



Assimilation of
atmospheric CH₄
products

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Assimilation of atmospheric methane products in the MACC-II system: from SCIAMACHY to TANSO and IASI

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Abstract

The Monitoring Atmospheric Composition and Climate Interim Implementation (MACC-II) delayed-mode (DM) system has been producing an atmospheric methane (CH₄) analysis 6 months behind real time since June 2009. This analysis used to rely on the assimilation of the CH₄ product from the SCanning Imaging Absorption spectroMeter for Atmospheric CHartographyY (SCIAMACHY) instrument on board Envisat. Recently the *Laboratoire de Météorologie Dynamique* (LMD) CH₄ products from the Infrared Atmospheric Sounding Interferometer (IASI) and the SRON Netherlands Institute for Space Research CH₄ products from the Thermal And Near-infrared Sensor for carbon Observation (TANSO) were added to the DM system. With the loss of Envisat in April 2012, the DM system has to now rely on the assimilation of methane data from TANSO and IASI. This paper documents the impact of this change in the observing system on the methane tropospheric analysis. It is based on four experiments: one free run and three analyses from respectively the assimilation of SCIAMACHY, TANSO and a combination of TANSO and IASI CH₄ products in the MACC-II system. The period between December 2010 and April 2012 is studied. This corresponds to a period during which the performance of SCIAMACHY was deteriorating. The SCIAMACHY experiment globally underestimates the tropospheric methane by 35 part per billion (ppb) compared to the HIAPER Pole-to-Pole Observations (HIPPO) data and the methane column by 23 ppb compared the Total Carbon Column Observing Network (TCCON) data, when the global bias of the free run against the same HIPPO and TCCON data is respectively -5 ppb and 4 ppb. The assimilated TANSO product changed in October 2011 from version v.1 to version v.2.0. The analysis of version v.1 globally underestimates the tropospheric methane by 18 ppb compared to the HIPPO data and the column by 11 ppb compared to the TCCON data. In contrast, the analysis of version v.2.0 globally overestimates the column by 10 ppb. When the high density IASI data are added in the tropical region between 30° N and 30° S, the resulting analysis is slightly less biased in the troposphere than the TANSO analysis when compared to the HIPPO

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in space from which column products of CH₄ were derived. Today, CH₄ columns can be retrieved from the Tropospheric Emission Spectrometer (TES) instrument aboard NASA's Aura satellite (launched in 2004), from the Infrared Atmospheric Sounding Interferometer (IASI) instrument aboard the first European meteorological polar-orbiting satellites, MetOp-A (launched in 2006) or the Thermal And Near-infrared Sensor for carbon Observation (TANSO) aboard the Greenhouse gases Observing Satellite (GOSAT, launched in 2009). SCIAMACHY and TANSO measurements are in the near infrared (NIR) while TES and IASI measurements are in the thermal infrared (TIR). NIR methane retrievals are sensitive to the entire tropospheric column, but due to their dependence on reflected sunlight, observations are unavailable at night. TIR methane retrievals can be performed day and night, over land and ocean, and for partly cloudy scenes but they have limited sensitivity to the lower troposphere (due to lack of thermal contrast) and their precision is limited mainly by uncertainties in atmospheric temperature and surface emissivity. The combination of all these measurements of atmospheric CH₄ columns from space are complementary to the surface observation to monitor the atmospheric CH₄. They provide nevertheless only vertically integrated information and with some uncertainties.

Data assimilation provides the the ability to combine all the information from different instruments and observing networks and to combining them with a model in an optimal way. This study presents one of the two data assimilation products provided by the Monitoring Atmospheric Composition and Climate Interim Implementation (MACC-II) project and using CH₄ measurement from space. The first product combines satellite measurements of CH₄ with inverse modelling in order to optimise the surface sources and sinks of CH₄ (Bergamaschi et al., 2009). The second product combines the satellite measurements with a forward transport model in order to monitor the tropospheric CH₄. In this paper, we will focus on this second product. It uses the European Centre for Medium-Range Weather Forecasts (ECMWF) Integrated Forecasting System (IFS). Methane satellite measurements and all the other operational meteorological data are assimilated in the IFS to produce the best estimate of the atmospheric state, referred to

hereafter as the analysis. But unlike most of the data assimilated in NWP, CH₄ data are not yet available in near real time, mainly due to their partial reliance on auxiliary data. As a consequence, the assimilation system is run with a 6 month lag in a so-called “delayed mode” (DM).

5 The MACC-II DM system started producing a CH₄ analysis from 1 June 2009 onwards. This analysis was initially based on the CH₄ product from SCIAMACHY provided by the SRON Netherlands Institute for Space Research in collaboration with the Jet Propulsion Laboratory (JPL, Frankenberg et al., 2005) and the version v00.50
10 CH₄ product from TANSO provided by the Japan National Institute for Environmental Studies (NIES). This TANSO product was not mature enough at this time and its assimilation increased the bias and the variability of the analysis. Therefore the SCIAMACHY CH₄ product remained for a while the only CH₄ product assimilated in the DM system. With the loss of Envisat in April 2012, the methane analysis of the DM system had to rely on other instruments. To continue the monitoring of the atmospheric CH₄ with
15 the DM system, TANSO and IASI CH₄ products were included in the assimilation system. TANSO has similar characteristics as SCIAMACHY but provides about 10 times less data than SCIAMACHY during a given assimilation window. Its data are nevertheless more accurate than that from SCIAMACHY. Since our first attempt at assimilating TANSO data, several retrieval algorithms processing the TANSO data have been developed by several teams (Parker et al., 2011; Schepers et al., 2012; Yoshida et al.,
20 2013). The MACC-II product is jointly developed by SRON and the Karlsruhe Institute of Technology (KIT, Butz et al., 2010). As a TIR sounder, IASI provides complementary information on the mid-troposphere during day and night over land and over sea. The MACC-II IASI product is from the *Laboratoire de Météorologie Dynamique* (LMD).
25 Currently it has a good enough precision in the tropics only (Crevoisier et al., 2009). The assimilation of TES was not considered because its data cannot be assimilated in the DM system as it is not available within the expected a 6 month latency period.

The aim of this paper is to document the impact of the assimilation of TANSO and IASI CH₄ products compared to the assimilation of the SCIAMACHY product in order

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to determine how the change in the observing system would impact the DM analysis. Section 2 provides more details on the assimilated data and describes the assimilation system as well as the experiments we ran to assess the impact of each data set on the analysis. Section 3 presents the results of our assimilation experiments and their comparison with independent measurements of atmospheric CH₄. The last section addresses the question of whether the combination of TANSO and IASI products is a good candidate to continue the monitoring of methane in our analysis system.

2 Analysis experiments

For the period between November 2010 and April 2012 we ran three data assimilation experiments based on the following CH₄ products: 1. SCIAMACHY; 2 TANSO; 3 TANSO and IASI. These experiments are referred hereafter as to SCIA, TANSO and IASI+TANSO, respectively (Fig. 1). To properly assess the impact of the assimilation of each product, we also ran for this period an experiment without assimilation of CH₄ observations. This experiment is referred to as the free run FREE. The free run and the assimilation system are described in the following subsection. Then we present the three assimilated data sets. The last subsection details the experiments we ran.

2.1 Analysis system

The MACC-II greenhouse gases DM system is based on ECMWF IFS. It runs at a global horizontal resolution of T511 (≈ 40 km) with 60 vertical levels from the surface up to 0.1 hPa. Because this horizontal resolution is too costly for long assimilation experiments, we chose in this study to keep the configuration of the DM system but to decrease the horizontal resolution to T255 (≈ 80 km), which is sufficient for resolving the large scale horizontal structures of the atmospheric CH₄ fields.

