

Inverse modeling of CH₄ emissions for 2010–2011 using different satellite retrieval products from GOSAT and SCIAMACHY

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Abstract. Beginning in 2009 new spaceborne observations of dry-air column-averaged mole fractions of atmospheric methane (XCH₄) became available from the Thermal And Near infrared Sensor for carbon Observations–Fourier Transform Spectrometer (TANSO-FTS) instrument onboard the Greenhouse Gases Observing SATellite (GOSAT). Until April 2012 concurrent methane retrievals were provided by the SCanning Imaging Absorption spectroMeter for Atmospheric Cartography (SCIAMACHY) instrument onboard ENVISAT. The GOSAT and SCIAMACHY XCH₄ retrievals can be compared during the period of overlap. We estimate monthly average methane emissions between January 2010 and December 2011, using the TM5-4DVAR inverse modeling system. In addition to satellite data, high-accuracy measurements from the Cooperative Air Sampling Network of the National Oceanic and Atmospheric Administration Earth System Research Laboratory (NOAA ESRL) are used, providing strong constraints on the remote surface atmosphere. We discuss five inversion scenarios that make use of different GOSAT and SCIAMACHY XCH₄ retrieval products, including two sets of GOSAT proxy retrievals processed independently by the Netherlands Institute for Space Research (SRON)/Karlsruhe Institute of Technology (KIT), and the University of Leicester (UL), and the RemoTeC “Full-Physics” (FP) XCH₄ retrievals available from SRON/KIT.

The GOSAT-based inversions show significant reductions in the root-mean-square (RMS) difference between retrieved and modeled XCH₄, and require much smaller bias corrections compared to the inversion using SCIAMACHY retrievals, reflecting the higher precision and relative accuracy of the GOSAT XCH₄. Despite the large differences between the GOSAT and SCIAMACHY retrievals, two-year average emission maps show a good overall agreement among all satellite-based inversions, with consistent flux adjustment patterns, particularly across equatorial Africa and North America. Over North America, the satellite inversions result in a significant redistribution of methane emissions from the northeastern to south-central USA. This result is consistent with recent independent studies suggesting a systematic underestimation of methane emissions from North American fossil fuel sources in bottom-up inventories, probably related in large part to natural gas production facilities. Furthermore, all four satellite inversions yield lower methane fluxes across the Congo basin compared to the NOAA-only scenario, but higher emissions across tropical eastern Africa. The GOSAT and SCIAMACHY inversions show similar performance when validated against independent shipboard and aircraft observations, and XCH₄ retrievals available from the Total Carbon Column Observing Network (TCCON).

1 Introduction

Atmospheric methane (CH₄) is the second-most important anthropogenic greenhouse gas (GHG) – after carbon dioxide – in terms of net radiative forcing (RF). Emissions of CH₄ have caused an RF of 0.97 W m⁻² (Stocker et al., 2013, p. 13), about twice the concentration-based estimate (0.48 W m⁻²). After a period of stabilization from 1999 to 2006 (Dlugokencky et al., 2003; Simpson et al., 2006), methane concentrations in the atmosphere have started to rise again (Dlugokencky et al., 2009; Rigby et al., 2008; Nisbet et al., 2014), and are currently estimated to be 160% higher than pre-industrial (1750) values (WMO, 2013). Previous research has identified the main sources and sinks of atmospheric methane; however, there remain considerable uncertainties regarding their relative importance (e.g., Kirschke et al., 2013).

Since large-scale regional or global methane fluxes cannot be directly measured, attempts at estimating these quantities have traditionally relied on two complementary techniques: “bottom-up” emission inventories, and inverse modeling (“top-down”). Bayesian inverse modeling (Tarantola, 2004) of CH₄ emissions operates under a well-defined mathematical framework that combines a priori information on methane emissions, atmospheric observations, and an atmospheric chemistry and transport model (CTM), to yield a statistical best estimate of methane emissions and concentrations over the time period of interest. The quality of the estimates obtained through inverse modeling depends in large part on the quality of the observation data available for the spatial and temporal domains of interest, and on the quality of the CTM.

Surface measurements of CH₄ concentrations are available from global networks such as the Cooperative Air Sampling Network of the National Oceanic and Atmospheric Administration Earth System Research Laboratory (NOAA ESRL) (Dlugokencky et al., 1994, 2009, 2013). However, surface observations provide only sparse global coverage, with the exception of certain regions, mainly Europe and North America, where regional monitoring stations, including tall towers and aircraft profiles, have been set up in recent years (e.g., Vermeulen et al., 2007)). Surface measurements provide effective constraints on regional emissions (Bergamaschi et al., 2010; Kort et al., 2008; Miller et al., 2013); however, they are not available in many important emission regions, such as the tropics. Inversions based on global background sites have provided a good picture of global and continental methane emissions, their trends, and inter-annual variability (Bergamaschi et al., 2013a; Houweling et al., 1999; Bousquet et al., 2006; Hein et al., 1997; Mikaloff Fletcher et al., 2004a,b). Smaller-scale regional patterns, however, remain in large part determined by the prior emission inventories due to lack of observations.

Since 2002 satellite retrievals of total-column methane mixing ratios have been available from the SCanning

Imaging Absorption spectroMeter for Atmospheric CHartography (SCIAMACHY) instrument onboard ENVISAT (Frankenberg et al., 2005, 2006, 2008, 2011; Buchwitz et al., 2005; Schneising et al., 2012). The SCIAMACHY data were the first space-borne XCH₄ retrievals sensitive to the atmospheric boundary layer. This new dataset, along with an extension in data coverage to previously observation-poor areas, such as the tropics, led to the first global and regional inversions of methane fluxes and XCH₄ (Bergamaschi et al., 2007, 2009; Frankenberg et al., 2008; Meirink et al., 2008a). Due to the relatively long operational lifetime of SCIAMACHY (almost one decade), the XCH₄ retrievals from this instrument were useful for analysing the inter-annual methane variability (IAV) during this period (Bergamaschi et al., 2013a). However, the impact of the serious detector pixel degradation, which occurred at the end of 2005, remains difficult to evaluate, despite overall good consistency of the SCIAMACHY time series with surface observations (Frankenberg et al., 2011).

Since 2009, XCH₄ retrievals have also become available from the Greenhouse Gases Observing SATellite (GOSAT) TANSO-FTS instrument (Parker et al., 2011; Yoshida et al., 2011; Butz et al., 2011). Given the limited lifetime of satellite instruments (the communication link to ENVISAT was lost in April 2012, while the GOSAT mission plans extend only until 2014), inverse modeling comparison studies using different satellite retrievals are of great importance for understanding the difference between products. Such analyses are a crucial step when using satellite data to analyse IAV and trends. Within the European project MACC-II (“Monitoring Atmospheric Composition and Climate – Interim Implementation”) pre-operational “delayed-mode” CH₄ flux inversions are performed, which are updated every six months (Bergamaschi et al., 2013b). Beginning in 2012 the assimilated satellite data set changed from SCIAMACHY IMAPv5.5 to GOSAT RemoTeC v2.0 (Bergamaschi et al., 2013b). Furthermore, alternative XCH₄ products from GOSAT and SCIAMACHY have been developed within the European Space Agency GHG Climate Change Initiative (ESA-GHG CCI) project (Buchwitz et al., 2013).

This study will present a detailed comparison of global CH₄ flux inversions constrained by different GOSAT and SCIAMACHY retrieval products and surface measurements, covering the two-year period between January 2010 and December 2011. The availability of multiple satellite retrieval products covering the same time interval allows for a detailed comparison of their consistency and added value in inverse modeling, which is the main objective of this paper. Three recent inverse modeling studies (Fraser et al., 2013; Monteil et al., 2013; Cressot et al., 2014) have made use of SCIAMACHY and GOSAT XCH₄ to estimate global CH₄ fluxes and concentrations. Our approach differs significantly from that of these studies. Herein we examine an extended time period, use a different inversion set-up, and employ several distinct (optimized) bias correction strategies for the SCIA-

MACHY and GOSAT retrievals. Another novel element of this paper is the comparison of two different satellite proxy retrievals: the GOSAT RemoTeC dataset (Schepers et al., 2012) from SRON/KIT, and the OCP R GOSAT retrievals from the University of Leicester (Parker et al., 2011). We also assimilate the “Full-Physics” (FP) GOSAT retrievals from SRON/KIT, which do not require the use of modeled CO₂ fields. Furthermore, we invert the SCIAMACHY IMAPv5.5 retrievals as used in the MACC reanalysis (Bergamaschi et al., 2013a). In addition to the GOSAT and SCIAMACHY satellite retrievals, all inversions are constrained by high-accuracy CH₄ surface data from the NOAA/ESRL Cooperative Air Sampling Network. We also present a detailed validation of the inversion results against independent NOAA ship and aircraft profile samples, the aircraft transects from HIPPO – the High-performance Instrumented Airborne Platform for Environmental Research (HIAPER) Pole-to-Pole observation campaigns from 2010 and 2011, and XCH₄ data from the Total Carbon Column Observation Network (TCCON) Fourier Transform Spectrometer (FTS) (Wunch et al., 2010). Finally, we discuss the impact of several bias correction approaches on the estimated total emissions.

This paper is organized as follows. Section 2 summarizes the main characteristics of the satellite and surface observations used in the inversion. The inverse modeling framework is described briefly in Sect. 3. In Sect. 4, we present and discuss the CH₄ emission estimates for the various inversion scenarios, and the validation of the model simulations against independent measurement data. Finally, the conclusions of the study are summarized in Sect. 5.

2 Observations

Table 1 gives an overview of the satellite data used in the inversions. The following sub-sections briefly discuss the characteristics of each set of satellite retrievals. For further details the reader is referred to, e.g., Parker et al. (2011); Butz et al. (2011); Frankenberg et al. (2011); Schepers et al. (2012).

2.1 The GOSAT retrievals

The Thermal And Near infrared Sensor for carbon Observation (TANSO)–Fourier Transform Spectrometer (FTS), onboard the satellite GOSAT (launched by JAXA in January 2009), provides dry-air column-averaged methane mole fractions that can be used in global and regional CH₄ source and sink inversions. The GOSAT XCH₄ are retrieved from a short-wave-infrared spectral analysis of sunlight backscattered by the Earth’s surface and atmosphere.

The proxy retrieval algorithms rely on the small spectral distance between carbon dioxide and methane sunlight absorption bands (1.65 μm for CH₄ and 1.6 μm for CO₂), using the CO₂ column-average dry-air mole fraction (XCO₂) as proxy for the sampled air mass. This helps minimize sys-

tematic errors which may arise due to aerosol scattering and instrument-related effects.

