

# Global CH<sub>4</sub> Fluxes Derived from JAXA/GOSAT Lower Tropospheric Partial Column Data and the CTE-CH<sub>4</sub> Atmospheric Inverse Model

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**Abstract.** Satellite-driven inversions provide valuable information about methane (CH<sub>4</sub>) fluxes, but the assimilation of total column-averaged dry-air mole fractions of ~~CH<sub>4</sub>~~CH<sub>4</sub> (XCH<sub>4</sub>) has been challenging. This study explores, for the first time, the potential of the new lower tropospheric partial column (pXCH<sub>4</sub>\_LT) GOSAT data, retrieved by the Japan Aerospace Exploration Agency (JAXA), to constrain global and regional CH<sub>4</sub> fluxes. Using the CarbonTracker Europe-CH<sub>4</sub> atmospheric inverse model, we estimated CH<sub>4</sub> fluxes between 2016–2019 by assimilating the JAXA/GOSAT pXCH<sub>4</sub>\_LT and XCH<sub>4</sub> data and surface CH<sub>4</sub> observations independently of each other. The Northern Hemisphere CH<sub>4</sub> fluxes derived from the ~~JAXA/GOSAT~~ pXCH<sub>4</sub>\_LT data were similar to the estimates derived from the surface observations, but was underestimated by about 35 Tg CH<sub>4</sub> year<sup>-1</sup> (~6% of the global total) using the ~~JAXA/GOSAT~~-XCH<sub>4</sub> data. For the Southern Hemisphere, the estimates from the both GOSAT inversions were about 15–30 Tg CH<sub>4</sub> year<sup>-1</sup> higher than ~~that-those~~ derived from surface data. The evaluations against independent data from the Atmospheric Tomography Mission aircraft campaign showed good agreement

in the lower tropospheric CH<sub>4</sub> from the inversions using the ~~JAXA/GOSAT~~-pXCH<sub>4</sub>-LT and surface data. However, from these inversions, the modelled North-South gradients showed significant overestimation in the upper troposphere and stratosphere, possibly due to relatively uniform inter-hemispheric OH distributions that control CH<sub>4</sub> sinks. Overall, we found that the use of the JAXA/GOSAT pXCH<sub>4</sub>-LT data shows considerable potential in constraining global and regional CH<sub>4</sub> fluxes, advancing  
15 our understanding of the CH<sub>4</sub> budget.

## 1 Introduction

Methane (CH<sub>4</sub>) is the second most important greenhouse gas (GHG) after carbon dioxide (CO<sub>2</sub>) with a radiative forcing of ~~~0.650~~-0.565 W m<sup>-2</sup> (for 20222023; <https://www.esrl.noaa.gov/gmd/aggi/aggi.html>, last access: ~~13 July 2024~~15 April 2025). Global and regional CH<sub>4</sub> budgets have been estimated using various data sources and methods, with recent estimates of global  
20 total emissions at 575 (553–586) Tg CH<sub>4</sub> yr<sup>-1</sup> over the past decade based on top-down estimates (Saunois et al., 2024). While top-down inverse models provide well-constrained global total emissions using atmospheric measurements of surface CH<sub>4</sub> and satellite total columns, regional estimates still vary significantly depending on model setups and assimilated data (Deng et al., 2024; Stavert et al., 2021).

One important factor controlling inverse model estimates is the type of data assimilated in the inverse models. Broad categories of assimilable data are 1) high-precision in-situ observations from ground-based stations, shipboard and aircraft, and 2) column-averaged dry-air mole fractions of GHGs retrieved from satellites and ground-based stations. Over the years, the column-averaged dry-air mole fractions of CH<sub>4</sub> (XCH<sub>4</sub>) from various satellites, such as SCanning Imaging Absorption spectroMeter for Atmospheric Cartography (SCIAMACHY) on board ENVironmental SATellite (ENVISAT) (Bovensmann et al., 1999), Thermal And Near infrared Sensor for carbon Observations–Fourier Transform Spectrometer (TANSO–FTS) on board  
30 the Greenhouse Gases Observing Satellite (GOSAT) (Kuze et al., 2009), TANSO-FTS-2 on board GOSAT-2 (Suto et al., 2021), TROPospheric Monitoring Instrument (TROPOMI) on board the Sentinel 5 Precursor (Hu et al., 2018), have been available and used in estimation of global and regional CH<sub>4</sub> fluxes (e.g. Alexe et al., 2015; Baray et al., 2021; Chen et al., 2022; Houweling et al., 2014; Lu et al., 2021; Miller et al., 2013; Lunt et al., 2019; Pandey et al., 2016; Wang et al., 2022; Tsuruta et al., 2023; Qu et al., 2021).

35 Due to their spatial coverage, satellite retrievals have shown high potential in estimating GHG budgets for regions with sparse surface observations, such as the tropics (Alexe et al., 2015; Houweling et al., 2014; Qu et al., 2021), central Africa (Lunt et al., 2019; Pandey et al., 2021) and China (Chen et al., 2022; Lu et al., 2021; Wang et al., 2022). However, it has been challenging to accurately model and retrieve vertical profiles of CH<sub>4</sub> concentrations, resulting in discrepancies between XCH<sub>4</sub> estimates from transport models and satellite retrievals. For transport models, key factors influencing the estimation  
40 of XCH<sub>4</sub> include model resolution (both horizontal and vertical), estimates of tropopause height and the representation of atmospheric chemical reactions with oxidants. The latter is significant since CH<sub>4</sub> is mostly oxidized by OH (Zhao et al., 2020). For satellite retrievals, prior profiles, clouds and aerosols, surface albedo and retrieval methods contribute to the uncertainty of retrieved XCH<sub>4</sub> values (Lindqvist et al., 2024; Sha et al., 2021). These factors contribute to large-scale latitudinal and

seasonal discrepancies between the satellite retrievals and transport model estimates using prior or posterior emissions derived  
45 by inversion estimates assimilating surface data.

Without addressing this issue, it could lead to unrealistic emission estimates from the inversions using satellite data that are significantly different from the estimates using surface observations. Previously in Tsuruta et al. (2023), we showed that the inversion using TROPOMI data without large-scale corrections could lead to smaller CH<sub>4</sub> emission estimates over the high northern latitudes compared to the inversion estimates based on surface observations. Various approaches have been  
50 developed to manipulate the large-scale discrepancies, which include: adjusting large-scale discrepancies before performing ~~satellite~~-satellite-based inversions (Houweling et al., 2017; Wang et al., 2022; Zhang et al., 2021), using the so-called proxy method that optimizes the CO<sub>2</sub>:CH<sub>4</sub> ratios based on GOSAT data that provide both XCO<sub>2</sub> and XCH<sub>4</sub> retrievals (Feng et al., 2017; Palmer et al., 2021; Pandey et al., 2016), or discarding high-latitude data, where the problem appears most severe (Alexe et al., 2015; Baray et al., 2021; Lu et al., 2022). Apart from the proxy method, these adjustments have been somewhat arbitrary,  
55 with the degree of adjustments varying between studies. With appropriate manipulations, results from ~~inversion~~-inversions using surface and satellite data seem to agree in general, while satellite data can also provide additional regional information about magnitude and seasonality of CH<sub>4</sub> emissions (e.g. Wang et al., 2022; Lu et al., 2021; Pandey et al., 2021; Lunt et al., 2019; Feng et al., 2017).

The TANSO-FTS has measured reflected sunlight with two orthogonal components of polarization in the short wave infrared (SWIR) and ~~thermal~~ emissions in the thermal infrared (TIR) simultaneously at the local time of 13:00. SWIR data constrain  
60 the total column density and TIR data provide vertical profile information. Recently, the Japan Aerospace Exploration Agency (JAXA) has developed a new retrieval product of the partial column CO<sub>2</sub> and CH<sub>4</sub> densities ~~in~~-of the lower troposphere (LT, typically 0–4 km), upper troposphere (UT, typically 4–12 km) and stratosphere from the SWIR and TIR by minimizing contamination by highly-polarized radiation scattered by aerosols and thin clouds (Kikuchi et al., 2016; Kuze et al., 2022).  
65 Compared to total column retrievals, this method offers the advantage that lower and upper tropospheric products contain more information about surface fluxes, making them particularly useful for detecting local CH<sub>4</sub> (Kuze et al., 2020) and CO<sub>2</sub> (Kuze et al., 2022) fluxes. Atmospheric transport models generally perform well representing the in lower troposphere and combined with inverse models, they can reproduce atmospheric CH<sub>4</sub> surface observations reasonably well. Therefore, the use of tropospheric partial column data may provide better constrains for global and regional CH<sub>4</sub> flux estimates than using total  
70 column data.

