

Methane variability in the upper troposphere and lower stratosphere and their relevance for emission inversions constrained by satellite observations

Michiel van Weele*

Chemistry and Climate Division, Royal Netherlands Meteorological Institute, PO Box 201, 3730 AE, De Bilt, The Netherlands;
Phone: +31 (0)30 2206410; Fax: +31 30 2210407; E-mail: weelevm@knmi.nl

Jason E. Williams

Chemistry and Climate Division, Royal Netherlands Meteorological Institute, PO Box 201, 3730 AE, De Bilt, The Netherlands;
Phone: +31 (0)30 2206418; Fax: +31 30 2210407; E-mail: williams@knmi.nl

Peter F.J. van Velthoven

Chemistry and Climate Division, Royal Netherlands Meteorological Institute, PO Box 201, 3730 AE, De Bilt, The Netherlands;
Phone: +31 (0)30 2206419; Fax: +31 30 2210407; E-mail: velthove@knmi.nl

Tanja J. Schuck

Atmospheric Chemistry Division, Max Planck Institute for Chemistry, Joh.-Joachim-Becher-Weg 27, D-55128, Mainz, Germany;
Phone: +49 (0) 6131 305-416; Fax: +49 (0) 6131 305-436; E-mail: tanja.schuck@mpic.de

Carl A.M. Brenninkmeijer

Atmospheric Chemistry Division, Max Planck Institute for Chemistry, Joh.-Joachim-Becher-Weg 27, D-55128, Mainz, Germany;
Phone: +49 (0) 6131 305-453; Fax: +49 (0) 6131 305-436; E-mail: carl.brenninkmeijer@mpic.de

* corresponding author

Abstract

Observation-based constraints on methane (CH₄) surface emissions can be obtained from measurements of atmospheric CH₄ concentrations. Small variations in mixing ratio can be traced back to variations in surface fluxes via *inverse modeling*. However, surface emissions are not the only source of CH₄ concentration variations in the atmosphere. Atmospheric variability in CH₄ – at all altitudes – has to be well simulated by the forward model that is used in inversions based on satellite-borne observations of the total column mixing ratio (CMR) of CH₄. In this study we examine the CH₄ variability in the upper troposphere and lower stratosphere (UTLS region) between ~6-25 km altitude. By comparing global chemistry-transport model simulations with *in-situ* aircraft observations of CH₄ it is shown that in the UTLS region over Europe variability in winter is predominantly controlled by atmospheric processes. In summer, and at lower latitudes, CH₄ surface emissions are effectively transported to the UTLS region by convection and, subsequently, quasi-horizontally transported over large distance. Simulated CH₄ variations in the UTLS critically depend on timing, location and the strength of convective uplift in combination with the assumed distribution and seasonal variability of the emissions. Other processes contributing to the observed variability in the UTLS are downward transport from the stratosphere, variations in tropopause height and long-range transport. Satellite-based CH₄ vertical profile information has the potential to reduce model uncertainties and provide additional constraints for CH₄ emission estimates based on variations in total CMR.

Keywords

Methane, inversion, satellite observations, global modeling, aircraft observations

1. Introduction

Methane (CH₄) is one of the *well-mixed* greenhouse gases. However, small regional variations in the atmospheric CH₄ concentrations occur throughout the atmospheric column. Such variations introduce associated variations in radiative forcing and, more importantly, contain useful information about the spatial distribution and temporal variation of CH₄ surface emissions. This information is what is exploited in *inverse modeling*. This tool allows observation-based (top-down) constraints to be placed on surface emissions that are otherwise based on (bottom-up) compilations of a composite of anthropogenic inventories (e.g. EC, 2009), biomass burning inventories (e.g. van der Werf et al., 2006), process modeling of natural fluxes from wetlands and rice paddies (e.g. Spahni et al., 2011) and uncertain up-scaling of local emission processes, e.g. from termites (Sanderson, 1996).

The aim of this study is to examine the atmospheric transport and mixing processes that contribute to the variability in the CH₄ total column mixing ratio (CMR) that is provided by present-day satellite observations such as those from SCIAMACHY on the ESA Envisat platform (Frankenberg et al., 2008). These atmospheric processes need to be well-represented by the forward model that is used to perform an accurate inversion. The forward model is usually a chemistry-transport model (CTM) that calculates the CH₄ concentration variations in the atmosphere for given distribution in surface emissions. Any bias or misrepresentation in the forward model may be inadvertently transferred to the derived surface emissions during the inversion (e.g. Meirink et al., 2008a).

