sites. This approach and the statistical analysis of XCH$_4$ model–TCCON residuals were conducted with three well-established CTMs: ACTM, TM5 and LMDz.

Our model–TCCON XCH$_4$ intercomparison reveals an inter-model spread in XCH$_4$ bias caused by an inter-model spread in stratospheric CH$_4$. For ACTM we find a large average XCH$_4$ bias of 38.1 ppb, in contrast to small average XCH$_4$ biases of 8.7 ppb for TM5 and 6.8 ppb for LMDz. The ACTM XCH$_4$ bias is reduced by the model correction to 13.7 ppb with MIPAS, and to 3.3 ppb with MIPAS adjusted to ACE-FTS, respectively. For TM5 and LMDz the impact of the model correction with satellite-based CH$_4$ fields is ambiguous, in that the model XCH$_4$ bias can be slightly reduced to 4.3 ppb with MIPAS, but can also be increased to 10.8 ppb for TM5 and 20.0 ppb for LMDz with MIPAS adjusted to ACE-FTS. This implies that for TM5 and LMDz the model representation of stratospheric CH$_4$ is located within the satellite data range mapped by MIPAS and ACE-FTS observations. The annual mean differences between the two satellite-based stratospheric CH$_4$ fields yield a global XCH$_4$ difference range of ~27 ppb.

Possible causes for the inter-model spread in stratospheric CH$_4$ have been discussed with an emphasis on model transport characteristics. Applying tropical transport diagnostics suggests that the poor representation of stratospheric CH$_4$ by ACTM originates from errors in the simulation of transport pathways into and within the stratosphere. However, this is only an interpretation based on a diagnostic and requires more process-oriented model evaluation of stratospheric transport. The inter-model spread in stratospheric CH$_4$ could be quantitatively investigated with a main focus
on model-to-model differences in the simulation of stratospheric transport (physical parameterizations, reanalysis data sets, vertical/horizontal resolution); e.g., model simulations could be performed with different reanalysis data sets, and/or different physical parameterizations, resulting in a model ensemble for each CTM or a multi-model ensemble consisting of multiple CTM data sets. This would allow the individual model errors in stratospheric CH$_4$ to be assessed more precisely.

Overall we state that there is a need for improvement in modeling of stratospheric CH$_4$ and, thus, XCH$_4$. At the same time, a better quantification of model errors in stratospheric CH$_4$ is limited by the uncertainty of satellite data products as used in this study. This implies that more stratospheric CH$_4$ in situ observations are required to validate both satellite-retrieved and modeled CH$_4$ data. A more accurate evaluation of modeled stratospheric CH$_4$ fields is particularly reasonable as these CTMs are used to invert CH$_4$ emissions from XCH$_4$ data. As surface emission signals in XCH$_4$ are small compared to co-resident XCH$_4$ atmospheric background levels, it is necessary to identify minor XCH$_4$ biases in the model as done in this study. Of course, an analogous quality requirement is also needed for ground-based and satellite-borne XCH$_4$ data. Indeed, as long as unallocated and poorly understood differences of several parts per billion remain between satellite-borne XCH$_4$ data and optimized model fields, it is difficult to make full benefit of satellite XCH$_4$ data to robustly retrieve regional methane emissions.

7 Data availability

TCCON data are publicly available at http://www.tccon.caltech.edu/; please follow the data use policy described there. For obtaining the model data used in this work, contact Prabir Patra (prabir@jamstec.go.jp) for ACTM, Sander Houweling (S.Houweling@uu.nl) for TM5, and Philippe Bousquet (philippe.bousquet@lsc.ei.phl.fr) for LMDz. MI-PAS and ACE satellite data are available from the official websites after signing a data protocol.

Acknowledgements. We thank H. P. Schmid (KIT/IMK-IFU) for his continual interest in this work. Our work has been performed as part of the ESA GHG-cci project via subcontract with the University of Bremen. In addition we acknowledge funding by the EC within the InGOS project. A part of work at JAXA was supported by the Environment Research and Technology Development Fund (A-1102) of the Ministry of the Environment, Japan. From 2004 to 2011 the Lander TCCON program was funded by the New Zealand Foundation of Research Science and Technology contracts CO1X0204, CO1X0703 and CO1X0406. Since 2011 the program has been funded by NIWA’s Atmosphere Research Program 3 (2011/13 Statement of Corporate Intent). The Darwin and Wollongong TCCON sites are funded by NASA grants NAG5-12247 and NNG05-GD07G, and Australian Research Council grants DP140101552, DP110103118, DP0879468, LE0668470, and LP0562346. We are grateful to the DOE ARM program for technical support at the Darwin TCCON site. The Białystok and Orléans TCCON sites are funded by the EU projects InGOS and ICOS-INWIRE, and by the Senate of Bremen. Nicholas Deutscher was supported by an Australian Research Council fellowship, DE140100178. We are also grateful to P. O. Wennberg for providing TCCON data.

The Atmospheric Chemistry Experiment (ACE), also known as SCISAT, is a Canadian-led mission mainly supported by the Canadian Space Agency and the Natural Sciences and Engineering Research Council of Canada.

The article processing charges for this open-access publication were covered by a Research Centre of the Helmholtz Association.

Edited by: H. Worden
Reviewed by: C. Frankenberg and one anonymous referee

References


A. Ostler et al.: Evaluation of column-averaged methane in models and TCCON

4858


Monteil, G., Houweling, S., Butz, A., Guerlet, S., Schepers, D., Hasekamp, O., Frankenberger, C., Scheepmaker, R., Aben, I., and Röckmann, T.: Comparison of CH₄ inversions based on 15