In this study, we present for the first time a way to assimilate JAXA/GOSAT lower tropospheric partial column CH<sub>4</sub> (pXCH<sub>4</sub>\_LT) data into the atmospheric inverse model CarbonTracker Europe-CH<sub>4</sub> (CTE-CH<sub>4</sub>; Tsuruta et al., 2017). We examined the global CH<sub>4</sub> fluxes for 2016–2019 derived from the pXCH<sub>4</sub>\_LT data and compare those to the flux estimates derived from JAXA/GOSAT XCH<sub>4</sub> data and surface CH<sub>4</sub> observations. We evaluated annual budgets, seasonal cycles and spa-  
75 tial distributions of the total and subcategory emissions (anthropogenic and wetlands). Additionally, we compared optimized atmospheric CH<sub>4</sub> to independent (i.e. not assimilated) data from the Atmospheric Tomography Mission (ATom) aircraft campaign and total and lower tropospheric partial column data from Total Carbon Column Observing Network (TCCON). The

study highlights the potential of JAXA/GOSAT pXCH<sub>4</sub>\_LT data to improve the constraints on global and regional CH<sub>4</sub> fluxes compared to total column data.

## 80 2 Method

### 2.1 CTE-CH<sub>4</sub>

CarbonTracker Europe-CH<sub>4</sub> (CTE-CH<sub>4</sub>; Tsuruta et al., 2017) is a modular atmospheric inverse modelling system (van der Laan-Luijkx et al., 2017) based on the ensemble Kalman filter (EnKF) (Peters et al., 2005). It minimizes the cost function  $J$ ,

$$J = (\mathbf{x} - \mathbf{x}^b)\mathbf{P}^{-1}(\mathbf{x} - \mathbf{x}^b) + (\mathbf{y} - \mathcal{H}(\mathbf{x}^b))\mathbf{R}^{-1}(\mathbf{y} - \mathcal{H}(\mathbf{x}^b)),$$

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$$J = (\mathbf{x} - \mathbf{x}^b)\mathbf{P}^{-1}(\mathbf{x} - \mathbf{x}^b) + (\mathbf{y} - \mathcal{H}(\mathbf{x}))\mathbf{R}^{-1}(\mathbf{y} - \mathcal{H}(\mathbf{x})), \quad (1)$$

where  $\mathbf{x}$  is the state vector,  $\mathbf{x}^b$  is the prior state vector,  $\mathbf{P}$  is the state covariance matrix,  $\mathbf{y}$  is the observation of atmospheric concentrations (see Section 2.3),  $\mathcal{H}$  is the observation operator, and  $\mathbf{R}$  is the observation covariance matrix. In this study, the state vector  $\mathbf{x}$  included the flux multiplication factors for anthropogenic and wetland fluxes (see Section 2.2). Fluxes were optimized at  $1^\circ \times 1^\circ$  (latitude  $\times$  longitude) resolution for land areas in northern Eurasia,  $2^\circ \times 3^\circ$  grid for other land areas, and region-wise over the ocean (Fig. A1). Note that we do not optimize natural ocean fluxes, but do optimize anthropogenic emissions over the oceans, such as shipping and flight tracks that were included in the prior fluxes (EDGAR v8.0, Section 2.2). The prior covariance  $P$  was a block diagonal matrix, assuming that  $1^\circ \times 1^\circ$  optimization regions were uncorrelated with  $2^\circ \times 3^\circ$  optimization regions, land and ocean regions were uncorrelated and wetland fluxes were uncorrelated with anthropogenic fluxes. The prior uncertainty (diagonal values) were defined as ratio to prior fluxes (Section 2.2): 80% over land and 20% over the ocean. Off-diagonals were defined based on distances between the grids and regions with spatial correlation lengths of 100 km for  $1^\circ \times 1^\circ$  optimization regions, 300 km over  $2^\circ \times 3^\circ$  optimization regions and 900 km over the ocean. Localization schemes as in Peters et al. (2007) were applied. Regarding the EnKF setups, we used an ensemble size of 500 members and optimization window of 7-days with lag of 5 following Tsuruta et al. (2017).

100 For the observation operator  $\mathcal{H}$ , the atmospheric transport model TM5 (Krol et al., 2005) was used. TM5 was run at  $1^\circ \times 1^\circ$  over Europe and  $6^\circ \times 4^\circ$  globally, following e.g. Thompson et al. (2021) and Tenkanen et al. (2025). The resolution is coarser than the flux optimization resolution outside Europe. We acknowledge that using different resolutions can be questionable because the atmospheric states may not be resolved in detail enough. However, this could be justified as the study mostly focus on large-scale fluxes. TM5 was constrained by 3-hourly European Centre for Medium-Range Weather Forecasts (ECMWF) ERA5 meteorology (Hersbach et al., 2020). Atmospheric chemistry included OH, Cl and O(<sup>1</sup>D). The OH concentrations were same as in Houweling et al. (2014), where which is based on Spivakovsky et al. (2000) distribution but scaled globally by 0.92. For reactions with Cl and O(<sup>1</sup>D), the reaction rates were calculated pre-calculated from the ECHAM/MESSy1 model (Jöckel et al., 2006) were used. The initial CH<sub>4</sub> concentration fields were taken from Tenkanen et al. (2024) Tenkanen et al. (2025).



## 2.2 Prior fluxes

110 For anthropogenic emissions, prior estimates were taken from the Emissions Database for Global Atmospheric Research (EDGAR) v8.0 (Crippa et al., 2023). For fluxes from wetlands and dry mineral soils (hereafter wetlands), the estimates from the LPX-Bern v1.4 process based ecosystem model (Lienert and Joos, 2018) were used with 2019 values replicated from 2018. Other sources include biomass burning, microbial (termites) and ocean emissions, and the estimates from Global Fire Emissions Database (GFED) v4.1s (van der Werf et al., 2017), the Vegetation Integrative Simulator for Trace gases (VISIT; Ito  
115 and Inatomi, 2012) and Saunio et al. (2020) were used, respectively. Among those, the fluxes from anthropogenic, wetlands and biomass burning were monthly and inter-annually varying. The emissions from termites were annual estimates, and ocean fluxes were climatological.