Based on the default IFS configuration, all the observations (operational and methane) are assimilated within the 12 h 4-D-Var window to constrain the meteoro-

logical variables as well as the atmospheric CH₄. Methane mass mixing ratio is directly transported within IFS as a tracer. But its evolution is also determined by fluxes and chemical sinks. Methane fluxes are prescribed in IFS using inventory and climatological data sets. These fluxes are consistent with those used as prior information in the CH₄ flux inversions from Bergamaschi et al. (2009). The anthropogenic fluxes are from the EDGAR 4.2 database (Janssens-Maenhout et al., 2012) for the year 2008, i.e. the last available year. All the anthropogenic categories are based on annual mean values, except for rice, which has been modulated with a seasonal cycle of the Matthews monthly inventory for rice (Matthews et al., 1991). The wetland fluxes are from the Kaplan climatology described in Bergamaschi et al. (2007). The biomass burning emissions are from the MACC-II GFAS dataset (Kaiser et al., 2012). The other sources/sinks include wild animals (Houweling et al., 1999), termites (Sanderson, 1996), oceans (Houweling et al., 1999; Lambert and Schmidt, 1993) and a soil sink (Ridgwell et al., 1999). For the chemical sink in the troposphere and the stratosphere, the climatological chemical loss rates from Bergamaschi et al. (2009) are used. These are based on OH fields optimised with methyl chloroform using the TM5 model (Krol et al., 2005) and prescribed concentrations of the stratospheric radicals using the 2-D photochemical Max-Planck-Institute model.

The background and observation error covariance matrices for the CH₄ assimilation are specified as for all the IFS meteorological variables. The background error is set to be constant in time. Its standard deviation (diagonal of the covariance matrix) is about 20 ppb in the troposphere and increases to a value of 150 ppb at the surface. The correlations are constant over the whole domain with a representative length scale of about 200 km. The system does not account for the spatial or temporal correlation between the errors of the observations. The observation error standard deviation is provided with the data and are specified later on for each used data set.

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2.2 Assimilated data

The MACC-II DM system is able to assimilate various kinds of data sets like radiances or geophysical products with and without averaging kernels. For methane, the choice was made to assimilate retrievals products in order to lower the level of complexity imposed by the radiance assimilation.

2.2.1 SCIAMACHY

SCIAMACHY measured the direct light from the sun as well as the solar light reflected by the Earth. The difference provides information on the atmosphere through which the Earth-reflected light has passed, especially information on the concentration of trace gases and aerosols in the troposphere and stratosphere. SCIAMACHY measured the light between 240 and 1700 nm, and in certain spectra between 2000 and 2400 nm at a moderate spectral resolution (0.2 to 0.5 nm).

Envisat flew in a sun synchronous polar orbit at an altitude of 790 km. It orbited the Earth in about 101 min with a repeat cycle of 35 days. Along the orbits, SCIAMACHY used different observation modes: nadir, limb, and solar or lunar occultation. For the nadir mode which is used in this study the swath was 960 km. For the methane product, the pixel size on the ground was 30 km (along track) by 60 km (across track). Per 12 h assimilation window SCIAMACHY provided about 1200 CH₄ data points with a distribution as shown in Fig. 2. In autumn the best observed places are the continents over a large tropical band. For example over Australia more than 10 observations per model grid point are provided for the whole month of October 2011. North Africa is also well observed with several model grid points having about 25 observations. Oceans south of 30° S are better observed than other oceans, but they have on average less than 5 observations per month. Later on in the season and until January, the coverage shifts to the South due to the dependence of the instrument on the sunlight. Early on and until June the coverage shifts to the North. But the main features of the coverage remain the same as for October 2011, the month displayed in Fig. 2.

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The column-averaged dry-air mole fractions of methane ($x\text{CH}_4$) product from SCIAMACHY used in this study is provided by an algorithm based on the proxy method developed at SRON in collaboration with JPL (Frankenberg et al., 2005, 2011). The algorithm uses the 1600 nm band of SCIAMACHY. In this paper, we used version v5.5 of the product. The product provides a dry molar fraction column of CH_4 with an a priori profile of CH_4 and its corresponding averaging kernel. Both the a priori and the averaging kernel have 12 vertical levels from the lower troposphere to the top of the atmosphere (Fig. 3). As shown by the shape of the averaging kernel, the retrieved $x\text{CH}_4$ from SCIAMACHY are more sensitive to the lower troposphere.

The proxy method provides $x\text{CH}_4^{\text{proxy}}$ based on the ratio between the CH_4 number column ($v\text{CH}_4$) and the CO_2 number column ($v\text{CO}_2$),

$$x\text{CH}_4^{\text{proxy}} = \frac{v\text{CH}_4}{v\text{CO}_2} \cdot x\text{CO}_2^{\text{prior}}. \quad (1)$$

The prior column-averaged dry-air mole fraction of CO_2 ($x\text{CO}_2^{\text{prior}}$) is computed from CO_2 molar fractions provided by CarbonTracker (carbontracker.noaa.gov). At the time this paper of writing, CarbonTracker provides CO_2 for 2011. But in 2010, when the 2010 $x\text{CH}_4$ SCIAMACHY product was produced, the available data set from CarbonTracker was from 2007. Assuming a yearly increase in CO_2 of 0.4%, a multiplicative factor of $(1.004)^3$ was applied to the 2007 CarbonTracker CO_2 fields in order to produce the 2010 $x\text{CH}_4$ product. When the 2011 $x\text{CH}_4$ SCIAMACHY product was produced, the 2010 release of CarbonTracker was available. So a multiplicative factor of 1.004 was applied to the 2010 CarbonTracker CO_2 fields in order to produce the 2011 $x\text{CH}_4$ product. The standard deviations of the observation error provided with the data do not vary much in time during the study period. Its averaged value is 60 ppb.

It is well known that SCIAMACHY NIR channels suffered from severe radiation damage (Kleipool et al., 2007; Frankenberg et al., 2011), thereby affecting its performance by increasing the measurement noise error (Gloudemans et al., 2008). This becomes worse towards the end of the mission which is the period used in this study.

2.2.2 TANSO

GOSAT is a joint effort from the Japan Aerospace Exploration Agency (JAXA), the National Institute for Environmental Studies (NIES) and the Japanese Ministry of the Environment (MOE) as part of the Global Change Observation Mission (GCOM) program of Japan. GOSAT was launched on 23 January 2009, into a sun synchronous orbit at an altitude of 666 km. This implies that the satellite comes back to the same location with a 3 day period.

GOSAT carries the TANSO observing instrument, which consists of a Fourier Transform Spectrometer (TANSO-FTS) and a Cloud and Aerosol Imager (TANSO-CAI). Two out of the four bands measured by TANSO-FTS are sensitive to CH₄, one in the short wave infrared (SWIR) and one in the thermal infrared (TIR) (for more details see: www.gosat.nies.go.jp/eng/gosat). Each detector has the same circular field of view of 10.5 km when projected on the Earth's surface (at exact nadir), and a swath of 750 km. Compared to SCIAMACHY that had a continuous coverage across the swath (for clear sky condition) and thus 16 observations, TANSO makes only 5 observations across the swath due to the measurement time.