In the free troposphere long-range transport and convection of surface emission plumes affect the total CMR simulated by the forward model. The shorter chemical lifetime of CH₄ in the lower stratosphere induces a profile in the vertical distribution, resulting in CH₄ variability in the upper troposphere following stratosphere-troposphere exchange. The tropospheric chemical lifetime of CH₄ is relatively long (~ 8 to 9 years, Stevenson et al., 2006) meaning biases in the chemical oxidation by the OH radical will affect inversions at large scale, e.g. with respect to the derived latitudinal distribution of the annual mean emissions.

Using aircraft observations in the UTLS region we evaluate the variability in the distribution of CH₄ in the global CTM TM5 (Krol et al., 2005; Huijnen et al., 2010). TM5 has been used as forward model in recent inversion studies, where it is constrained by total CMR observations from SCIAMACHY in tandem with *in situ* surface observations (e.g. Meirink et al., 2008b; Bergamaschi et al., 2009). Because the global surface network is sparse and has principally been set-up to observe variations in background CH₄ levels,

the total CMR observations provide crucial input to the emission inversions. Therefore it is important to analyze the variability in the total CMR as simulated by TM5. Multi-year simulations have been performed with the full tropospheric chemistry version of TM5 (Huijnen et al., 2010). The model set-up and the CH₄ inventories for the anthropogenic and natural CH₄ emissions are described in Section 2. The *in situ* observational aircraft datasets against which TM5 is evaluated is described in Section 3. Our results are presented in Section 4. Finally, the contribution of atmospheric processes to the variability in the total CMR simulated by TM5 is discussed in Section 5.

2. TM5 model set-up and CH₄ boundary conditions

This study exploits two decadal (2000-2009) global CTM simulations performed with the TM5 model on 3° x 2° resolution (lon x lat) with 34 levels up to 0.5 hPa. The TM5 model is driven off-line by ECMWF meteorological fields of the ERA-Interim reanalysis (Dee et al., 2011). A spin-up period of 4 years was used. A comprehensive description and benchmark evaluation of the tropospheric chemistry version of TM5 (Tracer Model 5, version TM5-chem-v3.0) used in this study is given in (Huijnen et al., 2010) and references therein, including the chemical mechanism, details on the tracer transport, the applied non-methane emissions, and extensive evaluation against observations for carbon monoxide and tropospheric ozone, as well as of the OH field ultimately determining CH₄ chemical lifetime.

A major difference between the TM5 model version described in Huijnen et al. (2010) and the version used in this work is the surface boundary condition used for constraining the distribution of CH₄. The *BASE* run uses the “traditional” approach where instantaneous forcing to a zonal-mean CH₄ surface climatology is performed, with the climatology assembled using from measurements taken at global background surface networks. Latitudinal interpolation between five remote stations (Barrow, Alaska; Mauna Loa, Hawaii; American Samoa; Cape Grim, Tasmania; South Pole) was used for the construction of this surface background. Given the long atmospheric chemical lifetime of CH₄ (~ 8-9 years, Stevenson et al. (2006)) this method of applying boundary conditions to constrain tropospheric CH₄ distribution has previously been considered adequate to provide sufficient accuracy to study tropospheric chemistry and composition changes. However, that the climatology is assembled using background measurements can result in a significant underestimation of the seasonal cycle of CH₄ concentrations near source regions (Williams et al., 2011).

The *CH4-HYBRID* run uses CH₄ surface emission inventories in combination with relaxation to observed surface concentrations in the background atmosphere. The CH₄ emission data sets which are adopted are the EDGAR V4.0 anthropogenic emissions (EC, 2009) between 1999 and 2005 (after which we use the estimates for 2005 due to availability), GFEDv2 observation-based biomass burning emissions (van der Werf et al., 2006) and the net natural surface CH₄ fluxes from Spahni et al.(2011) between 1999 and