## 2.3 Atmospheric observations

### 2.3.1 JAXA/GOSAT partial column data

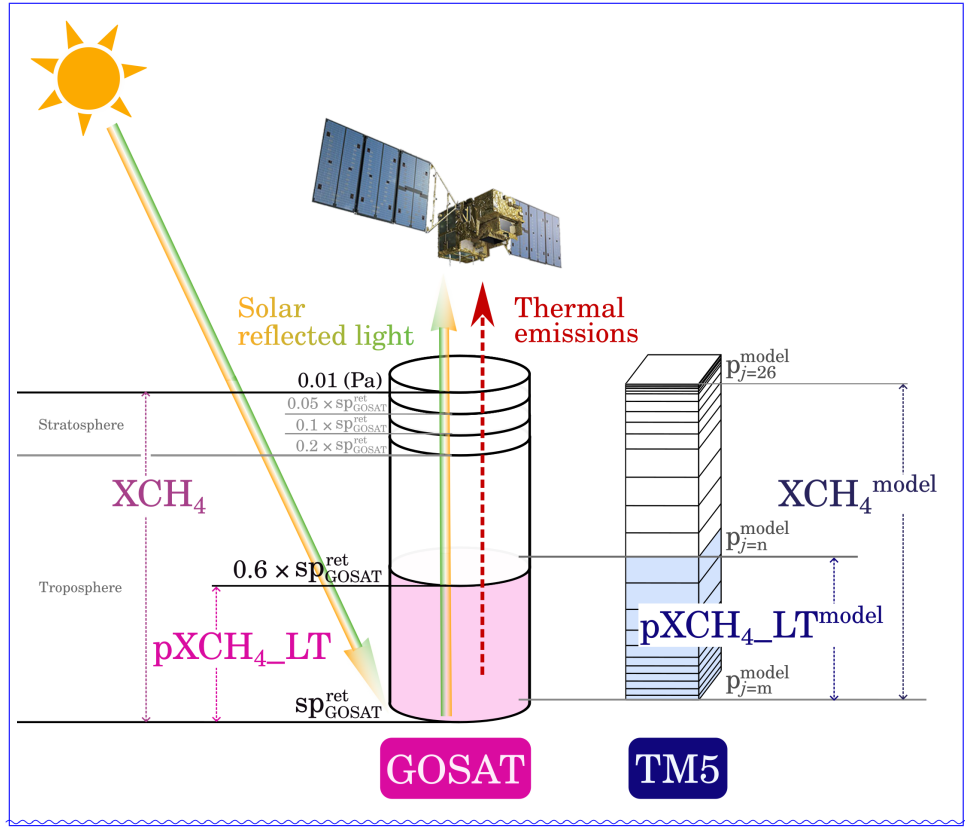
120 ~~The~~ JAXA/GOSAT retrieval algorithm is based on ~~the~~ Full Physics algorithm ~~extended-simultaneous-use-both-and-is extended to use both the~~ 2-orthogonal SWIR ~~signal-and-TIR. The algorithm is based on Kikuchi et al. (2017) and TIR signals simultaneously (Kikuchi et al., 2017).~~ JAXA's forward calculation constructs the vector radiance and ~~use-uses the~~ 2-orthogonal polarized SWIR observation in four windows with bi-directional reflection. For TIR, the scalar radiance is handled in ~~the~~ forward model for three windows. The ~~an~~-empirical orthogonal function (EOF) fitting ~~are-is~~ taken account in ~~retrieval-process. In~~ the retrieval  
125 process, ~~where~~ XCO<sub>2</sub>, XCH<sub>4</sub> ~~and~~ XH<sub>2</sub>O are simultaneously retrieved with solar-induced chlorophyll fluorescence (SIF) information. ~~Two layers in the tropospheric and three layers in the stratospheric~~ CO<sub>2</sub> and CH<sub>4</sub> partial column-averaged concentrations are derived ~~.For for two layers in the troposphere and three layers in the stratosphere. The~~ H<sub>2</sub>O ~~concentrations are derived on 11 layers of vertical concentration are derived~~ vertical layers.

The five ~~CO<sub>2</sub> and CH<sub>4</sub>~~ layers are defined by ~~the~~ pressure levels based on the retrieved surface pressure, denoted as  $sp_{GOSAT}^{ret}$   
130 ~~.The lower~~ (see also Fig. 1). ~~The two tropospheric layers, lower troposphere (LT) and upper troposphere,~~ are defined as ~~the~~ pressure levels between  $sp_{GOSAT}^{ret}$  and  $0.6 \times sp_{GOSAT}^{ret}$ , and between  $0.6 \times sp_{GOSAT}^{ret}$  and  $0.2 \times sp_{GOSAT}^{ret}$  ~~pressure levels~~, respectively. ~~For~~ ~~The~~ three stratospheric layers ~~are defined as the pressure levels~~ between  $0.2 \times sp_{GOSAT}^{ret}$  and  $0.1 \times sp_{GOSAT}^{ret}$ , between  $0.1 \times sp_{GOSAT}^{ret}$  and  $0.05 \times sp_{GOSAT}^{ret}$ , and between  $0.05 \times sp_{GOSAT}^{ret}$  and  $0.01$  ~~are defined as lower stratosphere, middle stratosphere, and upper stratosphere~~ Pa, respectively. In this study, all the GOSAT data is based on JAXA/GOSAT product (~~v2.0~~) ([https://www.eorc.jaxa.jp/GOSAT/Global\\_GHGs\\_Map/index.html](https://www.eorc.jaxa.jp/GOSAT/Global_GHGs_Map/index.html), last access: 4 June 2024).  
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During the study period, ~~the~~ TANSO-FTS and ~~the~~ Cloud and Aerosol Imager (CAI) were shut down and the observation had been suspended during 17–24 May 2018 due to the Command and Data Management System (CDMS) incident, and from 24 November 2018 until 28 December 2018 due to the rotation anomaly of the second solar-paddle.

For comparison to model estimates, the XCH<sub>4</sub> values from model results were calculated as:

$$140 \quad XCH_4^{model} = \frac{\sum_i (CH_{4i} \times dp_i)}{\sum_i dp_i}, \quad i = 1, \dots, 25 \quad (2)$$



**Figure 1.** A schematic figure illustrating how total column ( $XCH_4$ ) and lower tropospheric partial column ( $pXCH_4\_LT$ ) are calculated in the JAXA/GOSAT data and from TM5. JAXA/GOSAT retrieval algorithm uses information from both solar reflected light and thermal emissions to retrieve partial column  $CH_4$  mole fractions. In JAXA/GOSAT there are five layers between the retrieved pressure ( $sp_{GOSAT}^{ret}$ ) and 0.01 Pa, with two tropospheric and three stratospheric layers. The lower troposphere (LT) is defined as pressure levels between  $sp_{GOSAT}^{ret}$  and  $0.6 \times sp_{GOSAT}^{ret}$ . In TM5, there are 25 model layers and  $XCH_4^{model}$  is calculated using all layers, i.e. the pressure levels between  $p_{j=0}^{model}$  and  $p_{j=26}^{model}$ . For calculation of  $pXCH_4\_LT^{model}$ , the minimum level (m) and maximum level (n) vary depending on  $sp_{GOSAT}^{ret}$ .  $m = 0$  if  $sp_{GOSAT}^{ret} > p_{j=0}^{model}$  and otherwise the maximum level at which  $p_j^{model}$  exceeds  $sp_{GOSAT}^{ret}$ . n is the level at which  $p_j^{model}$  reaches  $0.6 \times sp_{GOSAT}^{ret}$ .

where  $CH_{4,i}$  is the dry air mixing ratio of  $CH_4$  at TM5 model layer  $i$ , level  $i$  \* temporally and horizontally interpolated to time and location of the GOSAT/JAXA/ \* data, and  $dp_i$  is the pressure thickness at the layer  $i$ , level  $i$  \* For calculating modelled partial lower tropospheric \* columns of methane,  $pXCH_4\_LT^{model}$ , the layers were selected based on  $sp_{GOSAT}^{ret}$  (see also Fig. 1). The minimum layer is  $i = 0$  if the surface pressure in TM5 levels were selected such that the minimum level  $i$  corresponded to the index where model layer is smaller than  $sp_{GOSAT}^{ret}$  and otherwise the maximum layer at which TM5 model pressure exceeds the GOSAT-retrieved surface pressure, and the maximum level \* to the point where TM5 pressure reached  $0.6 \times sp_{GOSAT}^{ret}$ . Vertical interpolation was not applied, leading for simplicity, and therefore, modelled mixing ratio was calculated using thinner (in case  $sp_{GOSAT}^{ret} > sp_{GOSAT}^{model}$ ) or thicker (in case  $sp_{GOSAT}^{ret} < sp_{GOSAT}^{model}$ ) air mass from the lowest layer. For the uppermost layer, due to the

selection method, the model nearly always contained thicker air mass than the retrievals. These likely lead to potential biases  
150 (see also Section 4.1).

Averaging kernels were not applied to ~~model-XCH<sub>4</sub>~~<sup>model</sup>-XCH<sub>4</sub><sup>model</sup> and pXCH<sub>4</sub>\_LT<sup>model</sup>, as ~~it was they were~~ unavailable for the v2.0 product. However, we are aware of a newer version, v3.0 data, where averaging kernels are now available. We did not apply any preprocessing of the JAXA/GOSAT data, such as averaging or removing large-scale differences that may have been raised in comparison to inversion estimates assimilating surface data. This way, we ~~can~~<sup>could</sup> examine the effect of observations  
155 directly. In the assimilation, we assumed observational uncertainty (retrieval error + transport model error) to be 1) 30 ppb globally with a rejection threshold of 60 ppb ~~in both cases assimilating for both of the~~ XCH<sub>4</sub> or pXCH<sub>4</sub>\_LT ~~assimilations~~, and 2) 50 ppb with a rejection threshold of 100 ppb ~~and assimilating in the case that assimilated~~ only pXCH<sub>4</sub>\_LT data over land (see also 2.4). ~~The rejection thresholds discriminates the observations if differences between observed and prior-modelled mole fractions exceed the threshold, i.e. the observations are rejected and would not be used to constrain the fluxes.~~ These values  
160 are somewhat arbitrary, but ~~following they follow~~ other inversion experiments ~~using that use~~ the GOSAT data (e.g. Janardanan et al., 2020; Lu et al., 2021; Maasakkers et al., 2021; McNorton et al., 2018). The larger uncertainty is ~~examined~~<sup>justified</sup> as retrieval errors in the lower tropospheric partial column data are probably higher ~~compare~~<sup>compared</sup> to the total column data.