The equation used to obtain the XCH₄ reads as follows:

$$XCH_4 = \frac{[CH_4]_{GOSAT}}{[CO_2]_{GOSAT}} \times XCO_{2,model} \quad (1)$$

The proxy retrieval algorithms considered herein use different XCO₂ model fields. The OCP R (OCO-Proxy) version 4 retrieval algorithm (Parker et al., 2011) from the University of Leicester (UL), developed under the ESA GHG-CCI initiative, derives the column-averaged mole fractions of carbon dioxide from the LMDZ model ((Chevallier et al., 2010); MACC-II CO₂ fields, optimized for the whole period until the end of 2011). The RemoTeC Proxy algorithm (version 1.9/2.0) (Schepers et al., 2012) uses modeled CO₂ total columns obtained from CarbonTracker (Peters et al., 2007) version 2013, with optimized carbon dioxide fields for 2009–2012. Perturbations in the optical path will mostly cancel out when taking the ratio $\frac{[CH_4]_{GOSAT}}{[CO_2]_{GOSAT}}$ of the two measurements. However, Eq. (1) implies that errors in the modeled CO₂ columns propagate directly into the derived XCH₄. The quality of the latter depends thus on the accuracy of the modeled carbon dioxide fields.

The third GOSAT XCH₄ data set used in this study is the RemoTeC FP version 2.1 from SRON/KIT (Butz et al., 2011). The methodology can be summarized as follows. CH₄ and CO₂ columns are retrieved simultaneously with three effective aerosol parameters (amount, size, and height) from GOSAT-FTS measurements at the O₂ A-band around 0.76 microns (μm), the CH₄ and CO₂ absorption bands around 1.6 μm, and the strong CO₂ absorption band around 2.0 μm. Dividing the CH₄ column by the dry air column from the European Centre for Medium-Range Weather Forecast (ECMWF) ERA-Interim data yields the methane dry air mixing ratios (XCH₄). The full physics approach does not require a proxy CO₂ field; instead, the amount of sunlight scattering is estimated directly, together with the XCH₄, from the measured spectra. However, this method can only account for a fraction of the total scattering (Butz et al., 2011). A further trade-off is the lower tolerance to cloud cover (i.e., the method requires a stricter cloud filter). Possible biases in the satellite data are corrected using XCH₄ observations from the Total Carbon Column Observation Network, or TCCON (Wunch et al., 2010), as anchor points.

The filter settings for the GOSAT SRON FP retrievals follow the approach of Butz et al. (2011). We use only observations taken over land (no sun-glint ocean data) that have been screened for clouds. Scenario S1-GOSAT-SRON-FP also assimilates M-gain data (recorded over highly reflective land surfaces). There are considerable differences in the total accepted pixel counts for the full physics vs. the GOSAT proxy methods. Furthermore, GOSAT has a generally much sparser spatial sampling (due to the FTS integration time) compared

to SCIAMACHY. Table 4 reports the total number of satellite data points that were used in each scenario (see also Fig. 4).

2.2 The SCIAMACHY retrievals

The SCIAMACHY Iterative Maximum A Posteriori (IMAP) version 5.5 retrievals used in this study (Frankenberg et al., 2011) are calculated by the proxy method outlined above. Variations in the CO₂ atmospheric columns are accounted for through the use of modeled CarbonTracker carbon dioxide fields (Frankenberg et al., 2011). Problems with the detector on the SCIAMACHY instrument occurred unexpectedly at the end of 2005, and led to a considerable degradation of the instrument performance in the 1.6 μm region relevant for CH₄ retrievals. The main feature of the IMAP v5.5 algorithm that set it apart from its predecessor, version 5.0 (Frankenberg et al., 2008), is the extension of the time series beyond 2005, using a coherent, uniform pixel mask for the entire retrieval period, so as to minimize the impact of pixel degradation (Frankenberg et al., 2011). The pixel deterioration remains visible in the IMAP v5.5 retrievals (higher noise levels are noticeable starting with November 2005). Nonetheless, comparisons with measurements at NOAA surface sites indicate a relatively good consistency of the satellite data time series (Frankenberg et al., 2011). There remain some systematic differences between IMAP v5.5 and v5.0 retrievals (Frankenberg et al., 2011; Bergamaschi et al., 2013a). Following Bergamaschi et al. (2013a), we use a reprocessed version of the IMAP v5.5 retrievals. This version includes CarbonTracker release 2010 CO₂ fields for the year 2009, while CO₂ fields for years 2010 through 2012 are based on non-optimized TM5 forward model runs using optimized CO₂ emissions from previous years (Bergamaschi et al., 2013a).

We assimilate only satellite data over land between 50° N and 50° S. We also discard all pixels whose average surface elevation is not within 250 m of the TM5 model surface height (Bergamaschi et al., 2009, 2013a). To avoid spurious outliers that may have a large impact on the inversion, we filter out any SCIAMACHY or GOSAT XCH₄ retrievals of less than 1500 nmol mol⁻¹ (henceforth abbreviated as ppb), or larger than 2500 ppb.

A SCIAMACHY pixel covers a ground area of 30 km (along track) times 60 km (across track), whereas TANSO-FTS has a ground pixel resolution of 10.5 km (at nadir). Single GOSAT and SCIAMACHY XCH₄ retrievals are averaged on a regular (longitude \times latitude) 1° \times 1° grid over the individual 3 h assimilation time slots. The TM5 XCH₄ are then obtained by vertical integration of the 3-D modeled CH₄ fields interpolated to the same 1° \times 1° grid, using the averaging kernels of the SCIAMACHY and GOSAT retrievals (Bergamaschi et al., 2009).

2.3 The NOAA surface observations

All inversions use high-accuracy CH₄ dry-air mole fraction measurements from a subset of 30 NOAA ESRL sites (Dlugokencky et al., 2013), globally distributed as shown in Fig. 1. Due to the coarse 6° \times 4° resolution of the model, we include only marine and continental background sites. Other locations, e.g., located near the coast or strongly influenced by sub-grid local sources, are excluded from the assimilation. Moreover, the list contains only sites with sufficient data coverage for 2010–2011. The NOAA surface measurements are calibrated against the NOAA2004 methane standard scale, or, equivalently, the World Meteorological Organization Global Atmosphere Watch (WMO GAW) CH₄ mole fraction scale (Dlugokencky et al., 2005).

2.4 Measurement data used for validation

2.4.1 NOAA observations

The simulated methane mixing ratios from all inversions are evaluated against independent observations which have not been assimilated. First, modeled CH₄ mixing ratios are compared against NOAA ship cruise data acquired in 2010 and 2011. These observations allow us to evaluate the simulated concentrations in the marine boundary layer, downwind of continental sources. Further important validation data sources are the NOAA aircraft-based vertical profiles (across North America and the Pacific Ocean, <http://www.esrl.noaa.gov/gmd/ccgg/aircraft/index.html>, and Fig. 1), to validate the modeled methane vertical gradients in the troposphere.

2.4.2 HIPPO aircraft campaigns

Simulated CH₄ fields are also validated against campaigns 3, 4 and 5 of the HIPPER Pole-to-Pole Observations (HIPPO) program (Wofsy, 2011). These three campaigns were run during March/April 2010 (HIPPO-3), June–July 2011 (HIPPO-4), and August–September 2011 (HIPPO-5), for the most part over the Pacific Ocean (see Fig. 1), but also partially above North America (between 87° N and 67° S). The HIPPO data consist of continuous profiles between ca. 150 m and 8500 m altitude. Several profiles extend up to 14 km altitude. For details on the measurement process, which makes use of a quantum cascade laser spectrometer (QCLS), the reader is directed to Kort et al. (2012). In addition, air samples collected using the NOAA Programmable Flask Package were taken during the HIPPO campaigns. Comparison of QCLS measurements and NOAA flask samples taken within the same 10 s interval showed a small bias in the HIPPO data which has been accounted for in our validation (see Fig. 11 and the Supplement): 6 ppb for HIPPO-3, 4.5 ppb for HIPPO-4, and 5.2 ppb for HIPPO-5.

2.4.3 TCCON XCH₄ retrievals

TCCON measures dry-air column-averaged mole fractions of atmospheric methane at several sites across the globe (Table T2 in the Supplement) using Fourier Transform Spectrometers. The TCCON XCH₄ observations have an uncertainty of 7 ppb, and a relative repeatability of 0.2% (Wunch et al., 2010). Only stations with sufficient data coverage during 2010–2011 are used for the validation. The modeled XCH₄ at the TCCON site locations were calculated using the TCCON a priori profiles and averaging kernels (Rodgers and Connor, 2003).

3 Modeling

3.1 Inverse modeling with TM5-4DVAR

We estimate the monthly averages of CH₄ surface fluxes between January 2010 and December 2011 using the TM5-4DVAR inverse modeling system (Meirink et al., 2008b). We also incorporate the further developments described in Bergamaschi et al. (2009, 2010). The statistical best fit of the model-generated 3-D methane fields and observations is achieved by minimization of the following cost functional:

$$\mathcal{J}(\mathbf{x}) = \frac{1}{2}(\mathbf{x} - \mathbf{x}_B)^T \mathbf{B}^{-1}(\mathbf{x} - \mathbf{x}_B) + \frac{1}{2} \sum_{i=1}^n (\mathcal{H}_i(\mathbf{x}) - \mathbf{y}_i)^T \mathbf{R}_i^{-1}(\mathcal{H}_i(\mathbf{x}) - \mathbf{y}_i). \quad (2)$$

Here $\mathbf{x} = (x_{\text{conc}}, x_{\text{em}}, s)$ is the state vector, which comprises the initial CH₄ fields at the beginning of each inversion series x_{conc} , the monthly average emissions x_{em} , and the bias parameters s (Bergamaschi et al., 2009, 2013a). The observations are denoted by \mathbf{y} , while $\mathcal{H}(\mathbf{x})$ is the corresponding model simulation. Finally, \mathbf{B} and \mathbf{R}_i are the parameter and observation error covariance matrices, where the index i indicates the observation window (set to 3 h). We ensured a posteriori CH₄ emissions were positive through the application of a “semi-lognormal” probability density function (PDF) for the a priori emissions $(x_{\text{em}})_B$ (Bergamaschi et al., 2009, 2010). This particular choice of a priori PDF introduces a non-linearity in Eq. (2). The 4DVAR functional \mathcal{J} in Eq. (2) is minimized using the algorithm M1QN3 (Gilbert and Lemaréchal, 1989). The adjoint model (Meirink et al., 2008b; Krol et al., 2008) allows for an efficient computation of the gradient of \mathcal{J} during the minimization process.

TM5 is an off-line transport model (Krol et al., 2005) driven by the ERA-Interim re-analysis meteorological data (Dee et al., 2011) from ECMWF. We use the standard TM5 version (cycle 1), with a global horizontal resolution of 6° × 4° (longitude-latitude), and 25 hybrid pressure vertical layers.