2008. The loss of CH₄ via microbial oxidation in soils is also accounted for. Minor natural sources include emissions from termites (Sanderson, 1996), wild animals (Olson, 1997) and the ocean (Lambert and Schmidt, 1993). The net natural fluxes from Spahni et al. (2011) vary as a function of the observed meteorology (temperature, precipitation). The global net natural emissions show an annual cycle which peaks from June to October. This maximum is caused by the peak in the net natural fluxes from the Indo-Gangetic plains during the South Asian monsoon (Spahni et al., 2011). At the surface, relaxation to background concentrations is performed in the *CH4-HYBRID* run. This relaxation prevents a model drift in the global CH₄ burden related to further uncontrolled differences between the applied total global CH₄ emission fluxes and the chemical oxidation by OH. The surface concentration at the dateline is used for determining the relaxation to the observed background. At the dateline the surface concentrations are assumed to be representative of the background atmosphere, i.e. the minimum in the zonal band. This zonal minimum-to-background relaxation is then applied every time-step across the entire zonal band using a 3-day relaxation time, thereby maintaining the distribution in the meridional concentration for each specific latitudinal band as determined by the distribution in surface emissions. To be effective for the global burden the relaxation is applied to the 10 lowest model layers up to ~500 hPa. The differences between the *BASE* and *CH4-HYBRID* runs in terms of tropospheric composition, oxidative capacity and chemistry budgets are analyzed in Williams et al. (2011).

The top boundary condition for CH₄ in the stratosphere is relaxation using a 45 days time scale towards a zonal-mean satellite climatology (Grooss and Russell, 2005) above 45 hPa (90 hPa) level in the tropics (extra-tropics). This relaxation gives a first-order approximation for the stratospheric chemical loss of CH₄ by the reaction with OH, O(¹D) and Cl, and corrects for potential long-term biases in the stratospheric circulation of the ERA Interim reanalysis. Using a 45 days timescale ensures that the relaxation does not significantly affect the dynamically-induced short-term variability in the UTLS-region.

3. Aircraft observations in the UTLS-region

The aircraft observations of CH₄ that are used for performing comparisons with the distribution of CH₄ in the TM5 simulations include those made as part of the SPURT campaign between November 2001 and July 2003 over Europe and North Africa (Engel et al., 2006) and those made as part of the CARIBIC project during 2008 (Schuck et al., 2010).

The SPURT flights were based from Hohn in northern Germany (54.3° N; 9.5° E). The primary scientific goal was to investigate how the trace gas distribution in the UTLS varies with both latitude and season, where the data would allow insight into the dynamical and chemical processes that govern the variability of trace gas mixing ratios in this region (Engel et al., 2006). Eight measurement campaigns (consisting of a total of 36 flights) were performed, distributed over all seasons and typically covering latitudes

between 35° N and 75° N in the European longitude sector (10° W–20° E). The flights reached altitudes up to 13.7 km, penetrating the lower stratosphere in northern Europe.

The flights from CARIBIC (Civil Aircraft for the Regular Investigation of the atmosphere Based on an Instrument Container; www.caribic-atmospheric.com) used in this study were based from Frankfurt, Germany (50.0° N; 8.5° E). Between April and December 2008, in total 16 measurement flights were performed by the CARIBIC aircraft between Germany and Chennai (India). A distinct monsoon signature in CH₄ was observed in the longitude range 50° – 80° E south of 40° N at flight altitudes 8–12.5 km (Schuck et al., 2010). It was argued that during the summer monsoon, strong and widespread convection occurs over India, the Bay of Bengal and over the Tibetan Plateau, partly over densely populated regions where strong emission sources are located. With typical cruising altitudes ranging from 8.5 to 12 km (corresponding to 300–180 hPa), the CARIBIC aircraft frequently crosses the tropopause at mid-latitudes. At lower latitudes the free tropical troposphere is probed.

4. Results

Daily variations in the CH₄ mixing ratio in the UTLS-region are analysed for the *CH₄-HYBRID* simulation. Figure 1 shows the CH₄ variability at the 150 hPa level (~14 km) and at the 500 hPa level (~ 6km) for the 1st Feb, 2003 and 1st Aug, 2003 to show the seasonal variability.

At the 150 hPa level the CH₄ mixing ratios are highest at tropical latitudes where the 150 hPa level is situated in the tropical tropopause layer (TTL). For most of the year the TTL is characterized by a strong easterly jet, which effectively transports convectively uplifted air from tropical emission areas (e.g. the floodplains in India) over the complete tropical belt. Filaments of upper-tropospheric tropical air mix into the lowermost stratosphere at mid-latitudes. At higher latitudes (>60°N/S) CH₄-poor stratospheric air masses fold into the upper troposphere introducing further variability. The simulated mixing ratios at the 150 hPa level typically vary between about 1400 and 1850 ppb.