The xCH₄ TANSO product used in this study is also provided by SRON using the proxy setup of the RemoTeC algorithm (Butz et al., 2010), a joint development between SRON and KIT. In this paper, we use version v.1 (from November 2010 to September 2011) and version v.2.0 (from October 2011 onward) of the product. These products also consist of a dry molar fraction column with an a priori profile and its corresponding averaging kernel. The a priori and the averaging kernel have 6 vertical levels for version v.1 and 4 vertical levels for version v.2.0 (Fig. 3). Note that the TANSO averaging kernels have a similar shape as the SCIAMACHY ones as they both measure in the near infrared. That makes TANSO also sensitive to the lower troposphere. The given values of the observation errors are lower than the ones from SCIAMACHY. Their averaged value is 12.6 ppb which means that the assimilation algorithm will give more confidence to the TANSO data than to the SCIAMACHY ones.

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The coverage of both versions of the product is similar over land even when the solar zenith angle limit was changed from 70° to 75° (Fig. 2). As for SCIAMACHY, the coverage of the Northern Hemisphere is larger during the summer reaching 70° N or 75° N (depending on the version of the product), and smaller during the winter, reaching 50° N. Contrary to SCIAMACHY, the coverage of the Southern Hemisphere does not vary much with the season. The measurements are mainly located north of 45° S with few points over Antarctica. Version v.2.0 of the product provides data over the oceans (sunglint instrument pointing) where no data were retrieved in version v.1. These observations over oceans are located in a tropical band that varies with the season, with more measurements in the Northern Hemisphere during JJA and more measurements in the Southern Hemisphere during DJF. These observations allow increasing the number of assimilated data per 12 h assimilation window from about 200 observations (for version v.1) to about 360 observations (for version v.2.0). This nevertheless remains more than 3 times less than the number of assimilated SCIAMACHY observations.

2.2.3 IASI

The first Infrared Atmospheric Sounding Interferometer (IASI) developed by the Centre National d'Etudes Spatiales (CNES) in collaboration with the European Organisation for the Exploitation of Meteorological Satellites (EUMETSAT) was launched in October 2006 onboard the European MetOp-A platform. IASI is a Fourier Transform Spectrometer based on a Michelson Interferometer coupled to an integrated imaging system that measures infrared radiation emitted from the Earth from 645 to 2760 cm⁻¹, with a spectral resolution of 0.5 cm⁻¹ apodized and a spectral sampling of 0.25 cm⁻¹.

MetOp-A flows in a sun synchronous polar orbit at an altitude of 817 km. IASI is an across track scanning system with a scan range of ±48.3° (symmetrically with respect to the nadir direction). A nominal scan line covers 30 scan positions towards the Earth. Each instantaneous field of view has a ground resolution of 12 km at nadir. This provides about 4,000 observations to assimilate per 12 h window.

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concentration from the DM analysis for the 1st November 2010 and used them as initial conditions. The SCIA experiment is then a clone of the DM but at a different resolution. The two other analysis experiments use the same initial condition so we can assess the impact of the change in the observing system. The FREE run also uses this initial condition in order to better evaluate the impact of the assimilation.

The study period runs from the beginning of November 2010 to the end of April 2012 (18 months), when SCIAMACHY stopped producing data (Fig. 1). Occasionally we split the period studied into seasons to interpret the results. Note that the first completed season studied (DJF 2010/2011) starts one month after the beginning of the study period. This first month can thus be regarded as a spin-up period.

2.4 Evaluation data

The non-satellite CH₄ data are mainly measurements of the near surface concentrations. For example, in situ measurements are provided by the National Oceanic and Atmospheric Administration (NOAA) network (www.esrl.noaa.gov/gmd) and the pre-operational Integrated Carbon Observation System (ICOS) network (icos-atc-demo.lsce.ipsl.fr). But as the assimilated data are columns and therefore they do not constrain well the surface level in the analysis, we decided to carry out the validation of the analyses using profiles and column measurements. We chose some campaigns from the HIPPER Pole-to-Pole Observations (HIPPO, Wofsy et al., 2011, hippo.ornl.gov) and the measurements from the Total Carbon Column Observing Network (TCCON, Wunch et al., 2011, tcon-wiki.caltech.edu). The flight data cover 3 months of the studied period with a good vertical resolution while the ground-based data cover the whole period but provide only the vertically integrated information in a few places over the Globe.

During the period under study, four HIPPO campaigns took place between 16 June and 9 September 2011. Three of them have a similar trajectory for the flights over the Pacific ocean (Fig. 4). The second campaign from 28 June to 11 July 2011 has its

flights more towards the West and crossing Australia and Indonesia. In this paper, the 10 s merged methane product was used (Wofsy et al., 2012).

At the time of writing, twenty one stations of the TCCON network have reported data for the period we are interested in. We chose to compute global statistics using all these stations and additionally differentiating them by regions and by period. Most of the used data are localised north of 20° N (about 70 %). The data collected over the tropical region represent 8 % and only 4 % for the period before October 2011. Moreover, eight stations were chosen to present time series (Fig. 4). Some stations were retained because they provided the most continuous data set (e.g. Park Falls). We also kept the stations located in the tropical area in order to assess the impact of the assimilation of the IASI data. The two stations located at the two highest latitudes (north and south) were also selected (e.g. Lauder and Sodankyla).

3 Results

In the following, we present the results of the FREE experiment and the three assimilation experiments by comparing the latter to the free run.

3.1 Free run

Unlike for a CTM, the FREE experiment consists here of forecasting both the CH₄ molar fraction and the meteorological parameters. The meteorological parameters are free to evolve without any constraint during the forecast. In order to have more accurate meteorological parameters, we replaced them each 12 h by the ones from one of our assimilation experiment (all our analyses presented in this paper having the same meteorological parameters). These analysed meteorological parameters result from the assimilation of all the operational meteorological data in IFS. This procedure makes the FREE experiment have a slightly different meteorological forcing than the other experiments.

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Methane is free to evolve without any constraint by observations in the FREE experiment. Its evolution is subject to the transport, the fluxes and the chemical loss rates. The fluxes are the largest source of uncertainty for CH₄ in this experiment and they are also the main driver of its evolution. Even if there is no constraint on the fluxes or on the CH₄ molar fraction, the forecast CH₄ is realistic enough to provide us the reference to assess the effect of the assimilation of CH₄ data. This simulation represents the main features of atmospheric CH₄ like the north–south gradient as well as the seasonal variability of the Northern Hemisphere (Fig. 5).

One can notice an increase in the global trend of the methane total column. This is due to the modelled CH₄ growth, i.e. a net positive budget of the sources and the sinks.

3.2 SCIAMACHY analysis

Due to the fact that SCIAMACHY measures reflected solar light, SCIAMACHY can only provide daytime measurements. This leads to gaps into the global coverage during local winter periods (Fig. 6a). To assess how the analysis deals with these gaps, we computed the seasonal averages of the analysis increment in terms of total column (Fig. 7a). An averaged increment is representative of a systematic mismatch between the background state (computed by a 12 h forecast from the previous analysis) and the observations. We do not discuss here the values of the averaged increment as the biases are not the focus of this paper. We use this diagnostic to present where the observations add information through the analysis.