3.2 Inversion settings

The prior emission inventories are identical to those used by Bergamaschi et al. (2013a). We independently optimize four groups of CH₄ emissions: wetlands, rice, biomass burning, and other remaining sources (Bergamaschi et al., 2010, 2013a). A priori uncertainties for each emission category are set to 100% (per model grid cell and month), with the exception of the “remaining sources” whose uncertainty is set to 50%. Wetland, rice, and biomass burning emissions are assumed to be uncorrelated in time, to allow the maximum flexibility when optimizing their seasonal variation. As in Bergamaschi et al. (2010), the temporal correlation of the remaining emissions – assumed to have little seasonal variation – is set to 9.5 months. A Gaussian function of the spatial distance between model grid cells is used to model the spatial emission error correlations, using a correlation length of 500 km, for all emission categories and all scenarios. Horizontal error correlations in the initial methane fields are modeled using a Gaussian distance of 500 km, while error correlations in the vertical direction are described by the National Meteorological Center (NMC) method (Parrish and Derber, 1992; Meirink et al., 2008a). For the satellite data, the reported error is taken as the measurement uncertainty. For the surface observations we prescribe a measurement uncertainty of 3 ppb, while also taking into account the model representation error, estimated from local emissions and 3D gradients of simulated CH₄ mixing ratios (Bergamaschi et al., 2010).

In all inversions the tropospheric methane sink is simulated using hydroxyl (OH) radical fields from a TM5 full chemistry run using the Carbon Bond Mechanism 4 optimized based on methyl chloroform measurements (Bergamaschi et al., 2009, 2010, 2013a). The lifetime of CH₄ is calculated at 10.1 years (total CH₄ vs. tropospheric OH). The fifth generation European Centre Hamburg general circulation model (ECHAM5) Modular Earth Submodel System version 1 (MESSy1) (Jöckel et al., 2006) is used to parameterize the stratospheric chemical destruction of methane by OH, Cl, and O¹(D), using sink averages from 1999–2002.

The number of optimization iterations required to minimize the cost function (Eq. 2) increases with the length of the assimilation window. For this reason, we have split all our inversions into 18 month blocks (Fig. 2), with 6 month spin-down periods (Bergamaschi et al., 2013a). Consecutive blocks overlap by 6 months. The first block starts on 1 January 2009; the third 18 month inversion block ends on 1 July 2012. The inversion for 2009 is considered as spin-up, and not further analysed in this study. The results for the 6 month spin-down periods are also not used in the analysis. A priori 3-D CH₄ concentration fields for 1 January 2009, are taken from a methane inversion constrained only by surface measurements (scenario S1-NOAA of Bergamaschi et al., 2013a), with the exception of scenario S1-SCIA, which uses the optimized concentrations from inversion S1-SCIA of Bergamaschi et al. (2013a). Sixty iterations of the M1QN3

optimization algorithm are used for the cost function minimization in each inversion block for all inversions which include satellite data, and 40 iterations for S1-NOAA (which assimilates only the NOAA surface data).

Initial CH₄ 3-D fields are optimized only for the first inversion block. The other two 18 month blocks start on 1 January from the optimized initial fields of the previous inversion block. This methodology guarantees a closed methane budget across the entire inversion period, i.e., total sources minus total sinks yield the variation in the global CH₄ burden. Additionally, the spin-down periods ensure that surface fluxes for 2010–2011 are constrained by all available observations for at least 6 months after emission.

The inversion scenarios considered in this study are summarized in Table 2. Scenario S1-NOAA is intended as a baseline for all the other inversions; it uses only NOAA ESRL surface station data. Scenarios S1-GOSAT-SRON-PX, S1-GOSAT-SRON-FP, and S1-GOSAT-UL-PX assimilate both NOAA surface data and GOSAT XCH₄ retrievals, whereas S1-SCIA uses SCIAMACHY retrievals and NOAA surface observations. The S1-satellite inversions make use of a second-order polynomial bias correction scheme that is a function of latitude and month (Bergamaschi et al., 2009, 2013a). Table 3 lists the main technical differences between the inversion system considered in the current study and the set-up used by Monteil et al. (2013).

To assess the impact of the bias correction scheme on the posterior emission estimates, we have considered four additional scenarios: S2-GOSAT-SRON-FP, S3-GOSAT-SRON-FP, S2-GOSAT-UL-PX and S3-GOSAT-UL-PX. These differ from S1-GOSAT-SRON-FP and S1-GOSAT-UL-PX by their bias correction scheme. Inversions S3-GOSAT-SRON-FP and S3-GOSAT-UL-PX use a “smooth” bias correction (Bergamaschi et al., 2013a): one bias parameter per degree of latitude and month, 10 ppb prior uncertainty, and a prescribed 20° latitude Gaussian error correlation length. The bias correction coefficients used for S2-GOSAT-SRON-FP and S2-GOSAT-UL-PX are variable in time, but constant with latitude. The choice of bias correction scheme is not found to have a significant impact on the posterior regional emission estimates (shown in Table 5).

The aim of this study is to quantify the impact of the different satellite retrievals on the inverted methane fluxes and concentrations. Hence, all inversions use the same a priori emission inventories (as in Bergamaschi et al., 2013a), and identical OH fields. It is important to note that the surface observations act as constraints (or “anchor points”) for the bias correction scheme.

4 Results and discussion

4.1 Assimilation statistics

The posterior statistics of S1-NOAA through S1-SCIA are summarized in Table 4. Figure 4 shows the frequency distributions of fit residuals (difference between model and observations). The data in Table 4 show that the bias is close to zero for both surface measurements and satellite XCH₄. Moreover, the model performance at the NOAA sites remains virtually identical when satellite data are assimilated: comparing the satellite-based inversions with S1-NOAA we note only a marginal increase in the bias of 0.1–0.2 ppb, and in the RMS difference of about 0.3–0.9 ppb (see also Fig. 3). The statistics of the three GOSAT inversions are almost identical in terms of posterior bias, standard deviation, and RMS difference between retrieved and assimilated XCH₄. While the large global bias in the SCIAMACHY XCH₄ retrievals is for the most part compensated by the bias correction mechanism (Fig. 4), the average standard deviation of the posterior distribution of SCIAMACHY–TM5 fit residuals (sigma = 32 ppb) is much larger than that of the GOSAT inversions (sigma = 9–10 ppb for S1-GOSAT-SRON-PX through S1-GOSAT-UL-PX). The significantly lower standard deviations of the fit residuals of all GOSAT-based inversions demonstrate the much higher precision and relative accuracy of the GOSAT XCH₄ products (compared to the SCIAMACHY retrievals). We note that the GOSAT inversions presented by Monteil et al. (2013) yielded a higher standard deviation (14.7–15.8 ppb). Since they used a previous retrieval version (RemoTeC Proxy v1.0 and Full-Physics v1.0 XCH₄), the lower standard deviation obtained in our study may reflect the further improvement of the GOSAT retrievals. Furthermore, the optimization of the bias correction probably plays some role: while Monteil et al. (2013) applied a constant correction to the GOSAT full physics retrievals before the inversion, based on the comparison with the TCCON data, they did not use any bias correction for the GOSAT proxy retrievals.

4.2 Modeled XCH₄

Figure 4 shows the column-averaged methane mixing ratios for 2010–2011 (two-year averages). The bias-corrected XCH₄ retrievals are plotted in the left maps, while the right-hand side maps show the assimilated XCH₄. Note the much denser data coverage of the SCIAMACHY XCH₄ retrievals (last row of Fig. 4) compared to that of the GOSAT products. For GOSAT, the more stringent selection criteria applied to the full-physics retrievals result in significantly lower pixel density than that achieved by the two proxy XCH₄ retrievals (see also Table 4).

The 4DVAR assimilation system is able to capture most major regional patterns of the observed XCH₄ fields, e.g., the pronounced XCH₄ enhancements over southeast Asia. Over

tropical South America, the agreement between retrieved and assimilated XCH₄ patterns is generally better for the three GOSAT-based inversions than for SCIAMACHY (e.g., over Columbia and Venezuela). Note, however, the lower data density of the GOSAT retrievals (especially of the GOSAT Full Physics retrievals) over those areas compared to SCIAMACHY. The different GOSAT products show overall very good consistency regarding the spatial XCH₄ patterns (in particular the two GOSAT proxy retrievals), and result in only small to moderate calculated bias corrections (maximum 10–20 ppb), indicating good consistency with the surface observations. In contrast, the SCIAMACHY XCH₄ require a significantly higher bias correction (varying with latitude by up to ca. 40 ppb). There are various indications that the SCIAMACHY IMAP v5.5 XCH₄ have a complex bias structure (e.g., the comparison with previous IMAP v5.0 XCH₄ retrievals examined by (Frankenberg et al., 2011)), which cannot be fully compensated by our polynomial bias correction. Furthermore, Houweling et al. (2014) showed recently that the bias of the SCIAMACHY IMAP v5.5 retrievals is strongly correlated with water vapour.

4.3 A posteriori methane fluxes

Figure 6 shows the spatial distribution of emissions, averaged over the two years (2010–2011). The maps on the left side show the a priori (top) and a posteriori fluxes, while the middle panel shows the longitudinal average partitioning among the 4 source categories optimized in the inversions. The right-hand side maps display the differences between a posteriori and a priori emissions for our baseline inversion S1-NOAA, and for the satellite inversions S1-GOSAT-SRON-PX through S1-SCIA the difference between the a posteriori emissions of these inversions and S1-NOAA. While the satellite inversions yield significantly different spatial emission patterns compared to the NOAA-only inversion (due to the constraints of the satellite data over the continents), they show overall good qualitative agreement across all satellite inversions. This is visible in particular in the difference plots on the right side of Fig. 6, that show similar regional emission increments relative to the NOAA-only inversion, especially over tropical Africa and the United States. While the NOAA-only inversion results in a significant increase of the emission hot spot over the Congo Basin (which is a prominent feature in the applied wetland inventory, see Bergamaschi et al., 2007, 2009), all satellite inversions reduce the emissions from this hotspot significantly, and instead increase the emissions in tropical eastern Africa (see also the “wetlands” panel in Fig. 7). Note that S1-GOSAT-SRON-FP calculates slightly lower emission rates for equatorial Africa, likely due to the absence of observations available directly over that region (Fig. 4). Especially for the NOAA-only inversion, the a posteriori CH₄ fluxes over the tropics depend in large part on the choice of prior inventory. Unfortunately, their uncertainties remain very high, and the comparison of global wetland

models by (Melton et al., 2013) shows large discrepancies in estimated CH₄ emissions among the models. The relatively consistent spatial patterns over tropical Africa found in this study for the different satellite inversions demonstrate that the satellite data combined with the inverse models provide significant constraints on the CH₄ emissions from this region.