At the 500 hPa level the CH₄ mixing ratios are highest at high latitudes and higher during boreal summer than winter. The range at the 500 hPa level ranges between 1730-1880 ppb, which is significantly smaller than at the 150 hPa level. The winter minimum is caused by enhanced downward transport from the stratosphere. Also the tropopause height is lower in winter than in summer. A summer maximum in mid-tropospheric CH₄ at northern high-latitude has also been seen in satellite observations (Xiong et al., 2010) and contrasts to the summer minimum observed at the surface. The mid-to-high latitude surface summer minimum is also captured in our simulations (see Figure 2) and is attributed to the seasonal cycle in the chemical loss by OH.

The two lowest panels show the effect of CH₄ emissions on the vertical concentration distribution as a function of latitude. The mean vertical cross-section during 1st Feb at 0°E

over Europe and Africa only shows marginal differences with the *BASE* run over Europe because the influence of convectively-uplifted surface emissions is rather small during this season. However, over Central Africa pockets of CH₄-rich air uplifted from the boundary layer can clearly be discerned. This effect is even more pronounced in the corresponding mean cross section for August 1st at 75°E over India. Upper tropospheric CH₄ concentrations up to ~1800 ppb are found up to the TTL around 100 hPa (~16-17 km altitude). Also at mid-latitudes over central Asia the CH₄ concentrations in *CH4-HYBRID* are enhanced by ~2-3% when compared to the *BASE* run. Once convectively uplifted, air masses can be quasi-horizontally transported over large distances. Long-range transport in the lower troposphere is the cause of the lower concentrations found in *CH4-HYBRID* compared to *BASE* around 70° N over Europe (blue colors in the lower left panel of Figure 1) which represents a filament of CH₄-poor Arctic air.