### 2.3.2 Surface CH<sub>4</sub> mole fractions

Surface CH<sub>4</sub> mole fraction observations mainly from ObsPack v4.0 (Schuldt et al., 2021) were assimilated in the inversion, as  
165 well as used for evaluation (see Fig. A1 for site locations and Table A1 for details). The data include ICOS ATC CH<sub>4</sub> Release and ICOS ATC NRT CH<sub>4</sub> growing time series data, which were downloaded among with other NOAA ObsPack data and the ICOS Carbon portal (Table A2). The data consisted of discrete and continuous observations from in-situ stations and ships. Similar to our previous studies (~~Tsuruta et al., 2017, 2019; Tenkanen et al., 2024~~)(Tsuruta et al., 2017, 2019; Tenkanen et al., 2025), all data were filtered by taking observations at well-mixed conditions based on quality flags given by the data providers. Continuous data were processed into daily means by averaging observations during local time afternoon (12–16) or night (0–4).  
170 Observational uncertainties (observational error + transport model error) ranged between 4.5–75 ppb, depending on each site (Table A1).

In addition, we used aircraft measurements from the Atmospheric Tomography Mission (ATom; Thompson et al., 2022), obtained from the ObsPack v6.0 (Schuldt et al., 2023) for independent evaluation. During 2016–2019, there were ATom obser-  
175 vations from four campaigns: 2016 July–August, 2017 January–February, September–October and 2018 April–May. Prior and posterior mole fractions were estimated at each sampling location and time, linearly interpolated within the TM5 model grid cells. ATom data were not assimilated in the inversions and therefore can be used as independent observations for validation.

### 2.3.3 TCCON

The Total Carbon Column Observing Network (TCCON) is a global network providing XCH<sub>4</sub> measurements retrieved from  
180 the spectrum of near-infrared radiation of direct sunlight using ground-based Fourier Transform Spectrometers (FTS) (Wunch et al., 2011). We used the GGG2020 data (Laughner et al., 2023, 2024) from 25 sites globally (Table A3) for evaluation of

inversion results. The sites were selected as those that provided GGG2020 data, and have at least one year of measurements between 2016–2019. The data were not assimilated in the inversions and so can also be used as independent observations for validation.

185 For comparison, hourly average mixing ratio interpolated horizontally to the TCCON locations were used. Temporal co-locations were done by selecting the TCCON observations that were closest to the model time step (hourly) and set the time limit of half an hour; if there was a TCCON observation made within  $\pm$  half an hour of the model time step, the TCCON and modelled values were taken into account.

For comparison to total column ( $XCH_4$ ), the model estimates were calculated by applying TCCON averaging kernels  
190 (Rodgers and Connor, 2003):

$$\hat{c} = c_a + (\mathbf{h} \circ \mathbf{a})^T (\mathbf{x} - \mathbf{x}_a) \quad (3)$$

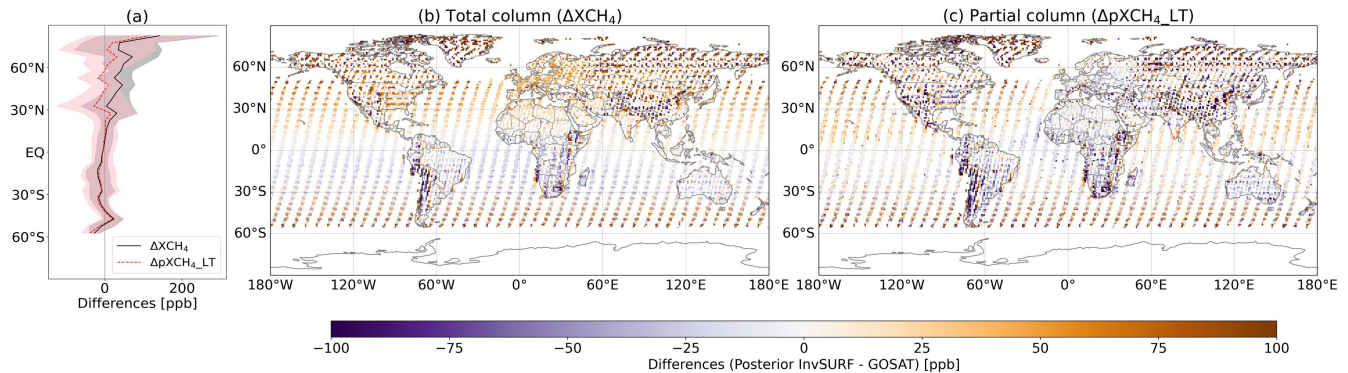
where  $\hat{c}$  is the averaging kernel corrected  $XCH_4$  value from the model,  $c_a$  is the TCCON prior  $XCH_4$ ,  $\mathbf{h}$  is the TCCON pressure weighting function,  $\mathbf{a}$  is the TCCON averaging kernel,  $\mathbf{x}$  is the model profile and  $\mathbf{x}_a$  is the TCCON prior profile. After applying the averaging kernel, daily means were calculated for evaluation.

195 In addition, the tropospheric partial column were calculated from TCCON total columns of  $CH_4$  and hydrogen fluoride (HF). Practically all of the HF exists in the stratosphere, where HF is produced from photodissociation of chlorofluorocarbons (CFCs). The concentrations of long lived tracers, such as  $CH_4$  and HF, are strongly correlated in the stratosphere (e.g. Plumb, 2007). Airmasses containing both  $CH_4$  and CFCs enter through the topical stratosphere, where  $CH_4$  is oxidized and HF is produced. In the stratosphere,  $CH_4$  shows a nearly linear inverse relationship with HF. By assuming a linear relationship in  
200 the stratosphere (Washenfelter et al., 2003; Saad et al., 2014) the ~~stratospheric~~-tropospheric partial column of  $XCH_4^{\text{tropo}}_{\text{TCCON}}$  is given as

$$XCH_4^{\text{tropo}}_{\text{TCCON}} = XCH_4 - \beta \times XHF \quad (4)$$

where  $\beta$  is the stratospheric  $CH_4$ :HF slope. As the tracer to tracer correlations typically exhibit distinct correlations in the tropics, extratropics and polar stratospheric vortices (Plumb, 2007), we derived slopes for the tropics ( $30^\circ$  S– $30^\circ$  N), SH ( $90^\circ$   
205 S– $30^\circ$  S), NH ( $30^\circ$  N– $90^\circ$  N), as well as for the polar stratospheric vortices. Profile data of  $CH_4$  and HF from the Atmospheric Chemistry Experiment - Fourier Transform Spectrometer (ACE-FTS) version 4.1 data products (Boone et al., 2020) were used to calculate the slopes. The polar vortices were identified from potential vorticity data from the ERA5 reanalysis (Hersbach et al., 2020). The vortex edge was assumed to be represented by the  $|36|$  PVU isoline. The interference of water can increase the error in the column averaged HF (XHF) (Saad et al., 2014), especially at high airmasses. Therefore, observations with  
210 column averaged  $H_2O$  ( $xH_2O$ ) above 2000 ppm and zenith angle larger than  $90 - 0.0075 \times xH_2O$  degrees were discarded.

For comparison to the tropospheric partial column, Eq. 2 was applied similarly to calculate  $pXCH_4_{LT}^{\text{model}}$ , but for the lowest and uppermost layer, vertical interpolation was applied to TCCON retrieval surface pressure  $sp_{\text{TCCON}}^{\text{ret}}$  and tropopause height, respectively. The pressure at dynamic tropopause was calculated based on ERA5 reanalysis data, defined by the  $|2|$  PVU isoline. Constant extrapolation was applied in case  $sp_{\text{TCCON}}^{\text{ret}} > sp^{\text{model}}$ .



**Figure 2.** Mean differences between posterior InvSURF and JAXA/GOSAT retrievals, averaged over 2016–2019. (a) five degree latitudinal means (solid line) with standard deviation (shaded area), and (b) and (c)  $1^\circ \times 1^\circ$  grid means. Positive values indicate posterior InvSURF mole fractions being higher than the JAXA/GOSAT retrievals.