The total emission latitudinal averages (shown in the bottom panel of Fig. 7) are relatively consistent among all five scenarios. The decrease in fluxes over the temperate northern hemisphere can be largely attributed to an attenuation of EDGAR anthropogenic emission hot spots over Eastern Europe, as seen in the panel labeled “other“. However, there remain noticeable inter-scenario differences in the category averages, particularly for wetland and biomass-burning emissions at sub-Equatorial latitudes.

Over North America, the satellite inversions result in a significant redistribution of CH₄ emissions from the northeastern USA to the middle south. A similar spatial pattern, with significantly higher methane emissions over the south-central United States compared to bottom-up inventories, has recently been reported by Miller et al. (2013), and attributed by the authors of that study mainly to fossil fuel emissions. Furthermore, a recent comprehensive review by Brandt et al. (2014), which analyzed a large number of bottom-up and top-down studies ranging from facility-level, over regional level and up to country level, suggested a systematic underestimation of methane emissions from North-American natural gas systems in bottom-up inventories. Although the spatial redistribution of CH₄ emissions over the USA calculated by our satellite inversions appears to be consistent with these studies, we emphasize that the applied coarse model resolution and the limitations of the inverse modeling system in differentiating between source categories do not allow us to attribute these emission increments to specific sources.

Methane fluxes aggregated over the TRANSCOM regions (Gurney et al., 2008) are shown in Fig. 8, and Table 5. All inversions show a small increase in the two-year global total emissions over the prior, from 1.8 Tg CH₄ yr^{−1} for S1-GOSAT-SRON-PX to 5 Tg CH₄ yr^{−1} for the SCIAMACHY-based S1-SCIA. Emissions in the mid latitudes of the northern hemisphere are reduced in all scenarios (mainly across Europe and Temperate Eurasia, see Fig. 8b) although there are considerable differences between the flux adjustments calculated for each inversion, ranging from −17 Tg CH₄ yr^{−1} for the GOSAT full-physics S1-GOSAT-SRON-FP, to −37.5 Tg CH₄ yr^{−1} for S1-SCIA. The negative increments in the northern hemisphere are compensated by across-the-board increases in tropical emissions (between 30° N and 30° S) over the prior, between 18.6 Tg CH₄ yr^{−1} for S1-GOSAT-SRON-FP, and 48.4 Tg CH₄ yr^{−1} for S1-SCIA. The net increase in the Southern Hemisphere fluxes can be mainly attributed to increased emissions over Brazil and sub-equatorial Africa. Part of the net increase in the SH

could be due to a bias in the inter-hemispheric mixing of TM5, as recently diagnosed by SF₆ simulations (Patra et al., 2011; Monteil et al., 2013). To tackle this problem, a new parameterization of convective fluxes has been implemented for TM5, based on the ERA-Interim convective fluxes (Berrisford et al., 2011), instead of the scheme of Tiedtke (1989). While it increases inter-hemispheric transport, the new parameterization has a significant impact on the simulated mixing ratios in the continental boundary layer (results not shown). Further investigations are needed to fully evaluate the impact and quality of the new convection scheme (which is beyond the scope of the present study).

Monteil et al. (2013) have reported that inversions using the GOSAT SRON proxy retrievals led to larger Asian emissions than those estimated using the SRON full-physics XCH₄ data set. We noticed a similar pattern in our inversions, particularly above tropical Asia where S1-GOSAT-SRON-FP flux estimates are circa 6.5 Tg CH₄ yr⁻¹ lower than those of the GOSAT SRON proxy scenario S1-GOSAT-SRON-PX. Schepers et al. (2012) attribute this discrepancy in the emission estimates to a regional overestimation of CH₄ mixing ratios in the proxy retrieval algorithm, caused by deficiencies in the applied CO₂ fields. The two GOSAT proxy retrievals yield overall relatively similar emission patterns. There are, however, some differences in the exact magnitude of the regional-scale fluxes calculated by S1-GOSAT-SRON-PX and S1-GOSAT-UL-PX, e.g., a larger decrease in temperate Eurasian fluxes when the GOSAT OCPD retrievals are assimilated (see Fig. 6).

Several recent studies (Butz et al., 2011; Schepers et al., 2012) indicated that the reported precision of XCH₄ satellite retrievals may be too high. To investigate the impact of applied satellite uncertainties, we considered one additional scenario, in which the reported errors of the GOSAT-SRON-PX retrievals were scaled by a factor of 1.5. This sensitivity experiment did not lead to significant changes in the a posteriori regional emission patterns (results not shown).

4.4 Model validation

All the inversion results are thoroughly validated against independent measurement data sets covering the atmospheric boundary layer (BL), the free troposphere (FT), as well as the upper troposphere and lower stratosphere (UTLS). Since the observations considered for validation have not been used in the assimilation, they provide an independent verification of the modeled XCH₄. Figure 9 gives an overview of the results for all inversions and validation data sets (for a total of slightly more than 80 900 observations). See Sect. 2.4 and Fig. 1 for details on each data set. The root mean square (RMS) differences shown in Fig. 9 have been averaged over all available measurements during 2010–2011. In general the optimized CH₄ mixing ratios have lower RMS differences than the prior concentrations. It is important to note that the a priori shown in Fig. 9 is already partly optimized, given

that inversion blocks 2 and 3 (for 2010, and 2011, respectively) start from optimized initial fields (see the discussion in Sect. 3.2). The validation performance of scenario S1-NOAA is generally no worse than that of the satellite inversions. This is likely due to the fact that validation data are generally located far from the regions where the changes in emissions patterns occur (see Fig. 1 and 6), an exception being the continental United States, where the agreement between the modeled mixing ratios and boundary layer NOAA data improves slightly when assimilating satellite retrievals (“BL” panel in Fig. 9). This result is, however, difficult to interpret given the coarse resolution of the model.

4.4.1 TCCON XCH₄ data

TCCON provides retrievals of CH₄ concentrations at globally distributed locations using ground-based Fourier Transform Spectrometers (Wunch et al., 2010). We compare our modeled XCH₄ with GGG2012 TCCON retrievals. Figure 10 shows the bias and RMS difference between the TM5 and TCCON XCH₄, averaged over the entire inversion period. Only stations with sufficient measurement data coverage for 2010–2011 are shown. The grey bars indicate the a priori bias and RMS. There is a noticeable improvement in the bias over the prior at the northernmost TCCON stations in Fig. 10. At other regional stations the improvement is modest, and at some stations, e.g., at Four-Corners (FCO), the XCH₄ bias slightly deteriorates after the assimilation. However, a recent high-resolution study of Kort et al. (2014) identified the Four-Corners area as a large methane anomaly likely caused by regional sources such as oil, gas, and coal-bed methane mining and processing.

We note a systematic trend in the bias from north to south (except for FCO). The positive bias at high northern latitudes could be partly due to overestimated CH₄ mixing ratios in the stratosphere (see the comparison of Bergamaschi et al. (2009) with balloon measurements, and comparisons with HIPPO data in Sect. 4.4.2 and Fig. 11). However, there is also some uncertainty in the TCCON FTS data, since the stratospheric contribution is not directly calibrated and validated (Wunch et al., 2010; Geibel et al., 2012). In future studies, the AirCore CH₄ data from NOAA ESRL (Karion et al., 2010) may also serve as an independent benchmark of both model and TCCON XCH₄ in the stratosphere.

4.4.2 HIPPO aircraft campaigns

Figure 11 shows the bias corrected HIPPO data for all three campaigns (leftmost panels), and modeled mixing ratios for scenario S1-GOSAT-SRON-PX. There is overall a good agreement between the model simulations and the HIPPO observations (similar results for scenarios S1-GOSAT-SRON-FP through S1-SCIA are reported in the Supplement).

The rightmost panels in Fig. 11 show the average bias as a function of altitude and latitude band: extra-tropical NH (red points), tropics (light green), and extra-tropical SH regions (blue). Agreement between model simulations and the HIPPO measurements in the free troposphere is generally very good for all inversions. However, the bias increases significantly above 300 hPa for all three HIPPO campaigns, particularly in the extra-tropical regions. A similar bias pattern has been reported by Bergamaschi et al. (2013a, Fig. 10). This abrupt deterioration of model performance in the stratosphere is likely caused by deficiencies of the parameterization of the stratospheric sink at high latitudes, and the inability of the coarse-resolution TM5 model to resolve the small-scale dynamics of the stratospheric–tropospheric exchange.

5 Conclusions

This study compares several inversions of global CH₄ emissions for 2010–2011, using four different satellite XCH₄ products: the SCIAMACHY IMAPv5.5 retrievals (Frankenberg et al., 2011), the SRON/KIT GOSAT RemoTeC Proxy v1.9/v2.0 and Full-Physics v2.1 (Butz et al., 2011; Schepers et al., 2012) retrievals, and the GOSAT OCPR v4.0 product from the University of Leicester (Parker et al., 2011). All inversions considered herein are further constrained by high-accuracy methane measurement data from the NOAA ESRL global station network (Dlugokencky et al., 2013). The modeled 3-D CH₄ fields have been validated against multiple sets of independent observations that were not assimilated.

The inversion results demonstrate clear improvements in the precision and relative accuracy of the GOSAT XCH₄ retrievals over SCIAMACHY. The standard deviations of the model to observation fit residuals of the GOSAT-based inversions (9–10 ppb) are significantly lower than the value calculated for the SCIAMACHY scenario (~ 32 ppb). Furthermore, the monthly bias corrections applied to the GOSAT retrievals (Fig. 4) are only a fraction of those estimated for the SCIAMACHY retrievals. All the satellite inversions yield qualitatively consistent regional emission patterns, particularly over tropical Africa and the United States. The inversions show areas of increased methane emissions relative to inventories over the south-central USA, a result consistent with the estimates of Miller et al. (2013), and the recent review of Brandt et al. (2014). The coarse resolution of the model used in this study, and the limitations of the inverse modeling system in differentiating between different source categories do not allow us to attribute these emission increments to specific sources. Nonetheless, the results warrant a more in-depth analysis of methane emissions over the North American continent.

It is very encouraging that the GOSAT proxy and GOSAT pull-physics retrievals (which are fundamentally different products of satellite data processing) yield qualitatively very similar spatial emission patterns in the inversions. However,

there remain some quantitative differences between the emission increments retrieved by each scenario (particularly over Europe, India, and South America), and the derived two-year average regional fluxes for the TRANSCOM regions differ by up to 15 Tg CH₄ yr⁻¹. For the GOSAT UL Proxy and SRON Full-Physics scenarios, the retrieved regional emission estimates show little sensitivity to the particular choice of optimized bias correction scheme (Table 5).