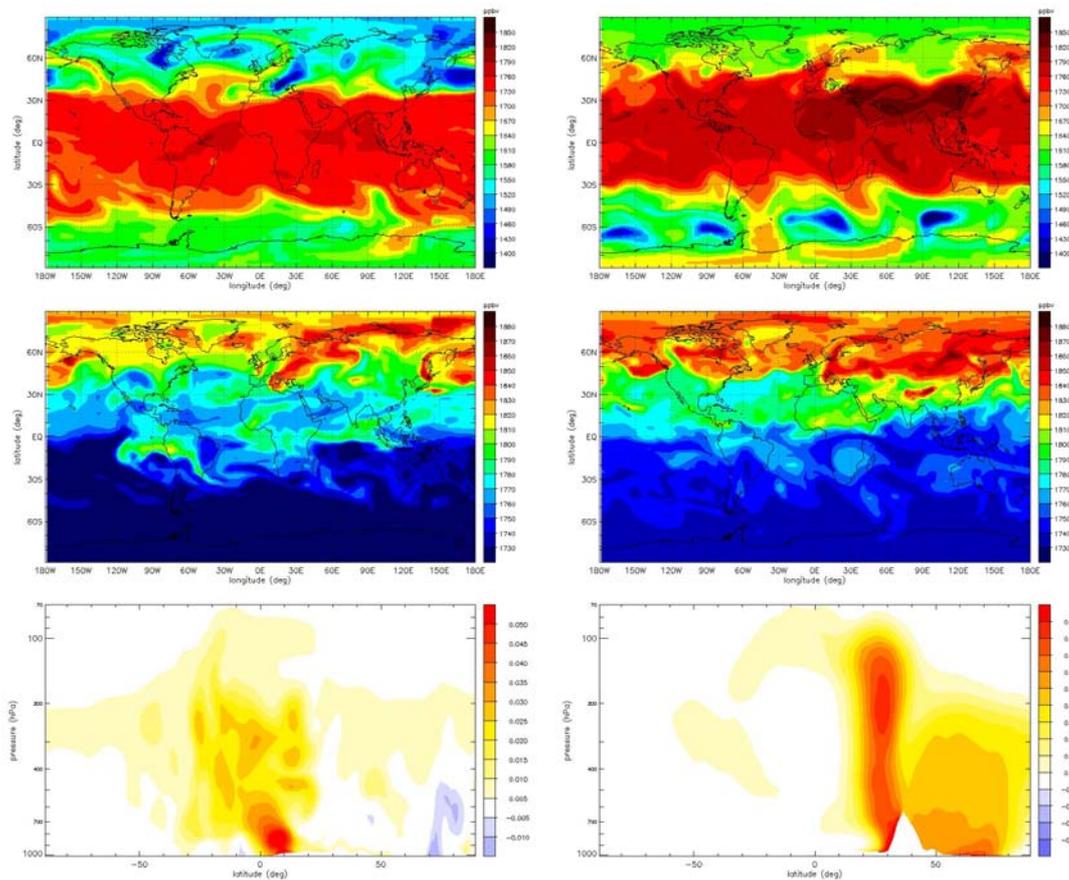


Figure 1: Global CH₄ concentration distribution on a northern winter day (1st February 2003, left) and a northern summer day (1st August 2003, right) at the 150 hPa level (top) and 500 hPa level (middle) of the *CH4-HYBRID* simulation including CH₄ surface emissions. The lower panels show the relative changes in the monthly mean CH₄ vertical profile for *CH4-HYBRID* with respect to *BASE*. Bottom left: at a longitude of 0°E over Europe for 1st February 2003. Bottom right: at 75°E over India for 1st, August 2003.

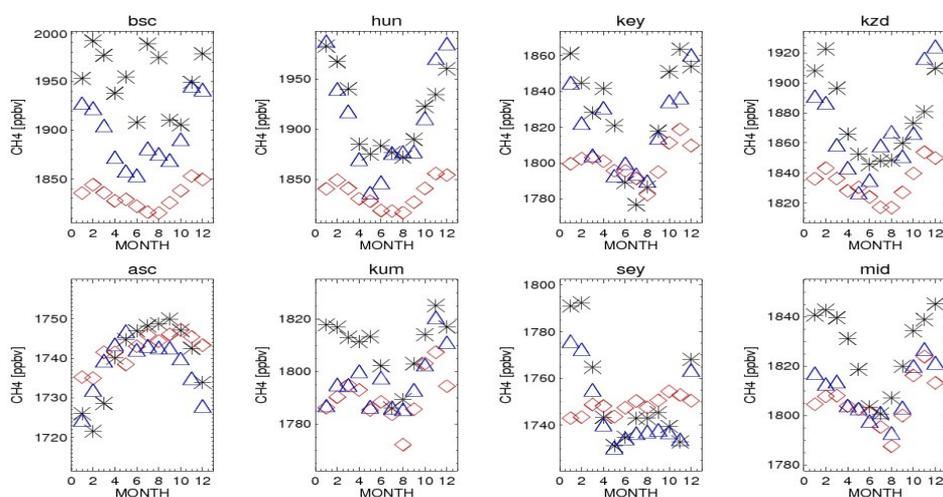


Figure 2: Comparison of monthly mean surface mixing ratios at 4 continental (upper panels) and 4 oceanic (lower panels) surface sites for the year 2003 (BSC = Black Sea Constanta, Romania; HUN = Hegyhatsal, Hungary; KEY = Key Biscayne, Florida; KZD = Sary Taukum, Kazakhstan; ASC = Ascension Island ; KUM = Cape Kumukahi, Hawaii ; SEY = Mahe Island, Seychelles; MID = Sand Island, Midway Islands)

Figure 2 shows a comparison of the monthly mean surface mixing ratios of *CH₄-HYBRID* with observations for a couple of surface sites from the NOAA CMDL global surface network (as available from: <ftp://ftp.cmdl.noaa.gov/ccg/ch4>). The differences between the *CH₄-HYBRID* and *BASE* simulation at the four continental sites show that these sites are influenced by regional distribution in *CH₄* emissions throughout the year. Improvements in the agreement between the *CH₄-HYBRID* volume mixing ratios and the observations occur at many sites, especially Hungary, both in their absolute values as well as for the amplitude of the seasonal cycle. At the oceanic sites (the four lower panels) differences between *CH₄-HYBRID* and *BASE* are much smaller. An exception is for the Dec-Jan-Feb season at the Seychelles which is clearly under influence of regional emissions at this time of the year, most probably of natural origin. It should be noted that none of these eight surface sites has been used to construct the global background distribution to which the surface concentrations are relaxed towards (c.f. section 2).