## 2.4 Simulation setups

In this study, we present the results from four inversions that differed in the observations assimilated:

InvSURF: surface  $\text{CH}_4$  data only

InvGLT: JAXA/GOSAT partial column data ( $\text{pXCH}_4\text{_{LT}}$ ) only

InvGLT\_land: JAXA/GOSAT partial column data ( $\text{pXCH}_4\text{_{LT}}$ ) over land only

InvGTOT: JAXA/GOSAT total column data ( $\text{XCH}_4$ ) only

In all of the GOSAT inversions, only the JAXA/GOSAT data were assimilated to examine the effect of the data in constraining fluxes independent of other datasets. The inversion without ocean data were tested in consideration of assimilating other satellite data, such as TROPOMI, which do not necessary provide ocean data from all retrieval products. Additionally, considering relatively small contribution of ocean fluxes to global total, compared to that of e.g.  $\text{CO}_2$ , we examine if land fluxes would be constrained equally well without ocean data.

For InvGLT and InvGTOT, observational uncertainty and rejection threshold was set to be 30 and 60 ppb, respectively, and for InvGLT\_land they were set as 50 and 100 ppb, respectively. All inversions were run for 2015–2019, but 2015 was considered as a spin-up, and not included in the analysis. The analysis on regional fluxes was done based on  $30^\circ$  latitudinal bands. Throughout the following sections, the prior and posterior mole fractions refer to those derived using prior and posterior fluxes, respectively.

### 3 Results

#### 3.1 Comparison of CH<sub>4</sub> mole fractions from InvSURF and JAXA/GOSAT

In this section, we analyse the differences between posterior estimates from the surface-based inversion (InvSURF) and JAXA/GOSAT retrievals for total and partial columns, i.e.

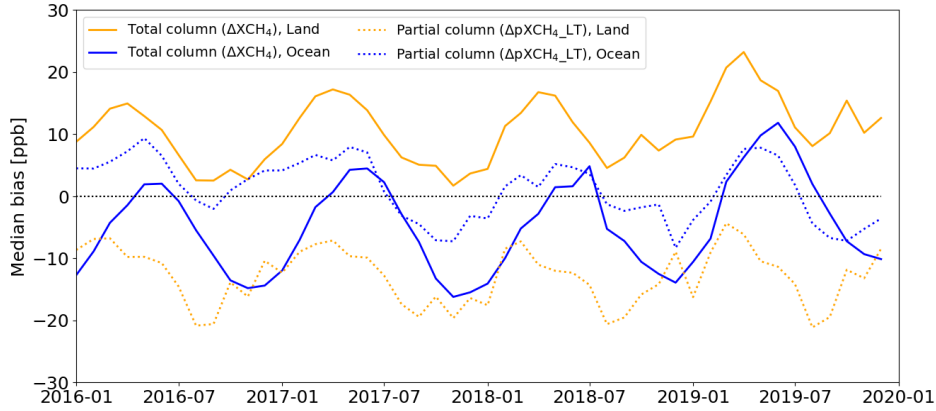
$$\begin{aligned} \Delta XCH_4 &= XCH_4^{\text{post}}_{\text{InvSURF}} - XCH_4^{\text{ret}}_{\text{GOSAT}} \\ 235 \quad \Delta pXCH_4\text{--}LT &= pXCH_4\text{--}LT^{\text{post}}_{\text{InvSURF}} - pXCH_4\text{--}LT^{\text{ret}}_{\text{GOSAT}} \end{aligned} \quad (5)$$

In the comparison of total column, latitudinal biases were found with positive  $\Delta XCH_4$  values in the extratropics and negative values in the tropics, especially in the SH tropics (Fig. 2 (b)). This feature was systematic in time, such that similar biases were found regardless of years and seasons, although the absolute value of the biases varied. In the comparison of lower tropospheric partial columns, such latitudinal biases were less prominent (Fig. 2 (c)), especially over land, indicating better agreement over  
240 NH extratropics (Fig. 2 (a)). This indicates the potential role of upper atmosphere in the total column biases, where posterior estimates and retrievals had difficulty agreeing.

Large biases were observed in regions such as Greenland, western South America, southernmost South Africa, eastern China and northern Russia in both comparisons (Fig. 2 (b) and (c)). Because of the challenges in retrieving data over ice-covered land, we assume that biases in Greenland were mostly associated with retrieval errors. Biases in other regions could be  
245 due to unresolved fluxes by surface observations, i.e. the inversion error in estimating fluxes, retrieval errors due to cloud cover and difficulties in retrieving surface pressure in regions with highly elevated surface. The horizontal resolutions of the transport model also contribute to the biases in highly elevated areas. The additional simulation with TM5 showed that increasing resolution from  $4^\circ \times 6^\circ$  (latitude  $\times$  longitude) to  $2^\circ \times 3^\circ$  decreased biases in mountain regions in Africa and Tibetan plateau, although the biases in regions such as Andes mountains in South America and edges of Tibetan plateau still remain (Fig. A2).  
250 We also acknowledge that the vertical interpolation and averaging kernel (AK) contribute to the biases in highly elevated areas, especially for  $XCH_4$  (Fig. A3) (see also Section 4.1).

Europe showed the smallest  $\Delta pXCH_4\text{--}LT$ , which is encouraging considering that InvSURF probably had constrained the emission in Europe the best compared to other regions globally, and modelled tropospheric CH<sub>4</sub> from InvSURF is well in line with ground-based observations (Section 3.2). Both the transport model and optimization resolutions were the smallest  
255 and number of observations were relatively large in Europe (Section 2.1). There was a notable shift between better agreement in Europe and a worse agreement in Russia and Africa, attributed to the CTE-CH<sub>4</sub> setup: TM5 was run with highest spatial resolution over Europe and an extended area (Tsuruta et al., 2015), with a coarser resolution elsewhere. This resulted to creating an artifact of a border between the zoom grid and the global grid that caused the shift. An additional simulation with TM5 with global  $2^\circ \times 3^\circ$  (latitude  $\times$  longitude) resolution and without zoom, eliminated these boundaries (Fig. A2).

260 Both lower tropospheric partial and total column comparisons showed seasonal and land-sea biases (Fig. 3). The global average of  $\Delta XCH_4$  and  $\Delta pXCH_4\text{--}LT$  were smaller during NH ~~winter-than-summer~~ autumn-winter than spring-summer. The amplitude of the seasonal biases was larger in the total column, especially over the oceans. The comparison of the land and



**Figure 3.** Global monthly median differences between posterior InvSURF and JAXA/GOSAT retrievals. Positive values indicate posterior InvSURF mole fractions being higher than the JAXA/GOSAT retrievals.

sea biases ~~it~~ showed that the biases in the total column were larger over land than ocean, whereas an opposite behaviour was observed for the lower tropospheric partial columns. In other words, the  $\Delta XCH_4$  and  $\Delta pXCH_{4\_LT}$  were closer over   
265 the ocean than land on average, with absolute median monthly differences of 5 ppb over the ocean and 23 ppb over land, indicating the possible influence of land fluxes to upper atmosphere. Overall, the average bias during 2016–2019 was smallest in  $\Delta pXCH_{4\_LT}$  over the ocean.

### 3.2 Evaluation against surface and aircraft data



Comparison of posterior atmospheric CH<sub>4</sub> to the surface ground-based observations showed the smallest overall bias, root mean squared error (RMSE) and strongest correlation for InvSURF (Table 1) as expected, since these observations were assimilated in the inversion. Among ~~the~~ GOSAT inversions, those using ~~lower-tropospheric~~ the lower tropospheric partial column data (InvGLT and InvGLT\_land) showed the best agreement to the surface ground-based observations, while the total column inversion (InvGTOT) showed large negative biases following the prior (Table 1). RMSE and correlation of the GOSAT inversions were not significantly different. The latitudinal gradient was best captured by InvSURF, InvGLT and InvGLT\_land compared to the surface stations (Fig. 4a). The inversions mostly underestimated the surface observations with the underestimation being the smallest in the Southern Hemisphere (SH) for InvGLT and InvGLT\_land and in the Northern Hemisphere (NH) for InvSURF. InvGTOT showed better agreement than InvSURF in the SH, but considerable underestimation in the NH (Fig. 4a), which was the reason for strong negative bias in the overall agreement (Table 1).