The structure of the averaged increment shows that the analysis of SCIAMACHY brings information mainly over land in the tropical region and at mid-latitudes and not over the ocean. Depending on the season, the analysis brings information at high latitudes (Northern Hemisphere in JJA and Southern Hemisphere in DJF) over ocean and land. The direct impact of the assimilation of the SCIAMACHY product should therefore be found over the tropics to mid-latitudes and at higher latitudes of each hemisphere during summer-time. When the CH₄ total columns from the SCIA experiment are compared to the ones from the FREE experiment, the main difference is at high latitudes

of the Northern Hemisphere in June (Fig. 5b). Total columns from the SCIA experiment are locally more than 50 ppb lower than those from the FREE experiment. This difference corresponds to less CH₄ molar fraction from the surface to the lower stratosphere, up to 50 hPa in the analysis (Fig. 8, left panels). The difference is largest in the troposphere and particularly at levels close to the surface.

At high latitudes in the Southern Hemisphere, the main differences are observed during DJF which corresponds to the local summer. There, the SCIA experiment has still less CH₄ from the surface up to 100 or 50 hPa (depending on the year). The largest differences of more than 30 ppb are found from the near surface to the upper troposphere. This corresponds to differences of about 20 to 30 ppb in terms of averaged total columns. One can notice that during the winter-time of each hemisphere, there is an indirect impact of SCIAMACHY as xCH₄ remains lower in the SCIA experiment compared to the FREE experiment. This is likely due to the transport that spreads the observation information in space and time or due to the model adjustment to its equilibrium state.

Over the tropical region, the analysis also has lower methane concentrations than the FREE experiment. The differences are higher in MAM and JJA 2011 with values between 15 and 25 ppb, and in MAM 2012 with values between 30 and 40 ppb. The result of these seasonal differences as well as the differences at highest latitudes is a modification of the north–south gradient and the seasonal cycle in the SCIA experiment compared to the FREE experiment. In the SCIA experiment the gradient is steeper in DJF and flatter in JJA, and the seasonal cycle is more pronounced.

3.3 TANSO analysis

The TANSO data are available during most of the period under study, apart for a few days in August 2011 (Fig. 6b). The Southern Hemisphere is less well sampled as there is little or no coverage of the oceans and the coverage mostly stops for latitudes south of 45° S. SCIAMACHY has thus an added value in this region south of 45° S as its data impact the analysis during the local-summer period as seen previously (Sect. 3.2). The

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averaged increments of the TANSO experiment show however that the TANSO data have an influence over Antarctica similar to the SCIAMACHY data (but to a lesser extent) in DJF 2011/2012 (Fig. 7). They also have an influence over oceans at southern mid-latitude while the SCIAMACHY data have an influence further on south. In JJA, where both datasets are available, their region of influence is similar. At mid and high latitudes north, SCIAMACHY data have additionally an influence over oceans. In summary, the averaged increments show that even if the TANSO data are sparser than the SCIAMACHY data, their region of influence is similar over the tropics and mid-latitudes.

The assimilation of the TANSO data has less impact on the analysis than the assimilation of the SCIAMACHY data when compared to the FREE experiment (Fig. 5c). The difference between the CH₄ column from the TANSO experiment and from the FREE experiment is mostly less than 10 ppb but here the analysis has higher CH₄ values than the FREE experiment. This is the main characteristic of this experiment compared to the SCIA one. The exception is for the summer period at northern high latitudes. As for the SCIA analysis, the TANSO analysis reduces the lower atmospheric CH₄ compared to the FREE experiment. But where the reduction exceeds 50 ppb for the SCIA experiment, it is here less than 40 ppb. The TANSO measurements are limited to 82° N, but due to the transport and the background error correlation, the information is spread to higher latitudes. This compensates the lack of TANSO data at the high latitudes.

The TANSO data influence the vertical distribution of the analysed CH₄ compared to the vertical distribution from the FREE experiment mainly over the northern extra tropical region from MAM to SON (Fig. 8, central panels) as for the total columns. The differences reach their maximum in JJA with a CH₄ molar fraction lower for the analysis (compared to FREE). The maximum value of the difference is more than 30 ppb and it is found from the surface to 500 hPa. The TANSO data thus allow constraining the whole troposphere, as is the case for SCIAMACHY. Note that the difference between the TANSO experiment and the FREE experiment in the stratosphere is similar to the one between the SCIA experiment and FREE. This supports the previous statement

that the difference in the stratosphere between the assimilation experiments and the free run experiment comes from slight differences in the meteorological forcing.

3.4 TANSO and IASI analysis

When the IASI data are added to the TANSO data in the analysis, their effect is evaluated by comparing the resulting analysis with the TANSO analysis (Fig. 5d). The comparison emphasizes the impact of the change in the version of the assimilated TANSO data in October 2011. The IASI+TANSO experiment has between 5 and 15 ppb less methane in total column after October 2011. Before this date, the difference is mainly between -5 and 5 ppb, except for DJF 2010/2011 when the total column from the IASI+TANSO experiment are 15 ppb higher. The difference between the two experiments is mainly located in the tropical and mid-latitudes regions south of 30° N and north of 30° S where the IASI data are provided. The transport and the background error correlation however spreads the information at highest latitudes but there the difference is never much more than 2 ppb.

The weighting function of the IASI product peaks at about 250 hPa. It reaches half of its maximum respectively at 100 hPa and 500 hPa. One can expect to see the influence of the assimilation of the IASI data mainly in this vertical region between 100 and 500 hPa. But looking at the differences between the zonal and seasonal averages from the IASI+TANSO experiment and from the TANSO experiment, the influence is observable from the surface up to 10 hPa (Fig. 8, right panels). This could be explained by the vertical transport as well as by the vertical correlation in the background error statistics or the interaction with the TANSO data.

From December 2010 to April 2011, the IASI data constrain the analysis so that it has higher values of methane concentration in the middle troposphere over the tropics compared to the TANSO analysis. For the extra-tropical regions, the impact is more dependent on the season. The IASI analysis tends nevertheless to decrease CH₄ in the middle troposphere and to increase it in the lower stratosphere. For the last two seasons (DJF 2011/2012 and MAM 2012) the shape of the difference between the two

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experiments (IASI+TANSO and TANSO) differs substantially from the previous seasons. The previously described decrease in terms of $x\text{CH}_4$ during these two seasons is associated with a decrease of CH_4 by more than 15 ppb from the surface to 100 hPa in the tropics and at the same time an increase (less than 10 ppb) above 100 hPa. Due to the transport, these differences spread to the extra-tropical regions.

3.5 Comparison with independent measurements

The three CH_4 analyses showed their own features in term of CH_4 spatial and temporal distribution when compared to the simulation without data assimilation. To further document the features of each analysis, they are compared to evaluation data, i.e. data that have not been directly included in the assimilation process. These evaluation data are not always independent as for example satellite products are bias-corrected using the measurements from the TCCON network.

3.5.1 Comparison with HIPPO

For each CH_4 concentration measured during the campaigns, we computed the model equivalent for each of our four experiments. Top panel of Fig. 9 presents the measurements for the four campaigns as a function of latitude and altitude (in pressure units). And for each experiment, we present the same figure but for the difference between the model and the measurement (Fig. 9 lines 2 to 5). We removed for each experiment the global bias between the model and the measurements in order to better visualise the difference between the experiments. The biases are computed for the four campaigns together and are available in Table 1.

From the four campaigns, we can clearly identify in the HIPPO data the north–south gradient in the CH_4 distribution with lower values in the Southern Hemisphere (Fig. 9 top panel). We can also clearly assess the vertical gradient when the flight crosses the tropopause at around 200 hPa at high latitudes.