Figure 3 shows the comparison of the *CH₄-HYBRID* and *BASE* simulations with the SPURT aircraft observations in the UTLS region. The model output was sampled at the location/time of the in-situ observations and all data are binned in 2° latitude bands for each month. The variability in *CH₄* mixing ratios observed in the UTLS region during the SPURT flights over Europe is significant. Many of the observed latitudinal variations are captured in both simulations. The absolute differences between the *CH₄-HYBRID* and *BASE* simulations are smaller than those observed across the latitudinal range covered by the SPURT flights, especially in the two winter months. In July 2003 significant differences between both simulations are seen.

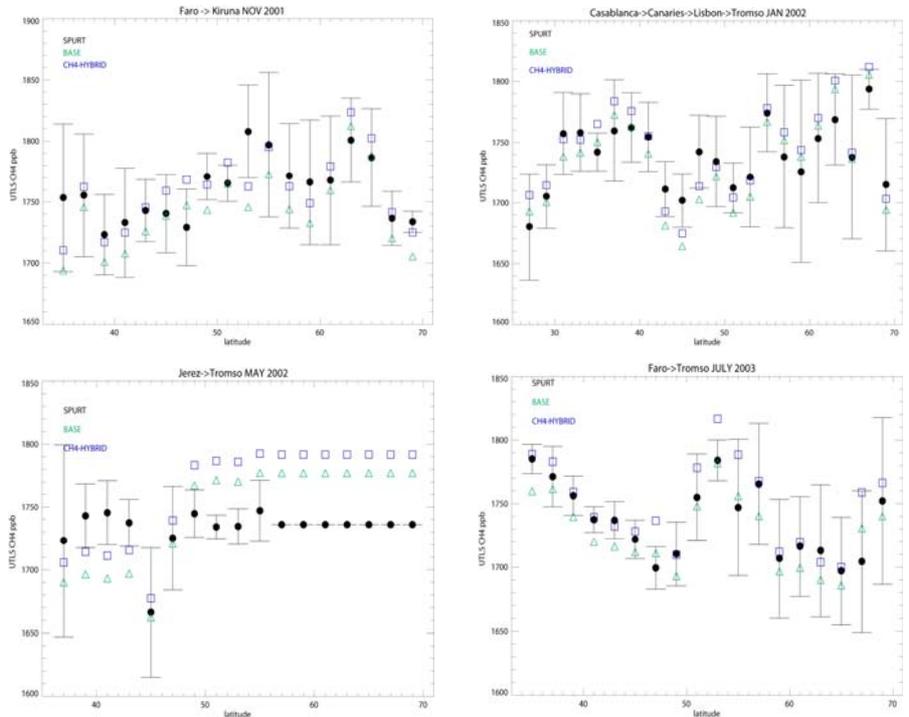


Figure 3: Comparison of *BASE* (green triangles) and *CH4-HYBRID* (blue open squares) with SPURT *in situ* CH₄ aircraft observations (black dots) in the UTLS-region over Europe. In November 2001 from Faro to Kiruna (top left), in January 2002 from Casablanca to Tromsø (top right), in May 2002 from Kiruna to Lisbon (bottom left) and in July 2003 from Faro to Tromsø (bottom right).