The satellite inversions show similar validation performance. The posterior CH₄ mixing ratios have, in general, a lower RMS difference to the observations than the prior concentrations. However, validation against the HIPPO profiles demonstrates that a significant bias remains present in the UTLS at higher latitudes, indicating possible deficiencies of the parameterization of the stratospheric sink, or potentially also transport within the stratosphere. Furthermore, increased horizontal and vertical model resolutions may improve the representation of stratospheric–tropospheric exchange, leading to a better agreement with observations in the upper atmosphere. The observed deficiencies of TM5 in the UTLS and stratosphere at high latitudes may partly explain the noticeable north-south trend in the bias between TM5 and TCCON XCH₄ (Fig. 10).

An important diagnostic of the observational constraints are the a posteriori uncertainties. Our choice of non-Gaussian prior statistics for the methane fluxes precludes the use of the Lanczos algorithm for uncertainty quantification (Meirink et al., 2008b). Alternative approaches are currently being investigated, including the ensemble method described by Chevallier et al. (2007).

Acknowledgements. The authors thank the TCCON PIs for making their measurement data available. The TCCON XCH₄ data (GGG2012) were obtained from the TCCON Data Archive, operated by the California Institute of Technology, and hosted at the website <http://tcon.ipac.caltech.edu/>. US funding for TCCON comes from NASA's Carbon Cycle Program, grant number NNX11AG01G, the Orbiting Carbon Observatory Program, and the DOE/ARM Program. The European TCCON groups involved in this study acknowledge financial support by the EU infrastructure project InGOS. The University of Bremen acknowledges financial support of the Bialystok and Orleans TCCON sites from the Senate of Bremen and EU projects IMECC, GEOMon and InGOS, as well as maintenance and logistical work provided by AeroMeteo Service (Bialystok) and the RAMCES team at LSCE (Gif-sur-Yvette, France), and additional operational funding from the National Institute for Environmental Studies (NIES, Japan). The authors acknowledge Nicholas Deutscher for his kind assistance with the TCCON data processing. CarbonTracker CT2013 results provided by NOAA ESRL, Boulder, Colorado, USA from the website at <http://carbontracker.noaa.gov>. This work has been supported by the European Commission Seventh Framework Programme (FP7/2007–2013) projects MACC under grant agreement 218793 and MACC-II under grant agreement 283576. The ECMWF meteorological data has been preprocessed by Philippe Le Sager into the TM5 input format. We thank Greet Janssens-Maenhout for provid-

- ing the EDGARv4.2 emission inventory, and Christoph Brühl for providing the stratospheric CH₄ sinks from the ECHAM5/MESSy1 model. ECMWF has kindly provided the necessary computing resources, under the special project “Global and Regional Inverse Modeling of Atmospheric CH₄ and N₂O” (2012–2014). H. Boesch and R. Parker acknowledge funding by the NERC National Centre for Earth Observation and the ESA Climate Change Initiative. André Butz acknowledges support by the Emmy-Noether programme of the Deutsche Forschungsgemeinschaft (DFG) through grant number BU2599/1-1 (RemoteC). Finally, we thank Peter Rayner and the anonymous reviewers for their insightful comments on the manuscript.
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Table 1. Satellite data used in the inversions.

Satellite/Instrument	Algorithm	Proxy CO ₂ model	Data provider	Temporal data coverage
ENVISAT/SCIAMACHY	IMAP v5.5	CarbonTracker	SRON	Jan 2009–Mar 2012
GOSAT/TANSO-FTS	OCPR v4.0	LMDZ	Univ. of Leicester	Jun 2009–Dec 2011
GOSAT/TANSO-FTS	RemoTeC Proxy v1.9/v2.0	CarbonTracker 2013	SRON/KIT	v1.9: Jan 2009–Oct 2011 v2.0: Oct 2011–Jun 2012
GOSAT/TANSO-FTS	RemoTeC FP v2.1	–	SRON/KIT	Jun 2009–Jun 2012

Table 2. Inversion scenarios.

Inversion	Assimilated observations
S1-NOAA	NOAA ESRL surface measurements only
S1-GOSAT-SRON-PX	NOAA ESRL surface measurements and GOSAT SRON RemoTeC v1.9/v2.0 XCH ₄ retrievals
S1-GOSAT-SRON-FP	NOAA ESRL surface measurements and GOSAT SRON FP v2.1 XCH ₄ retrievals
S1-GOSAT-UL-PX	NOAA ESRL surface measurements and GOSAT OCPR v4.0 XCH ₄ retrievals
S1-SCIA	NOAA ESRL surface measurements and SCIAMACHY IMAP v5.5 XCH ₄ retrievals
S2-GOSAT-SRON-FP	as S1-GOSAT-SRON-FP, with a constant bias correction instead of 2nd order polynomial
S3-GOSAT-SRON-FP	as S1-GOSAT-SRON-FP, with a smooth bias correction
S2-GOSAT-UL-PX	as S1-GOSAT-UL-PX, with a constant bias correction instead of 2nd order polynomial
S3-GOSAT-UL-PX	as S1-GOSAT-UL-PX, with a smooth bias correction

Table 3. Inversion settings: current study vs. Monteil et al. (2013)

	Current study	Monteil et al. (2013)
Prior PDFs	Semi-lognormal	Gaussian (may result in negative a posteriori emissions)
Satellite retrievals	ENVISAT/SCIAMACHY IMAP v5.5 GOSAT/TANSO-FTS RemoTeC Proxy v1.9/2.0 GOSAT/TANSO-FTS RemoTeC FP v2.1 GOSAT/TANSO-FTS OCPR v4.0	ENVISAT/SCIAMACHY IMAP v5.5 GOSAT/TANSO-FTS RemoTeC Proxy v1.0 GOSAT/TANSO-FTS RemoTeC FP v1.0
Bias correction	Function of latitude and month, optimized in the inversion (for all satellite products).	GOSAT RemoTeC FP v1.0: Correction by a single coefficient (1.0037). GOSAT RemoTeC Proxy v1.0: no bias correction applied. SCIAMACHY IMAP v5.5: Constant factor, plus seasonally varying bias correction term based on specific humidity (Houweling et al., 2014).
Stratospheric sink	ECHAM5/MESSy1.	Cambridge 2D model (Velders, 1995) with a correction based on HALOE/CLAES climatology applied above 50 hPa.
Tropospheric OH	TM5 full chemistry run based on CBM4 (see Section 3.2)	Spivakovsky et al. (2000), with a scaling factor of 0.92.
Satellite retrieval errors	Uses reported XCH ₄ errors.	The reported GOSAT retrieval uncertainties are scaled by a factor of 1.5 before the inversion.
Emission categories	Four categories optimized independently.	Total emissions.
Prior emission uncertainties	50–100% per category, grid cell, and month (see section 3.2).	50% per grid cell and month.
Target period	January 2010 - December 2011	April 2009 - August 2010

Table 4. Statistics for inversions S1-NOAA through S1-SCIA: NOAA surface measurements (left) and satellite data (right). See Fig. 3 for the frequency distributions of fit residuals.

Inversion	NOAA ground stations			Satellite		
	<i>n</i>	Bias [ppb]	RMS [ppb]	<i>n</i>	Bias [ppb]	RMS [ppb]
S1-NOAA	3418	0.2	11.5	–	–	–
S1-GOSAT-SRON-PX	3418	0.3	12.4	106 854	−0.3	9.2
S1-GOSAT-SRON-FP	3418	0.4	12.1	31 201	−0.3	10.4
S1-GOSAT-UL-PX	3418	0.4	11.8	129 916	−0.1	8.9
S1-SCIA	3418	0.3	12.0	432 008	−0.9	32.3

Table 5. Two-year average CH₄ emissions (Tg CH₄ yr^{−1}) for the TRANSCOM land regions and 30° latitude bands. The prior emission inventories are as used by Bergamaschi et al. (2013a). The global total includes the contribution of ocean regions.

Region	Prior	S1-NOAA	S1-GOSAT-SRON-PX	S*-GOSAT-SRON-FP			S*-GOSAT-UL-PX			S1-SCIA
				S1	S2	S3	S1	S2	S3	
BNA	13.0	11.5	11.0	12.2	13.3	12.2	10.3	11.4	10.2	10.3
TNA	38.5	47.6	44.7	44.8	41.3	43.1	52.1	47.3	51.5	45.6
TrSA	63.7	74.9	68.9	79.4	79.6	80.7	70.7	72.4	71.6	71.8
TSA	37.5	40.9	40.9	41.7	41.5	40.5	41.3	42.3	40.7	40.2
NAf	36.7	43.0	36.1	48.0	52.7	48.1	38.3	41.7	40.2	50.6
SAf	28.5	36.4	41.6	36.4	37.7	36.2	38.4	40.0	35.7	42.0
BEr	18.1	18.1	20.6	16.8	16.7	17.0	17.0	17.4	16.7	15.4
TEr	131.4	110.1	107.5	110.4	104.7	108.9	104.0	98.1	103.0	109.6
TrAs	69.6	75.9	74.2	67.7	73.4	68.6	77.2	81.4	77.4	76.8
Aus	5.8	4.8	9.1	4.7	3.5	4.4	6.9	6.2	7.8	4.3
Eur	46.4	29.5	38.6	33.8	29.0	35.7	36.8	32.9	38.0	28.9
Global total	535.5	538.1	537.3	537.7	537.9	537.2	538.2	538.2	538.4	540.5
Arctic	19.9	17.6	21.1	18.3	19.7	18.3	17.7	20.1	17.3	18.2
NH-mid	183.8	156.2	163.9	166.7	156.0	165.2	161.7	149.7	158.5	146.2
NHTr	193.8	202.8	182.9	192.1	196.7	193.8	197.2	199.7	207.7	215.1
SHTr	127.7	153.9	157.4	148.1	156.5	148.5	150.6	160.1	141.6	154.8
SH-mid	14.3	11.6	15.0	16.1	12.9	14.8	13.7	11.5	16.0	8.9

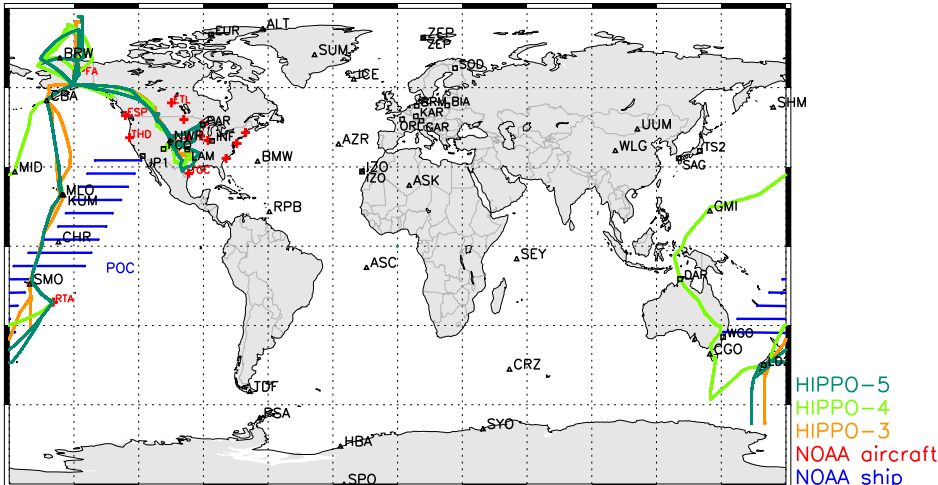


Fig. 1. Observation data map indicating the locations of NOAA surface stations used in the inversions (triangle symbols; see also Table T1 in the Supplement). The squares indicate the TCCON station locations. Some of the NOAA and TCCON stations are co-located. The regions covered by NOAA ship cruises (labeled as POC) are displayed through the horizontal blue lines, which indicate the longitudinal ranges within each 5° latitude band. In addition, we show the NOAA aircraft profile locations (red crosses), and the HIPPO 3–5 transects used for validation.