In the mid- and high northern latitudes (above 30° N) the seasonal cycle amplitude (SCA) was generally overestimated by model estimates in prior, which was worsened by inversion (SCA of posterior estimates were larger than prior) (Fig. A4). In addition, seasonal minima in the NH ~~occured~~ occurred one to two months later than observations, although the results in InvSURF were slightly better than in the prior and GOSAT inversions (Fig. A4). This indicates either possible errors in seasonal cycles of posterior emission, or atmospheric chemical sinks.

Comparison to AToM aircraft data also showed that on average, InvSURF, InvGLT and InvGLT\_land resulted in the best statistics (Table 1), and the latitudinal gradient agreed better with the observations compared to InvGTOT ~~below 4000 meter at~~ the altitude bands 0–2000 and 2000–4000 meters above sea level (masl) (Fig. 4b, A5 and A6). In ~~this~~ these altitude zones, the mean bias was significantly larger in InvGTOT, which was in line with the results compared to surface ground-based stations (Table 1). This height corresponds approximately to the height where most of the surface ground-based stations were situated and from which JAXA/GOSAT XCH<sub>4</sub>\_LT data were calculated. Between 4000–8000 masl, the latitudinal gradients from InvGTOT were better captured whereas the other inversions overestimated these gradients (Fig. ~~A5 and~~ A6). Considering that the tropopause height is around 9000 masl or above, these results indicate that the transport model has problems in representing upper tropospheric concentrations. This is consistent with the finding that all model estimates worsened at high altitudes (> 8000 masl). All model estimates, both prior and posterior, failed to capture low mole fractions observed in high latitude (> 50° N/S) regions (Fig. A5). Such low CH<sub>4</sub> mole fractions were observed especially in the winter of 2017 and the spring of 2018 when the tropopause height was lower and the AToM aircraft operated in the stratosphere (Fig. A7). This is when the disagreement was strongest, possibly indicating the improper modelling of vertical profiles in polar vortex conditions.

### 3.3 Evaluation against TCCON

The latitudinal gradient was generally weaker in TCCON tropospheric partial and total columns (Fig. 4) compared to the surface observations, indicating a smaller influence of surface fluxes. Therefore, the evaluation against the TCCON data provides limited information about how well the inversions constrained the surface fluxes but focuses more on the model performance regarding long-range transport, tropopause mixing and atmospheric chemistry, which are important to take into account when

**Table 1.** Bias, root mean squared error (RSME) and Pearson’s correlation against observations at surface ground-based stations assimilated in InvSURF, ATom aircraft measurements, and TCCON data. The statistics were calculated for each ground-based and TCCON station over 2016–2019 and four ATom campaigns separately, and the mean of all stations or campaigns are shown. The value followed by  $\pm$  sign is the standard deviation (std) of statistics from all stations. For ATom, the minima and maxima of the statistics are shown in square brackets instead of std due to the limited number of campaigns (four) to calculate std from. Biases were calculated as modelled values subtracted by observed values, and therefore, positive values indicate model overestimation. The simulation with the best statistics are highlighted in bold.

<i>Observations/Inversions</i>	Bias [ppb]	RMSE [ppb]	Correlation
<i>Surface ground-based</i>			
Prior	-42.2 $\pm$ 15.6	20.3 $\pm$ 11.1	0.69 $\pm$ 0.15
InvSURF	<b>-9.2 <math>\pm</math> 10.7</b>	<b>18.1 <math>\pm</math> 10.5</b>	<b>0.80 <math>\pm</math> 0.12</b>
InvGLT	-12.3 $\pm$ 13.6	20.8 $\pm$ 13.2	0.74 $\pm$ 0.15
InvGLT_land	-11.7 $\pm$ 13.8	20.6 $\pm$ 13.1	0.74 $\pm$ 0.15
InvGTOT	-36.9 $\pm$ 18.8	20.0 $\pm$ 12.1	0.74 $\pm$ 0.16
<i>ATom, &lt; 2000 m</i>			
Prior	-35.2 [-40.6, -31.1]	16.0 [11.0, 22.2]	0.97 [0.93, 0.98]
InvSURF	-13.4 [-20.8, -8.7]	<b>15.8 [12.2, 18.0]</b>	<b>0.97 [0.95, 0.98]</b>
InvGLT	-11.1 [-19.0, -5.1]	18.3 [14.3, 25.9]	0.95 [0.91, 0.97]
InvGLT_land	<b>-8.5 [-15.9, -4.0]</b>	19.9 [15.4, 26.3]	0.94 [0.90, 0.97]
InvGTOT	-28.6 [-37.4, -21.9]	22.0 [15.9, 34.9]	0.94 [0.87, 0.98]
<i>ATom, all</i>			
Prior	-22.8 [-34.3, -15.6]	<b>34.6 [17.3, 48.9]</b>	0.77 [0.63, 0.92]
InvSURF	3.3 [-11.2, 8.1]	35.2 [18.1, 49.9]	0.77 [0.63, 0.92]
InvGLT	<b>1.2 [-8.3, 13.6]</b>	34.7 [17.5, 48.1]	0.76 [0.64, 0.92]
InvGLT_land	3.5 [-5.4, 16.0]	<b>34.6 [18.8, 47.7]</b>	0.76 [0.65, 0.90]
InvGTOT	-13.8 [-26.0, -0.7]	34.9 [19.2, 45.8]	<b>0.76 [0.66, 0.92]</b>
<i>TCCON, partial column</i>			
Prior	-3.3 $\pm$ 16	15.4 $\pm$ 4.0	0.38 $\pm$ 0.20
InvSURF	23.4 $\pm$ 20.3	<b>13.6 <math>\pm</math> 4.2</b>	<b>0.68 <math>\pm</math> 0.15</b>
InvGLT	24.2 $\pm$ 15.8	15.8 $\pm$ 5.5	0.56 $\pm$ 0.21
InvGLT_land	25.4 $\pm$ 15.4	15.7 $\pm$ 5.4	0.56 $\pm$ 0.20
InvGTOT	<b>0.7 <math>\pm</math> 9.0</b>	14.7 $\pm$ 4.5	0.59 $\pm$ 0.19
<i>TCCON, total column</i>			
Prior	<b>3.2 <math>\pm</math> 15.3</b>	12.4 $\pm$ 7.9	0.39 $\pm$ 0.23
InvSURF	23.8 $\pm$ 20.4	<b>10.1 <math>\pm</math> 6.4</b>	<b>0.59 <math>\pm</math> 0.28</b>
InvGLT	25.2 $\pm$ 18.4	11.9 $\pm$ 8.8	0.50 $\pm$ 0.28
InvGLT_land	26.3 $\pm$ 18.3	11.8 $\pm$ 8.7	0.50 $\pm$ 0.27
InvGTOT	8.1 $\pm$ 10.8	11.2 $\pm$ 7.4	0.52 $\pm$ 0.28

assimilating total column satellite data. In the northern high latitudes, the extracted TCCON partial columns successfully separated the stratospheric component, showing no significant low biases under polar vortex conditions (Fig. A8).

305 Compared to the surface data comparison, the TCCON comparison showed better overall agreement and improved representation of the latitudinal gradient in InvGTOT compared to other inversions for both total and tropospheric partial columns (Table 1, Fig. 4c and 4d). In the NH, all model estimates exhibited overestimation, driven by larger latitudinal gradients, with InvSURF, InvGLT and InvGLT\_land performing worse than InvGTOT. Considering also that InvGTOT showed the best agreement with the comparison to Atom data between 4000–8000 masl on latitudinal gradients, this indicates the importance of the upper troposphere in the estimation of XCH<sub>4</sub>. This finding also denotes that model biases in the estimation of XCH<sub>4</sub> are not  
310 solely caused by errors in resolving stratospheric concentrations, but also the errors in upper troposphere contribute to total column biases.

### 3.4 CH<sub>4</sub> fluxes

**Table 2.** Global and regional CH<sub>4</sub> emissions (Tg CH<sub>4</sub> yr<sup>-1</sup>), averaged over 2016–2019. Uncertainty presented in numbers after ± sign is the standard deviation of 500 ensemble members in CTE-CH<sub>4</sub>.