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The FREE experiment represents the north–south gradient reasonably well for the two last campaigns (Fig. 9 line 2), despite a global underestimation of about 4 ppb in the CH₄ tropospheric content (Table 1). But it fails to represent correctly the gradient for the two first campaigns with an underestimation over the Southern Hemisphere and an overestimation over the Northern Hemisphere. On average for the campaigns, the underestimation over the Southern Hemisphere is around 7 ppb (as well as for the tropics) and the overestimation over the Northern Hemisphere is also around 7 ppb. This leads to an overall underestimation of less than 2 ppb.

As shown in the previous section, the SCIA analysis reduces the north–south gradient during the summer. This leads to an improvement in the inter-hemispheric gradient when compared to the HIPPO data (Fig. 9 line 3). This experiment has a better gradient than the one from the FREE experiment. But overall the analysis underestimates CH₄ by about 30 ppb on average. This suggests an underlying bias of the SCIAMACHY product.

We already mentioned the change in the north–south gradient in the TANSO experiment due to a reduction of the tropospheric CH₄ in the Northern Hemisphere (Sect. 3.3). When compared to the HIPPO data this reduction appears to be slightly too large (Fig. 9 line 4). The Northern Hemisphere overestimation in the FREE experiment (7 ppb) is replaced by a larger underestimation of 22 ppb in the analysis (Table 1). For the Southern Hemisphere, the impact of the assimilation of the TANSO data is mainly positive (from 7 ppb for FREE to 3 ppb for the analysis) even at high latitudes where there is no data to constrain the model. This suggests a latitude dependent bias in the version v.1 of the TANSO product. The Northern Hemisphere bias increases however at high latitudes during the last campaign with a difference larger than 40 ppb when compared to the HIPPO data. Note that this large underestimation is also present in the SCIA analysis.

When comparing the difference between the IASI+TANSO analysis and the HIPPO data, with the difference between the TANSO analysis and the HIPPO data, we see the impact of the IASI data on the analysis. Globally the IASI+TANSO experiment is slightly

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less biased than the the TANSO experiment (Table 1). The improvement comes from the tropics where the bias is reduced by about 4 ppb and from the Southern Hemisphere where it is reduced by about 2 ppb. In contrast, the bias increased by about 2 ppb in the Northern Hemisphere. In terms of vertical distribution, the difference between the experiments is located mainly above 700 hPa (Fig. 9, last line). The largest effect of the assimilation of the IASI data is found for the first campaign during which the inter-hemispheric gradient is less well represented in the IASI+TANSO experiment.

In order to have another picture of how well the experiments are able to represent the CH₄ features as measured during the HIPPO campaigns, we computed the correlation between the measurements and the model equivalent (Table 2). The FREE experiment presents already a strong correlation of 0.95 which means that the model is able to well represent the spatial distribution of the tropospheric CH₄. The SCIA experiment has a similar correlation but with a different regional distribution compared to the FREE experiment. The correlation is higher over the Northern Hemisphere and the tropics but lower in the Southern Hemisphere. This shows that the assimilation system correctly uses the information content of the SCIAMACHY data. In contrast, the system has more difficulties with the TANSO data especially in the Southern Hemisphere where only few data are available to the assimilation. There the correlation drops from 0.75 (from the FREE experiment) to 0.68 (for the TANSO experiment). This means that the propagation of the information to the Southern Hemisphere has to be improved and the output of the analysis in this region should be used with caution. Even if the assimilation of the IASI data allows reducing the bias compared to the HIPPO measurements, it degrades the correlation in the tropics with a value of 0.57 for the IASI+TANSO experiment when the FREE experiment has a correlation of 0.83 there. This suggests an issue with the IASI data or that the assimilation system does not make a proper use of the IASI data, at least when they are combined with the version v.1 of the TANSO product. We saw in Sect. 3.4 that the impact of the IASI data in the vertical distribution of CH₄ changed when the data are combined with version v.1 or version v.2.0 of the TANSO data (left column of Fig. 8). Unfortunately we do not have other profile mea-

surements for the second period to document if the IASI+TANSO experiment presents better profiles with the version v.2.0 of the TANSO data.

3.5.2 Comparison with TCCON

For each TCCON measurements we computed the model equivalent using the TCCON a priori information and averaging kernel. To summarise the behaviour of each experiment compared to the TCCON measurements, we then computed the monthly averaged differences between the model and the measurements.

Period before October 2011

The initial condition of all the experiments comes from the methane analysis of the DM which is known to have a negative bias of about 20 ppb in terms of total column when compared to independent measurements (Massart et al., 2013). Therefore, at the beginning of the period, the FREE experiment underestimates $x\text{CH}_4$ when compared to the measurements of the selected TCCON stations (black lines of Fig. 10). The underestimation varies from 10 to 30 ppb depending on the station without a clear difference between the stations in the southern or Northern Hemisphere. We previously described an increase over time of $x\text{CH}_4$ in this experiment (Sect. 3.1 and Fig. 5a). This increase allows reducing the bias against the TCCON measurements after the first few months and leads to a global bias close to zero on average between November 2010 and October 2011 (Table 3), that is consistent with the global bias found compared to the HIPPO data. But this increase is too large in the Northern Hemisphere and the FREE experiment starts to overestimate $x\text{CH}_4$ especially in JJA 2011 (e.g. black lines of Fig. 10 for Parkfalls). This leads the FREE experiment to have a bias of 2 ppb in this region on average before October 2011 (Table 3). Its bias is opposite over the tropics and over the Southern Hemisphere with a value of respectively -4 and -9 ppb. The FREE experiment therefore fails on average at correctly representing the north-south gradient.

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The assimilation of the TANSO data partially helps in reducing the biases with respect to the TCCON measurements on average with strong differences between the two versions of the data. In the Southern Hemisphere during the period from November 2010 to September 2011 the assimilation of version v.1 of the TANSO data helps in reducing the bias with respect to the TCCON measurements from -9 ppb for the FREE experiment to -5 ppb for the TANSO experiment (Table 3). In the meantime the assimilation of the SCIAMACHY data increases the bias up to -27 ppb. This means that the version v.1 of the TANSO product provides more accurate information than the SCIAMACHY product in the Southern Hemisphere. This corroborates the conclusions drawn from the comparison with the HIPPO data. In the Northern Hemisphere, during the same period, both SCIA and TANSO analyses underestimates the methane column compared to the TCCON data by respectively 24 and 13 ppb. The two experiments are nevertheless similar from July to September 2011 looking at specific TCCON stations (four last panels of Fig. 10, cyan – TANSO – and green – SCIA – lines). This indicates again a latitude-dependent bias in the TANSO experiment and not in the SCIA one.

The effect of the assimilation of IASI data combined with TANSO data was found to be mainly located in the tropical band where the IASI data are available (Fig. 5). When the IASI+TANSO analysis (red lines of Fig. 10) is compared to the measurements from stations situated north of 20° N (Park Falls, Garmisch, Karlsruhe and Sodankyla) there is hardly any significant difference with the TANSO analysis. On average over all the stations of the Northern Hemisphere, the bias is slightly increased by one ppb with the assimilation of the IASI data (Table 3). In contrast, for the stations situated south of 20° S (Lauder and Wollongong), IASI has a positive impact as the bias of the IASI+TANSO experiment (-2 ppb) is lower than for the TANSO experiment (-5 ppb) and FREE experiment (-9 ppb). In the tropical region, the statistics are driven by the comparison at the Darwin station. There the IASI+TANSO experiment has a larger bias than the TANSO experiment during DJF 2010/2011 (red and green lines, Darwin panel, Fig. 10). On average in this region, the effect of adding the IASI data in the assimilation system is an increase of the bias from -1 ppb to 9 ppb. This is related to the previously

Whole period

The TANSO experiment and accordingly the IASI+TANSO experiment showed a very different behaviour before and after October 2011. Their statistics over the whole period are therefore not relevant. For the FREE experiment, its behaviour was also different during the two periods. But it shows a constant characteristic which is a highest bias over the Northern Hemisphere (and over the tropics) than over the Southern Hemisphere, with a difference of more than 8 ppb between them on average for the whole period. The statistics of the SCIA experiment are consistent over time with global bias of -23 ppb which is consistent but lower compared to the value of -37 ppb found by Houweling et al. (2013) for 2009 when they directly compare the SCIAMACHY data with the TCCON ones.