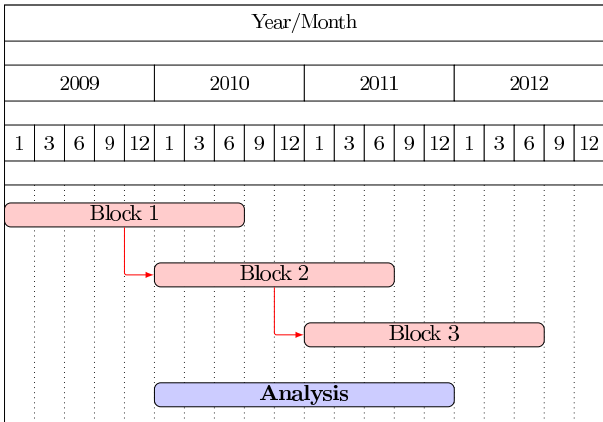


Fig. 2. The inversion settings, as described in Sect. 3.2. Inversion blocks 2 and 3 start (on 1 January 2010, and 1 January 2011, respectively) from the optimized 3-D CH₄ fields calculated by the previous block.

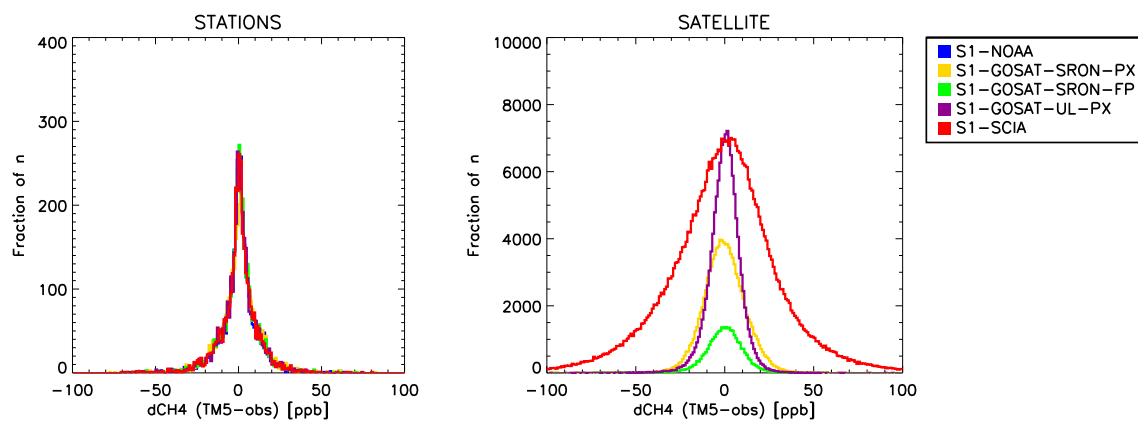


Fig. 3. Frequency distributions of model–observation residuals ($d\text{CH}_4$) for satellite and station data (2010–2011). Both station and satellite data are distributed across 1 ppb bins. The total number of surface measurements or retrievals is denoted by n . The bias and RMS of each inversion are shown in Table 4.

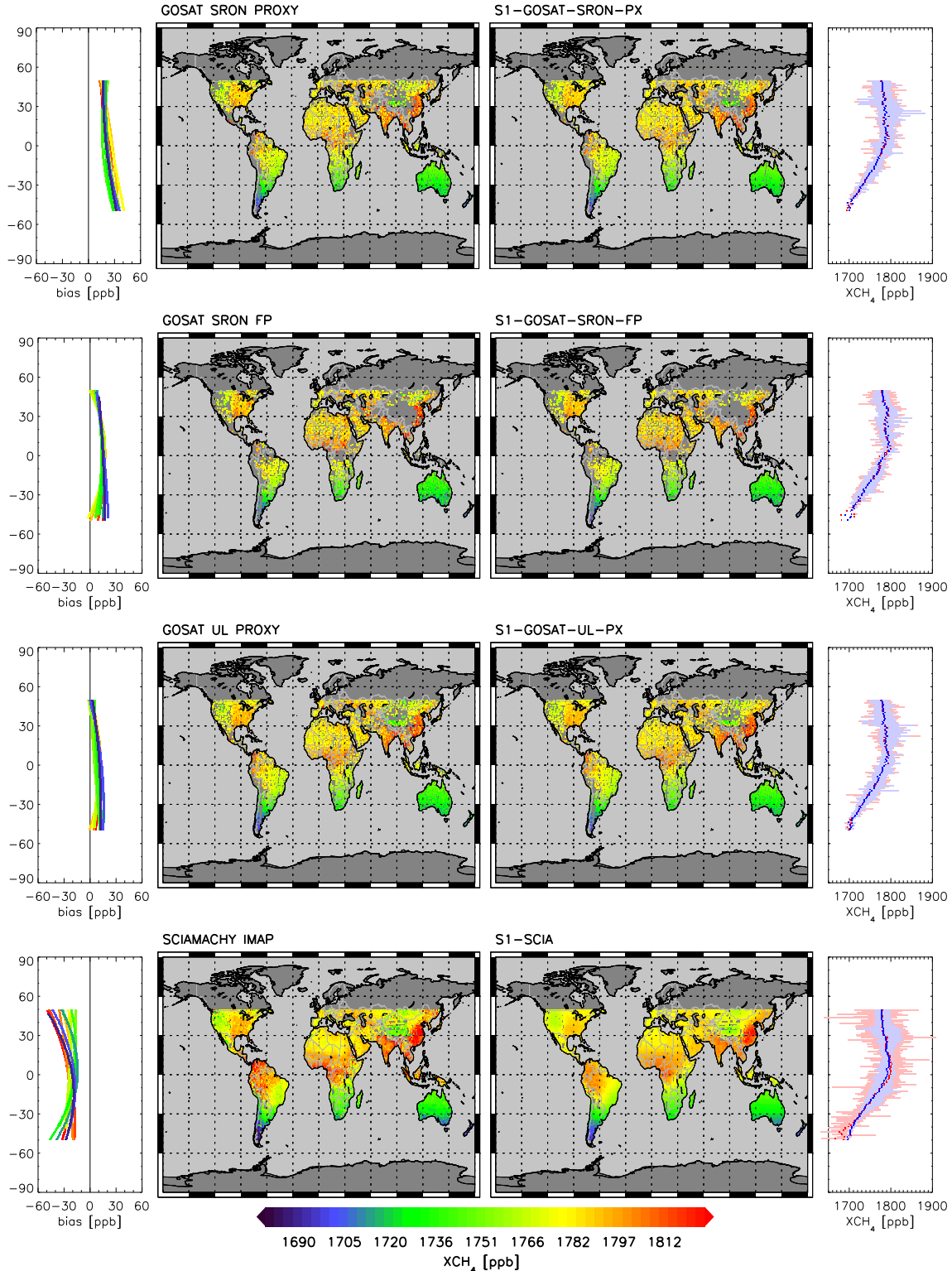


Fig. 4. Column-averaged CH₄ mixing ratios (XCH₄): bias-corrected satellite retrievals vs. TM5-4DVAR. The leftmost plots show the monthly average bias corrections (in ppb) applied to the satellite data for January 2010–December 2011. The panels on the right display the two-year latitudinal average XCH₄ values (red: satellite, blue: TM5-4DVAR) and the corresponding minimum and maximum values across the longitude.

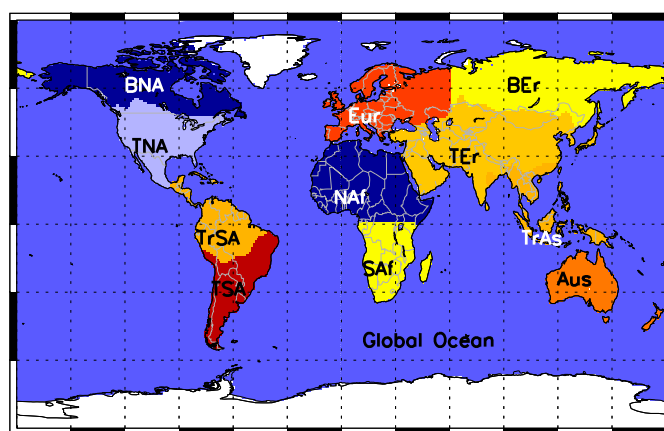


Fig. 5. The TRANSCOM emission regions used in this study (at $1^\circ \times 1^\circ$ resolution). The land regions are labeled as follows: boreal North America (BNA), temperate North America (TNA), tropical South America (TrSA), temperate South America (TSA), Europe (Eur), North Africa (NAf), South Africa (SAf), boreal Eurasia (BEr), temperate Eurasia (TEr), tropical Asia (TrAs), and Australasia (Aus). White areas (ice) are not assigned to any region.