Category / Region	Prior	InvSURF	InvGLT	InvGLT_land	InvGTOT
<i>Total</i>					
Global	523 ± 30	547 ± 26	550 ± 20	552 ± 21	544 ± 23
60° N–90° N	17 ± 1	20 ± 1	18 ± 1	17 ± 1	17 ± 1
30° N–60° N	179 ± 19	196 ± 14	191 ± 10	189 ± 10	154 ± 12
EQ–30° N	197 ± 18	200 ± 17	195 ± 13	199 ± 14	211 ± 15
30° S–EQ	115 ± 13	115 ± 12	129 ± 10	130 ± 10	146 ± 11
90° S–30° S	14 ± 2	15 ± 2	17 ± 2	17 ± 2	16 ± 2
<i>Anthropogenic</i>					
Global	356 ± 28	373 ± 67	382 ± 17	383 ± 18	368 ± 20
60° N–90° N	5 ± 1	6 ± 1	5 ± 1	5 ± 1	5 ± 1
30° N–60° N	150 ± 18	164 ± 14	161 ± 10	159 ± 10	125 ± 12
EQ–30° N	133 ± 16	136 ± 15	132 ± 11	135 ± 12	143 ± 13
30° S–EQ	57 ± 11	57 ± 10	70 ± 8	70 ± 8	82 ± 9
90° S–30° S	11 ± 2	12 ± 2	14 ± 2	14 ± 2	13 ± 2
<i>Wetlands</i>					
Global	120 ± 12	126 ± 11	121 ± 10	122 ± 10	130 ± 11
60° N–90° N	8 ± 1	10 ± 1	8 ± 1	8 ± 1	8 ± 1
30° N–60° N	21 ± 3	24 ± 2	22 ± 2	22 ± 2	22 ± 2
EQ–30° N	49 ± 7	49 ± 7	48 ± 6	49 ± 6	53 ± 7
30° S–EQ	40 ± 6	40 ± 6	41 ± 6	41 ± 6	45 ± 6
90° S–30° S	1 ± 1	1 ± 1	2 ± 1	2 ± 1	2 ± 1

The 2016–2019 average global total and regional (30° latitudinal band) emissions are presented in Table 2. The posterior global total emissions were similar in all inversions (544–552 ± 20–26 Tg CH<sub>4</sub> yr<sup>-1</sup>) and showed increases from the prior with reduction in uncertainty (523 ± 30 Tg CH<sub>4</sub> yr<sup>-1</sup>). Most of the increase from the prior was attributed to anthropogenic emissions (c.a. 57–96%). The sectorial emissions showed that anthropogenic emissions were higher and wetland emissions lower in InvGLT and InvGLT\_land compared to InvSURF and InvGTOT. The anthropogenic emissions in the SH in InvGLT and InvGLT\_land were higher compared to InvSURF but comparable in magnitude to InvSURF in the NH. InvGTOT also showed higher anthropogenic emissions in the SH compared to InvSURF, especially in the tropics (30° S–EQ), but lower anthropogenic emissions in 30° N–60° N.

Between the tropics and SH (90° S–30° N), the posterior anthropogenic emissions in the GOSAT inversions deviated more from the prior compared to InvSURF (Table 2, Fig. 5). This difference was primarily associated with emissions over South America, Africa and India (Fig. 6). The monthly variations of anthropogenic emissions deviated largely from the prior in the

GOSAT inversions (Fig. 5). All GOSAT inversion showed an emission peak in November in the region from the equator to 30° N, which was three months later than prior and InvSURF. In 30° S–EQ, all GOSAT inversions showed a clear seasonal cycle with emissions peaking during April–November. In contrast, the prior had no clear seasonal cycle, and InvSURF showed a small seasonal cycle with a peak in July. In 90° S–30° S, InvGLT and InvGLT\_land showed two emission peaks, one around April and another in October. InvGTOT did not show the April peak, but it aligned with InvGLT and InvGLT\_land in presenting an emission reduction in August and peak in October. InvSURF showed a less distinct seasonal cycle with emissions constantly increasing from the prior in all months, except January. In the tropics, there are only a few surface stations (Fig. A1), which are often far from emission sources, measuring background mixing air. As a result, the JAXA/GOSAT data likely contained more valuable information to constrain the fluxes in these regions compared to the sparse surface data.

In 30° N–60° N, posterior anthropogenic emissions in InvGTOT were about 35 Tg CH<sub>4</sub> year<sup>-1</sup> lower than those in the other inversions (Table 2), representing about 6% of the global total. These differences were not associated specifically with a specific region, but InvGTOT showed mostly negative emission increments indicating that posterior emissions were lower than the prior in this region (Fig. 6). Unlike the tropics, there are relatively large number of surface stations in Europe and the best optimization setup in terms of model spatial resolution, and thus, we suspect that the results from InvGTOT in Europe may have been too low. The comparison to ground-based observations in Europe also showed strong underestimation in InvGTOT (-44 ppb on average compared to stations within [12° W–37° E, 34° N–73° N] domain). For other regions, considering that the regions such as USA and China are large CH<sub>4</sub> emitters (Petrescu et al., 2024), we also suspect that the results in InvGTOT were underestimated, possibly due to the ability of the transport model representation of XCH<sub>4</sub> (see also Section 4.2).

In 60° N–90° N, posterior wetland emissions in the GOSAT inversions stayed close to the prior compared to InvSURF (Table 2). While InvSURF showed an increase in summer wetland emissions, such change was not found in the GOSAT inversions. Therefore, the seasonal cycle amplitude of wetland emissions was smaller in the GOSAT inversions (Fig. 5). It is known that the GOSAT data has sampling limitation during winter due to polar nights and very low solar zenith angles. However, JAXA/GOSAT data were available above 60° N during summer (Fig. 2). Further, as we found from the comparison of JAXA/GOSAT retrievals to InvSURF (see Section 3.1), posterior mole fractions in InvSURF were higher than the JAXA/GOSAT retrievals, indicating that emissions from the GOSAT inversions would likely be lower than those from InvSURF. Therefore, we argue that the lower emissions from the GOSAT inversions were not due to limited number of data, but rather reflected the agreement between the inversion and the prior. However, the inversion setups, such as the observation and transport model biases and uncertainties, rejection threshold and prior emission uncertainty, may need to be investigate further to better constrain the fluxes using the satellite data.

### 3.5 Uncertainty estimates

Despite a rather arbitrary choice of prior observational uncertainty (diagonals of **R** in Eq. 1, see Section 2.1 and 2.3), the posterior emission uncertainties were generally lower in the GOSAT inversions compared to InvSURF, and lowest in InvGLT. These differences were most pronounced in the NH tropics (EQ–30° N) (Table 2). The differences between InvSURF and GOSAT inversions were probably driven by the number of available observations, since GOSAT had much more data in the

tropics. This could also explain in part the latitudinal bias found in Section 3.1. Within the GOSAT inversions, the number of assimilated data were higher in InvGTOT than InvGLT (Fig. A9), ~~and even though~~ the observational uncertainty and rejection thresholds were the same in both inversions. This indicates that the lower uncertainty in InvGLT was not simply due to number of assimilated observations but also related to sensitivity of the data to surface fluxes. Due to the nature of the retrieved quantity, total columns have less sensitivity to surface fluxes than ground-based data or lower tropospheric partial column data, i.e. the total column data would have less power to constraint on surface fluxes. Therefore, the larger number of assimilated observations in InvGTOT did not necessary lead to higher uncertainty reduction rates than in InvGLT. However, it is possible that the prior observational uncertainty prescribed in InvGLT may have been underestimated, considering that retrieval errors are probably higher for lower tropospheric partial columns ~~compare~~ compared to total columns. Consequently, the posterior uncertainty of the fluxes may have been also underestimated. On the other hand, the transport model error may be lower for the lower tropospheric partial column data, considering that the transport model performs better in representing tropospheric concentrations than stratospheric, so the total observational uncertainty could have been reasonable. The global total flux uncertainty of InvGLT\_land was slightly higher compared to InvGLT. The posterior uncertainty was higher in the region with the largest uncertainty (EQ–30° N). Since the flux horizontal and seasonal distributions in InvGLT\_land did not change significantly from InvGLT overall, we could argue that the effect of ocean data as a constraint was minor.

The spatial distribution of uncertainty reduction rates is shown in Fig. 7. This figure indicates that the GOSAT inversions constrained fluxes mostly in tropical regions and northeast China, while InvSURF showed strongest reductions in the USA, Canada and Europe. InvSURF also showed a reduction hot-spot areas in northeast China, but the signal was much weaker compared to the GOSAT inversions. The uncertainty reduction rates over land in InvGTOT was weak in general, with fewer reduction in hot-spots compared to InvGLT and InvGLT\_land, indicating that the use of the lower tropospheric partial column data was more effective in constraining fluxes compared to total column data. The inversions with lower tropospheric partial column data showed stronger uncertainty reduction rates in Africa and India, which were not as prominent in other inversions. The weaker uncertainty reduction rates from the GOSAT inversions in North America and Europe were probably due to 1) larger uncertainty in the observations, where many surface observations in these regions had less than 30 ppb observation uncertainty (Table A1), and 2) lack of observations in high latitudes over winter.