4 Conclusions

After the loss of Envisat in April 2012, the MACC-II delayed mode system for methane had to rely on the assimilation of the TANSO and the IASI products. In order to assess the impact of this change in the observing system, we have documented how the assimilation of the products of each of these instruments constrains the model. First, the model without any assimilation of methane data showed a good behaviour even if the initial condition was negatively biased. It nevertheless overestimates the inter hemispheric gradient in June and July 2011 when compared to the HIPPO aircraft measurements. This was due to an underestimation of the methane concentration in the Southern Hemisphere and an overestimation in the Northern Hemisphere, the difference between them being 8 ppb. This was confirmed when the experiment was compared against measurements from the TCCON network. The CH_4 sources and sinks are climatological in the model and the difference between them produces a positive global budget. As a consequence, the free run model will have the same positive

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methane growth each year and therefore it will deviate from the observed growth. Using data assimilation is thus needed in order to properly monitor atmospheric CH₄.

SCIAMACHY provided dense and global measurements but only during daylight hours. This limited the amount of data to assimilate, especially during the winter period of each hemisphere. When the data were available, the largest impact of the assimilation of the SCIAMACHY data was found during MAM and JJA at high latitudes in the Northern Hemisphere. There the assimilation decreased the methane concentration from the surface up to 50 hPa. This allowed improving the north–south gradient in the analysis. But on the other hand, this also introduced a negative bias in the analysis estimated to 30 ppb for the tropospheric CH₄ concentration when compared to HIPPO data and to 23 ppb for the column when compared to TCCON measurements. This bias may result from the degraded performance of the SCIAMACHY during the studied period. In the context of the DM system, the bias of the assimilated product could not be computed as there are no independent data in near-real-time to anchor a bias correction scheme. For consistency purposes, SCIAMACHY data have not been bias corrected in this study. However, if this version of the SCIAMACHY product had to be assimilated again in the future for a reanalysis purpose, a bias correction would therefore have to be envisaged.

TANSO also provides measurements only during daylight hours. But compared to SCIAMACHY, the data are much less dense (about 3 times less) and cover only land in version v.1 of the used product and partially the ocean in version v.2.0. Over land the density of the TANSO data is nevertheless sufficient to produce a similar constraint on the analysis as with the SCIAMACHY data thanks the background error correlations. Over tropical oceans, the density of SCIAMACHY data was too low to affect the analysis and to have any appreciable difference with TANSO. The change of version for the TANSO product had a significant impact on the analysis. When the analysis of the first version has a global negative bias of –11 ppb on average compared to the measurements from the TCCON network, the analysis of the second version has a positive bias of 10 ppb. The bias of the first analysis is confirmed by the comparison

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evaluate the change in the profile of the analysis. In terms of total column the impact of the IASI data was positive reducing by about 5 ppb the extra-tropical bias when compared to data from the TCCON network.

The combination of the IASI data and the version v.2.0 of the TANSO data has been shown to be a good candidate to continue the monitoring of the tropospheric methane in the MACC-II system. Despite residual biases present in the products, the main concern is the deficiency of data to constrain the system south of 45° S and north of 45° N in winter or 80° N (in summer), this last region being also not observed by SCIAMACHY. Extending the coverage of the IASI CH₄ data to the extra-tropical regions could help by increasing the number of data in these regions.

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Table 1. Averaged difference (in ppb) between the free run or the analyses and the HIPPO data. The average is computed for the four campaigns and for data below 300 hPa. It is also split into regions: Northern Hemisphere above 20° N (NH); tropics between 20° S and 20° N (Trop); Southern Hemisphere below 20° S (SH).

	NH	Trop	SH	Global
FREE	2.68	-6.43	-12.55	-4.98
SCIA	-36.38	-31.41	-34.45	-34.64
TANSO	-27.55	-13.98	-8.80	-17.65
TANSO+IASI	-29.68	-12.56	-7.35	-17.69

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Table 2. Same as Table 1 but for the correlation.

	NH	Trop	SH	Global
FREE	0.781	0.831	0.752	0.951
SCIA	0.792	0.859	0.726	0.952
TANSO	0.791	0.816	0.685	0.948
TANSO+IASI	0.792	0.572	0.649	0.928

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Table 3. Averaged difference (in ppb) between the free run or the analyses and the TCCON data. The average is computed for all available TCCON stations and split into regions as for Table 1 and into periods: from November 2010 to April 2012 (top), before October 2011 (middle) and after October 2011 (bottom).

	NH	Trop	SH	Global
FREE	5.67	5.03	-2.81	3.72
SCIA	-22.33	-19.28	-25.75	-22.86
TANSO	-5.23	8.35	1.57	-2.63
TANSO+IASI	-7.74	11.74	1.14	-4.17
Before Oct. 2011				
FREE	1.94	-4.37	-9.10	-0.52
SCIA	-23.58	-21.95	-27.02	-24.18
TANSO v.1	-13.07	-1.19	-4.84	-10.92
TANSO v.1+IASI	-14.01	9.03	-2.42	-10.69
After Oct. 2011				
FREE	12.75	9.82	4.28	10.12
SCIA	-19.89	-17.92	-24.33	-20.82
TANSO v.2.0	9.65	13.19	8.79	9.89
TANSO v.2.0+IASI	4.55	13.12	5.15	5.87

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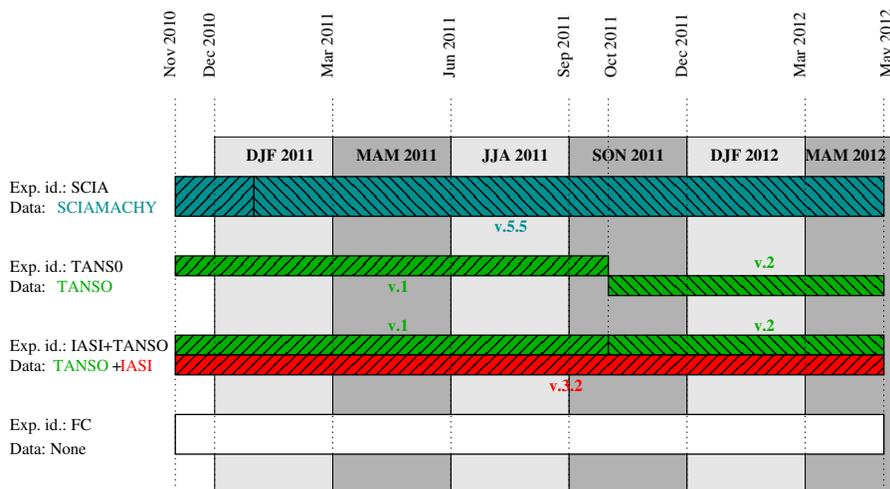


Fig. 1. Configuration of the experiments. Each line represents an experiment and the box represents the time-line when the experiment was run. The name of the experiment is given by “Exp. Id.”. The assimilated data is given by “Data”. The text near the boxes is the version of the data that were assimilated.