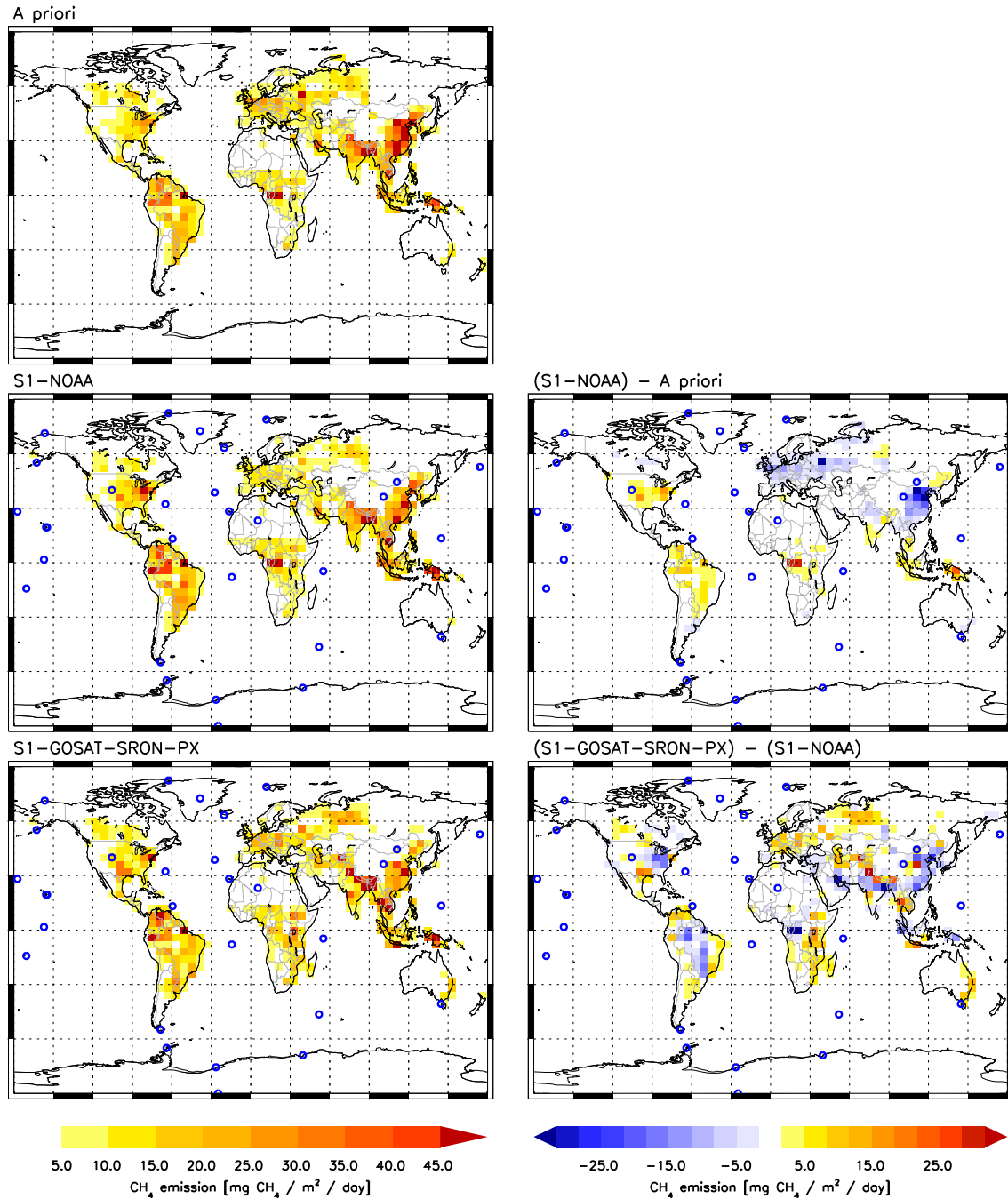


Fig. 6. Left: a posteriori two-year average emissions for S1-NOAA and S1-GOSAT-SRON-PX. The a priori emissions are shown in the topmost plot. White areas indicate grid cells with very low emissions (less than $5 \text{ mg CH}_4 \text{ m}^{-2} \text{ day}^{-1}$). Right column: for S1-NOAA the difference between posteriori and a priori emissions is shown, while for all satellite inversions the panels show the difference between the a posteriori emissions of these inversions and S1-NOAA.

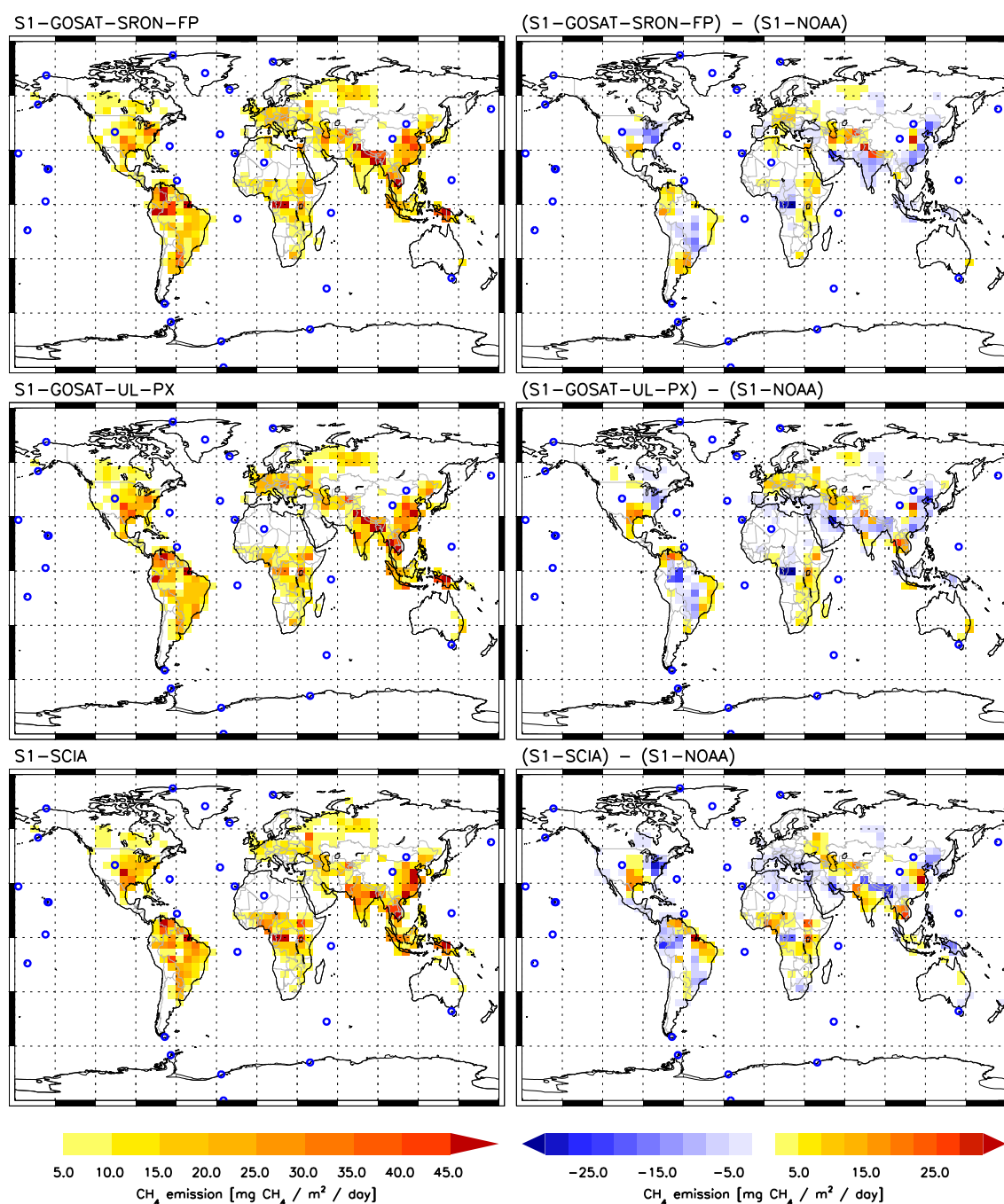


Fig. 6. Continued – scenarios S1-GOSAT-SRON-FP – S1-SCIA.

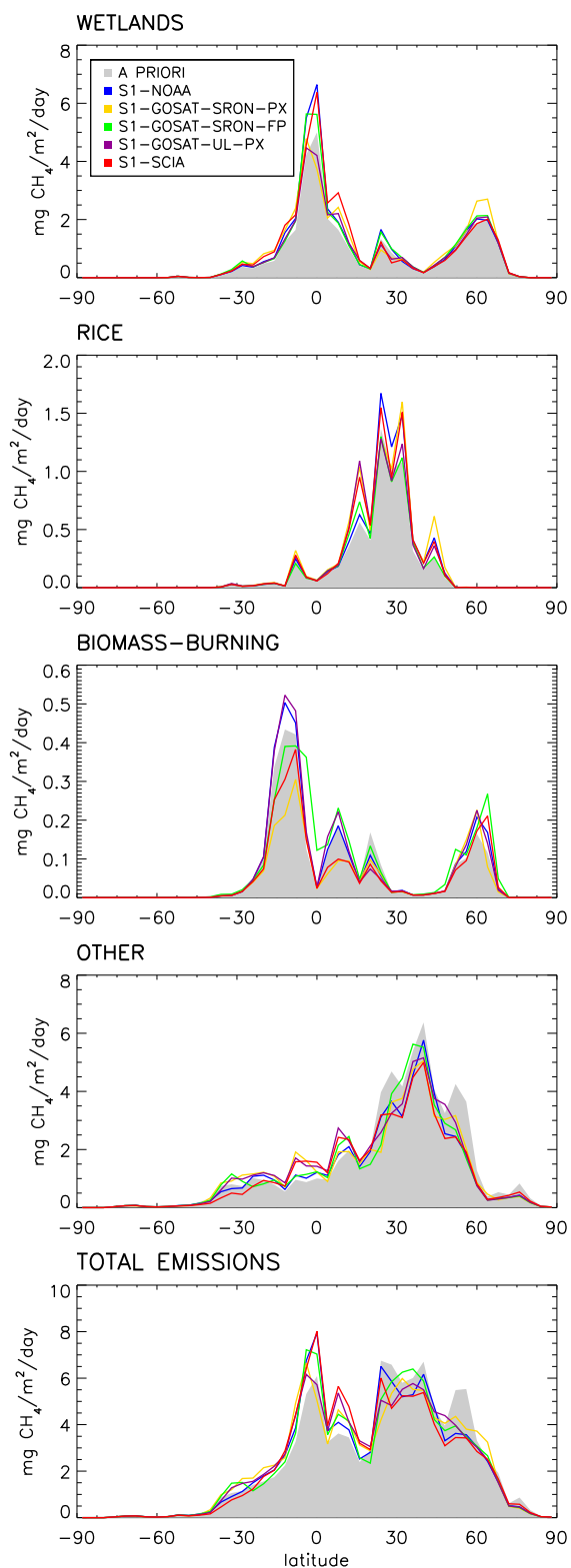


Fig. 7. Two-year latitudinal averages of CH_4 emissions, shown for the different source categories optimized in the inversions, and for the total emissions. The gray areas correspond to the prior emissions.