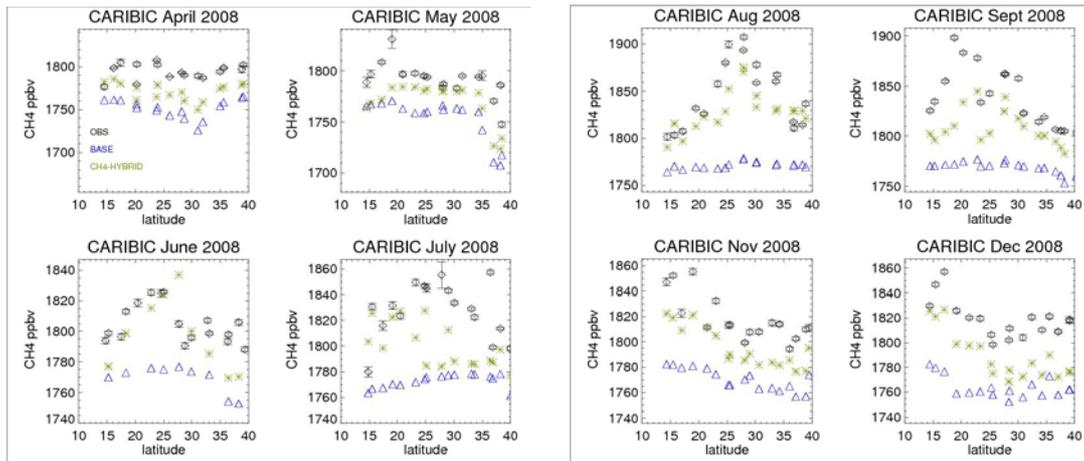


Figure 4: Comparison of *BASE* (blue triangles) and *CH4-HYBRID* (yellow-green crosses) with CARIBIC *in situ* CH₄ observations (black open dots) in the UTLS-region from Europe to Chennai, South-India during 2008.

In this instance the *CH4-HYBRID* simulation seems to overestimate the CH₄ mixing ratios at certain locations. This may be an illustration of the difficulty to correctly simulate convective uplift over emission areas.

Figure 4 shows the comparisons over Eurasia and India against the CARIBIC aircraft observations. Both observations and simulated mixing ratios are again binned as a function of latitude and month. Much larger differences between the *CH4-HYBRID* and *BASE* simulations are found here because of the large difference in the seasonal surface concentrations (Williams et al, 2011) which are convectively-uplifted from the boundary layer. The effect is most pronounced during the Asian monsoon months from June to September 2008. Many of the CH₄-rich air-masses observed by CARIBIC are captured in the *CH4-HYBRID* simulation, but missed in the *BASE* simulation. However, biases of up to 2% in volume mixing ratio between *CH4-HYBRID* and the observations do persist during some months.

5. Discussion

The comparison of two multi-year CTM simulations with TM5 against in-situ aircraft and surface observations has provided better insight into the causes of CH₄ variability that occur in the atmosphere at different altitudes. The evaluation with *in situ* aircraft observations shows that the CH₄ variability observed in the UTLS-region is related to convectively-uplifted CH₄-rich air masses from the surface, along with other transport and mixing processes including stratosphere-troposphere exchange, tropopause height variations, and long-range transport. The evaluation of both simulations at different surface sites reveal that the application of emission inventories improves the seasonal variability at many continental sites, illustrating that these sites are affected by long-range transport of regional CH₄ emissions for at least some months of the year.

The CH₄ variations in the UTLS region contribute significantly to the variations in the total column mixing ratio (CMR). The mixing ratio at the 150 hPa level varies between ~1400-1850 ppb in the *CH4-HYBRID* simulation. At the 500 hPa level the mixing ratio varies between ~1730-1880 ppb. Although the relative variations are smaller at the lower altitude, the contribution to total CMR variations is somewhat larger because of the increased air density. To illustrate this effect the contribution of CH₄ variability to the total CMR is calculated for the atmosphere above the 150 hPa level and above the 500 hPa level. The column above 150 hPa contributes ~12% to the total column and the spatial variability – represented by the standard deviation of the global mean field, is (~0.3%). The column above the 500-hPa level contributes about half (48%) to the total column. The standard deviation for the column above this level is about 0.5%. Both are significant fractions of the total CMR variations which are typically less than 2%.

Atmospheric processes lead to CH₄ variability in the upper atmosphere and need to be well captured by the forward model used for emission inversions. Forward model

uncertainties have the potential to be reduced by putting additional constraints on the model using satellite observations of the CH₄ vertical profile in the UTLS region. Such information is already becoming available from the AIRS (e.g. Xiong et al., 2010) and IASI (e.g. Razavi et al., 2009) satellite instruments and could potentially be further enhanced with CH₄ limb observations that would penetrate into the mid-upper troposphere. Applying such information in inverse modeling studies associated with determining surface emission fluxes could potentially have a significant influence.

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Figure captions

Figure 1: Global CH₄ concentration distribution on a northern winter day (1st February 2003, left) and a northern summer day (1st August 2003, right) at the 150 hPa level (top) and 500 hPa level (middle) of the *CH4-HYBRID* simulation including CH₄ surface emissions. The two lower panels show the relative changes in the monthly mean CH₄ vertical profile for *CH4-HYBRID* with respect to *BASE*. Bottom left: at a longitude of 0° E over Europe for 1st February 2003. Bottom right: at a longitude of 75° E over India for 1st, August 2003.

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