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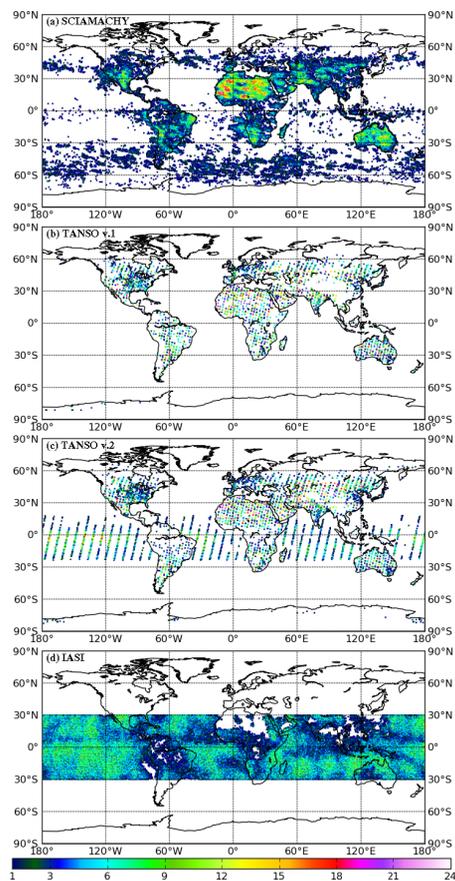


Fig. 2. Number of assimilated observations in October 2011, for **(a)** SCIAMACHY, **(b)** TANSO v.1 and **(c)** TANSO v.2.0, and **(d)** IASI. The observations are gridded on a $0.7^\circ \times 0.7^\circ$ grid.

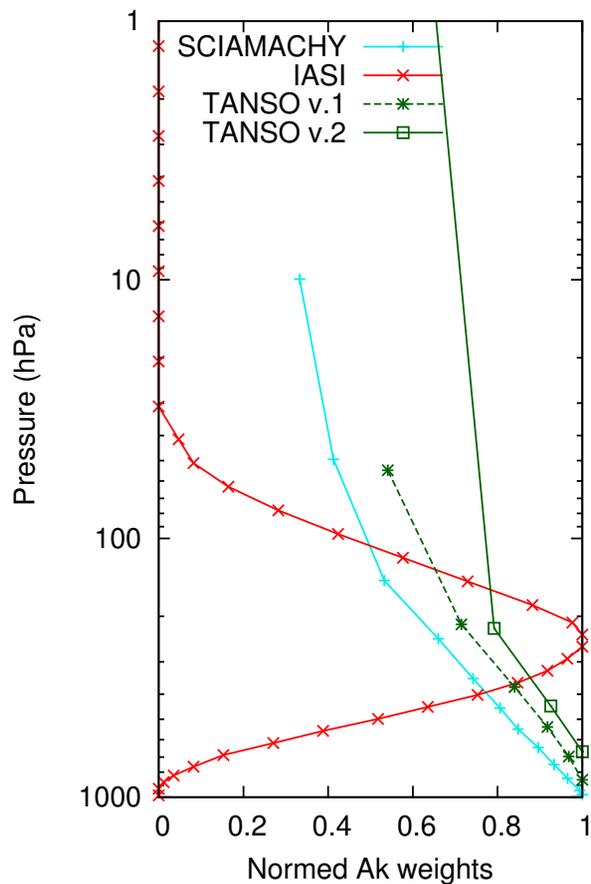


Fig. 3. Example of averaging kernels (cyan: SCIAMACHY, dotted green: TANSO v.1 and full green: TANSO v.2.0) and weighting function normalised by it maximum value (red: IASI) of the CH_4 products from the different used instruments.

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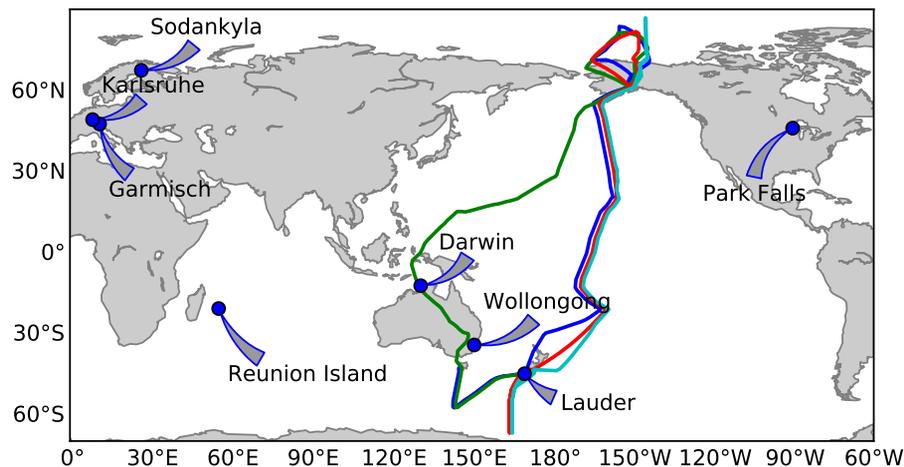


Fig. 4. Location of some particular TCCON stations used in this paper (blue points) and trajectories of the 2011 HIPPO campaigns. The campaign periods are respectively: 16 June–29 June (blue), 28 June–11 July (green), 19 August–30 August (red), 29 August–09 September (cyan). Note that for the plot, some trajectories are shifted one or two degrees east or west in order to avoid superposition between them.

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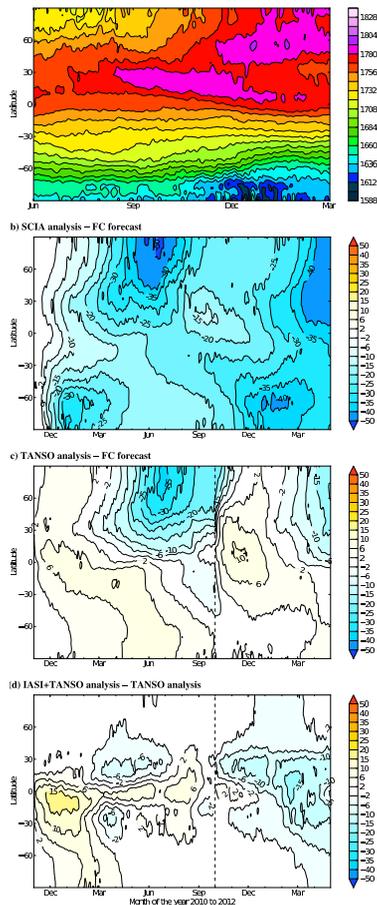


Fig. 5. Time series of xCH₄ zonal mean (in ppb). From top to bottom: FREE free run, SCIA analysis – FREE, TANSO analysis – FREE, IASI+TANSO analysis – FREE. The vertical dotted line corresponds to the date when version v.2.0 of the data replace version v.1.