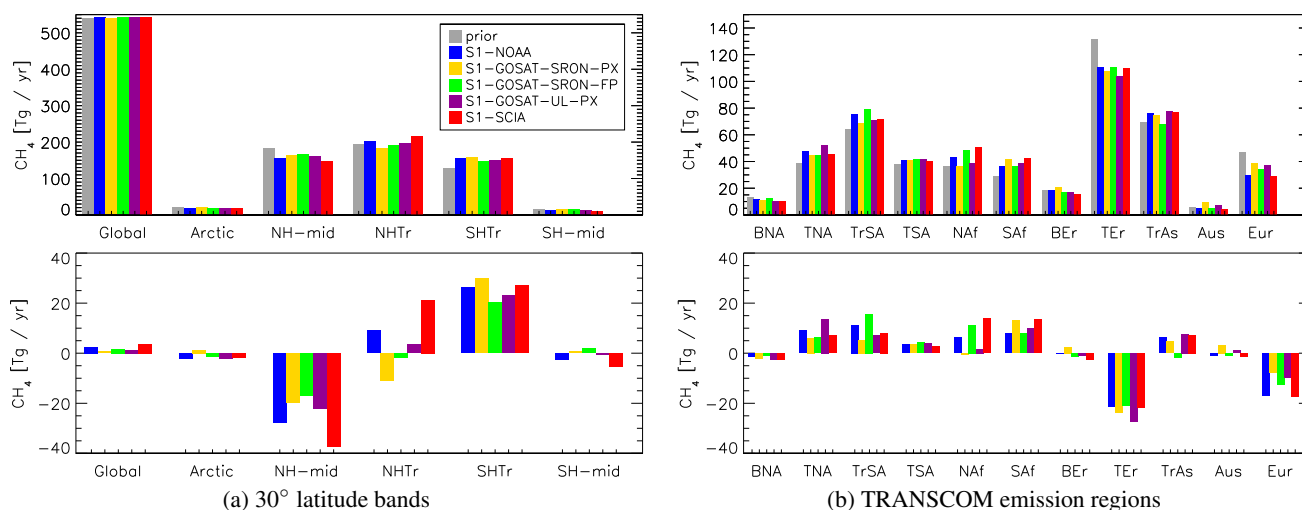


Fig. 8. Average yearly CH₄ emissions for the pre-defined regions. Top panels show total surface fluxes (in Tg CH₄ yr⁻¹), while increments from the prior are given in the bottom panels. Yearly totals are shown in (a), along with surface fluxes attributed to each 30° latitude band. The Antarctic region (not shown here) is estimated to be responsible for less than 0.1 Tg yr⁻¹ of CH₄. See Fig. 5 for the definition of the modified TRANSCOM regions shown in (b).

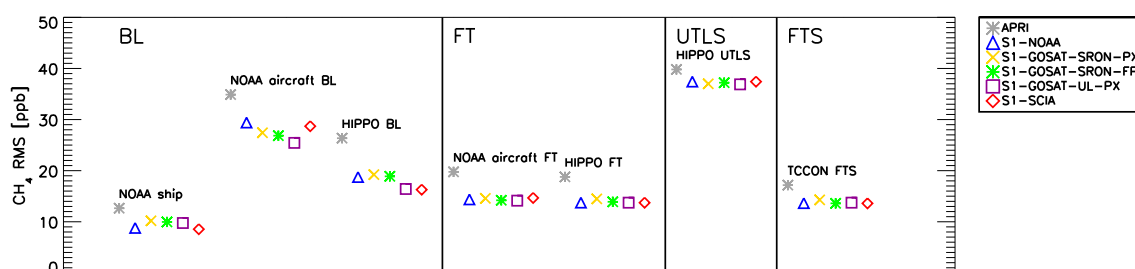


Fig. 9. Validation against independent measurement data sets for all inversions. The plot shows the RMS (in ppb) of differences between modeled methane mixing ratios, and observation data in the boundary layer (“BL”), free troposphere (“FT”), and upper troposphere/lower stratosphere (“UT/LS”). Observation data sources: NOAA shipboard samples, vertical profiles from NOAA aircraft sampling, and the HIPPO campaigns 3–5. Validation results for the Fourier Transform Spectrometer CH₄ total column data from TCCON are shown in a separate panel (“FTS”). The prior (APRI) is already partly optimized (see section 3.2).

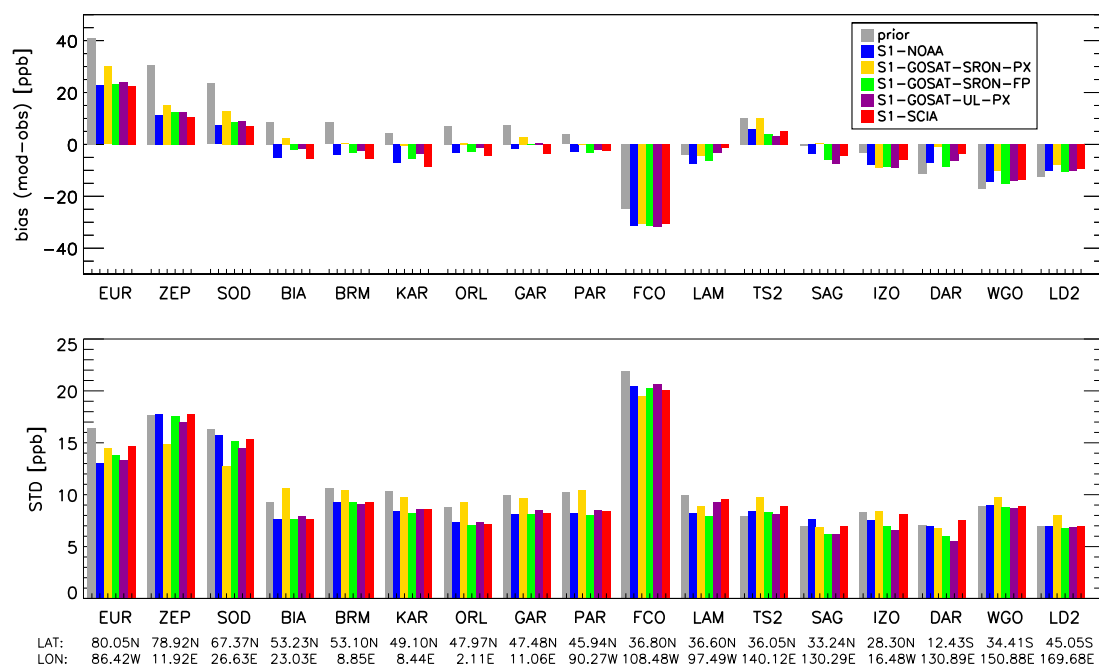


Fig. 10. Model validation against TCCON data across all measurement stations with significant data coverage during our inversion period. Prior values are given by the grey bars. Upper panel: bias (in ppb). Lower panel: standard deviation.

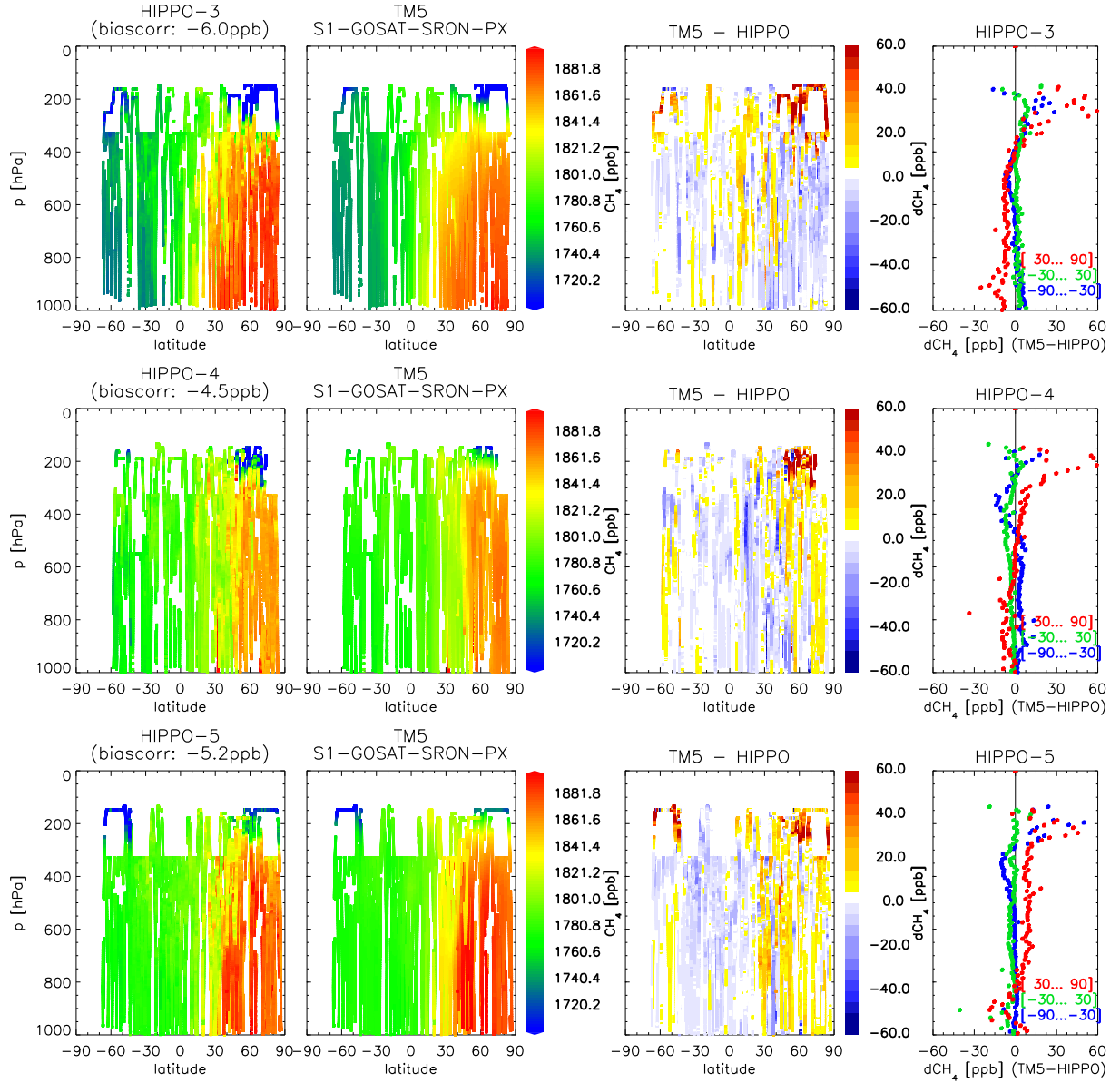


Fig. 11. Scenario S1-GOSAT-SRON-PX: validation against HIPPO campaigns 3–5 (southbound and northbound flights). Rightmost panels show the average bias as a function of latitude: extra-tropical northern hemisphere (NH) in red, extra-tropical SH in blue, and the tropics in green. HIPPO validation results for the other inversions are shown in the Supplement.