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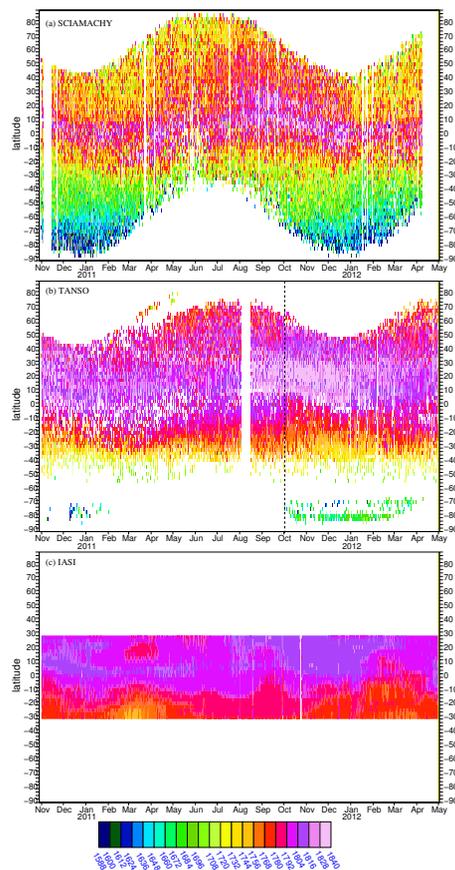


Fig. 6. Time series of xCH₄ zonal mean (in ppb) from SCIAMACHY, TANSO and IASI products (from top to bottom).

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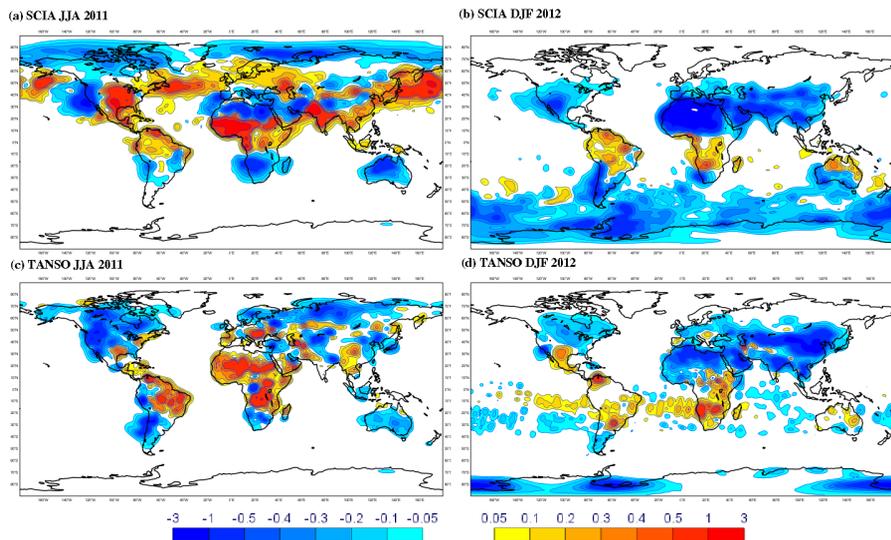


Fig. 7. Seasonal average of the increment in terms of CH₄ total column, for JJA 2011 (left) and DJF 2011/2012 (right) in ppb per 12 h. From top to bottom: SCIA analysis and TANSO analysis.

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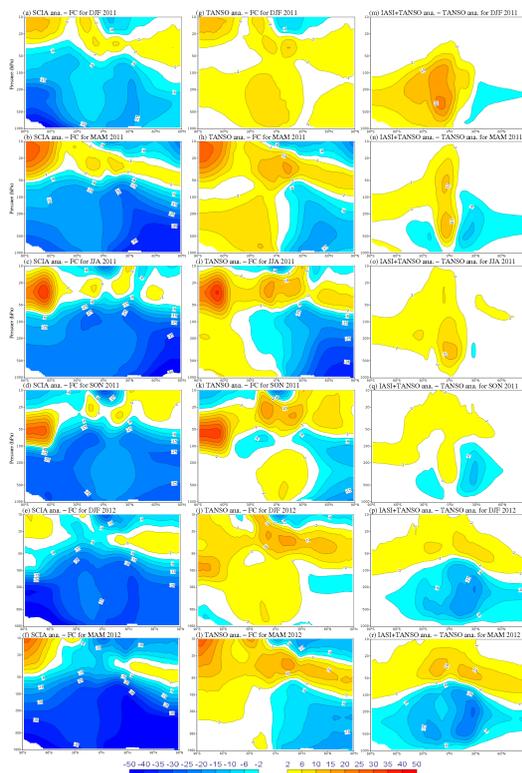


Fig. 8. Seasonal and zonal mean difference between the CH₄ molar fraction (in ppb): from the SCIA experiment and the FREE free run (left); from the TANSO experiment and the FREE free run (middle); from the IASI+TANSO experiment and the TANSO experiment (right). The seasonal means are from top to bottom: DJF 2010/2011, MAM 2011, JJA 2011 and SON 2011, DJF 2011/2012 and MAM 2012.

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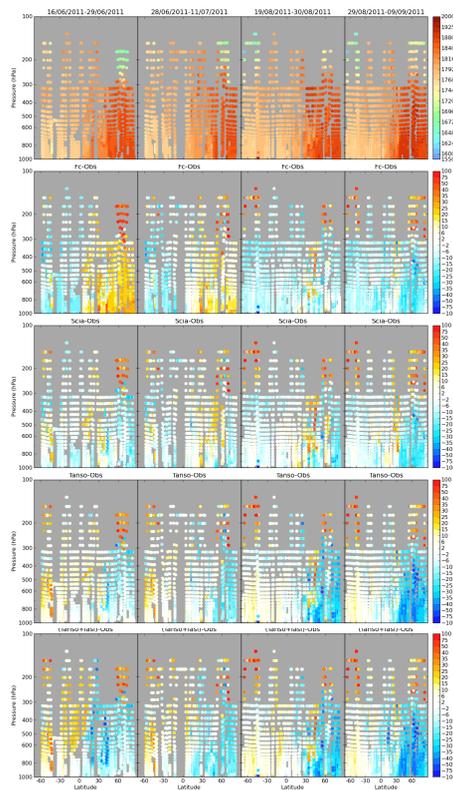


Fig. 9. Comparison between the analyses and the HIPPO data. From left to right: latitude vs. pressure plots for the four flights. From top to bottom: HIPPO observations, observations minus SCIA analysis, observations minus TANSO (v.1) analysis and observations minus IASI analysis. A global bias has been removed from the free run and from each analysis (see Table 1).

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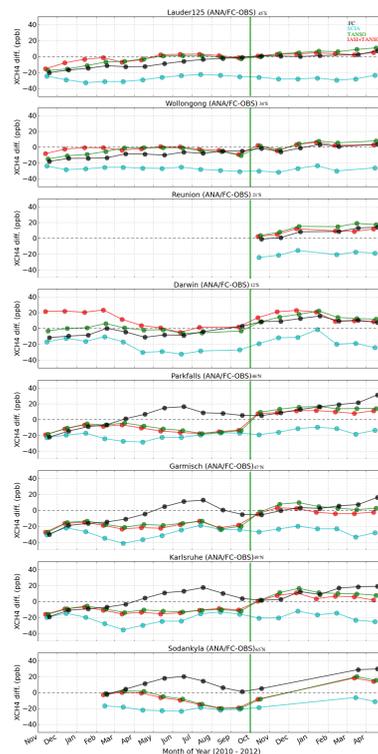


Fig. 10. Time series of the monthly differences (in ppb) between xCH₄ measurements from the TCCON network and xCH₄ from the free experiment (black) and from the analyses (cyan: SCIA, green: TANSO, red: IASI + TANSO). The stations are ordered from top to bottom by their latitude, from south to north: Lauder, Wollongong, Reunion, Darwin, Park Falls, Garmisch, Karlsruhe, Sodankyla.

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