The uncertainty reduction rates over the ocean were relatively large compared to those over land (Fig. 7). This is mainly due to the assigned prior uncertainty, which was significantly smaller over the ocean; the emissions were low (Fig. 6) and prior uncertainty was set to 20% over the ocean (see Section 2.1). This has led to prior uncertainty of  $\sim 10^{-22} \text{ mol m}^{-2} \text{ s}^{-1}$  over the ocean, which is approximately  $10^{12}$  smaller than those over land. The uncertainty reduction rates are sensitive to small changes when the prior uncertainties are small, and therefore, they became larger over the ocean, although the absolute changes in the uncertainties were small.

## 4 Discussion

### 4.1 Effect of vertical interpolation and averaging kernels

In this study, we did not apply vertical interpolation for simplicity or averaging kernels (AK) as JAXA/GOSAT v2.0 did not provide the information. However, we acknowledge that these are important to take into account when possible. With test simulations with TM5, we found that the latitudinal biases improved in  $\Delta XCH_4$  when compared to JAXA/GOSAT v3.0 data when vertical interpolation and AK were applied (Fig. A3). In the NH temperate regions, the biases in  $\Delta XCH_4$  turned from positive to negative and became similar to the biases in the tropics. Therefore, using v3.0 would possibly increase the  $CH_4$  flux estimates from the NH temperate regions in InvGTOT, aligning better with the inversions using surface and lower tropospheric partial column data. There are still positive biases in the high latitudes, although they are smaller than these compared to v2.0 data without interpolation and AK.

For lower tropospheric partial column,  $\Delta pXCH_4$  LT, the overall latitudinal biases did not change significantly by applying vertical interpolation and AK (Fig. A3). However, the large positive biases in highly elevated regions (e.g. Andean region in South America and Tibetan plateau in China and surroundings) and northeastern part of China, where  $CH_4$  emissions are large, turned to large negative biases. Nevertheless, large biases, both in positive and negative, means that the observations are likely to be rejected during assimilation and would not affect flux estimates. The  $\Delta pXCH_4$  LT over the ocean showed less latitudinal biases – the negative biases in the SH tropics turned positive. Overall, the robustness of modelling lower tropospheric partial column could be considered as an advantage over total column, having potential to provide more robust estimates regardless of differences in how transport models represent long-range transport, tropopause mixing, and atmospheric chemistry.

### 4.2 Role of upper atmosphere and OH in the estimation of $XCH_4$ values

Latitudinal differences between posterior from the surface inversion and satellite total column retrievals have been seen in earlier studies using TM5 and other global Eulerian atmospheric chemistry transport models (Alexe et al., 2015; Qu et al., 2021; Tsuruta et al., 2017; Turner et al., 2015; Zhang et al., 2021). Such discrepancies were reported regardless of the satellites' retrieval products, prior fluxes, years or seasons. Part of the misrepresentation of  $CH_4$  mole fractions in the upper troposphere could be due to convection schemes in the transport models (Saito et al., 2013), but the exact effect on the representation of total column values are to be examined. In high northern latitudes, the stratospheric profile is one of the challenges especially in polar vortex conditions (Tsuruta et al., 2023). However, polar vortex occurs only occasionally in spring, and our comparison to GOSAT, TROPOMI and TCCON  $XCH_4$  values in this and previous studies showed biases even during the periods without polar vortex conditions (Tsuruta et al., 2017, 2023). As shown in this study, latitudinal biases occur already in the upper troposphere (Fig. A6), which confirms the findings from Lindqvist et al. (2024) who argued that the role of the stratosphere in the estimation of  $XCH_4$  was minor.

In addition, we found that using higher horizontal resolution in TM5 improves agreement in the lower tropospheric partial and total columns (Fig. A2), indicating the importance of the transport model resolution. The latitudinal biases indeed seem to be slightly lower using higher spatial resolution (Stanevich et al., 2021), although exact effect is to be examined. This study was



limited in its exploration of the impact of the horizontal resolution, but increasing model resolution in the vertical dimension could also improve the representation of upper atmospheric CH<sub>4</sub>.

The discrepancies in the latitudinal gradient could also be caused by the choice of chemistry schemes in the transport models.

425 The distribution of OH, the largest sink of CH<sub>4</sub> (Saunois et al., 2024), plays an important role in regulating ambient methane levels. The OH concentration fields by Spivakovsky et al. (2000) used in this study and several others (Patra et al., 2011; Saunois et al., 2024) have relatively uniform inter-hemispheric distributions, possibly underestimating the OH concentration in the NH and overestimating them in the SH (Zhao et al., 2019). This leads to higher atmospheric CH<sub>4</sub> in the NH and lower in the SH, as shown by this and previous studies (Tsuruta et al., 2017, 2023). Zhao et al. (2020) showed that the differences in the  
430 inter-hemispheric distribution of OH could lead to about 25–50 Tg CH<sub>4</sub> yr<sup>-1</sup> differences in the inter-hemispheric distribution of CH<sub>4</sub> emissions, where Spivakovsky et al. (2000) based inversion lead to lower emissions in the NH and higher in the SH. This outcome is in line with the conclusion that the emission distributions in InvGTOT, which estimated the lowest CH<sub>4</sub> emissions in the NH, could be unreliable. The vertical OH profiles used in this study that have distinct peaks at around 500–600 hPa (Zhao et al., 2019) may also partially explain why ATom profiles and GOSAT total columns were not accurately reproduced.

### 435 4.3 Land-sea discrepancies

In CH<sub>4</sub> inversions using the GOSAT data, it has not been common to correct land-sea biases in the retrievals or exclude data over [the](#) ocean. CH<sub>4</sub> fluxes over the ocean are minor compared to those over land (Saunois et al., 2024). Consequently, the effect of land-seas biases in the JAXA/GOSAT retrieval data is expected to be small in estimation of CH<sub>4</sub> fluxes. This study showed that the emission estimates and posterior mole fractions from InvGLT and InvGLT\_land were very similar, despite the  
440 different systematic and seasonal biases over land and sea compared to InvSURF (Fig. 3), confirming that the effect of ocean data as constraints has minimum influence to the outcome of inversions. Therefore, to significantly decrease the amount of data in the inversions and increase the computational efficiency.

### 4.4 Global and regional emissions

The global total estimates from InvSURF were slightly lower than from the inversions using JAXA/GOSAT\_LT data, and on  
445 the lower edge of the range of the top-down (TD) estimates from the latest Global Methane Budgets (553–586 Tg CH<sub>4</sub> yr<sup>-1</sup>, 2010–2019 average; Saunois et al., 2024). The breakdown of anthropogenic and wetlands sources showed that anthropogenic emissions from this study were slightly higher compared to Saunois et al. (2024) (350–391 Tg CH<sub>4</sub> yr<sup>-1</sup> vs 145–214 Tg CH<sub>4</sub> yr<sup>-1</sup>, 2010–2019 TD averages). These differences ~~is~~ [occur](#) probably because of smaller wetland prior emissions (120 Tg CH<sub>4</sub> yr<sup>-1</sup>), which are on the lower boundary of bottom-up estimates from Saunois et al. (2024) (119–203 Tg CH<sub>4</sub> yr<sup>-1</sup>, 2010–2019  
450 bottom-up averages). This indicates that the prior uncertainty may need to be revised, and for example, spatially and seasonally varying uncertainty ratios ~~Tenkanen et al. (2024)~~ [Tenkanen et al. \(2025\)](#) could provide better freedom in the inversion. It is worth pointing out that we did not use a separate prior for freshwater emissions, and, thus, the freshwater emissions could be wrongly attributed to anthropogenic emissions, if there is a spatial overlap with the anthropogenic emissions.

The total emissions in the eastern part of North America agree well with previous studies (Alexe et al., 2015; Baray et al., 2021; Lu et al., 2022). Higher emissions in the northeastern part of the US compared to EDGAR (Fig. 6) point to underestimation of emissions from oil and gas. It should be noted that although we used the newer version of EDGAR (v8.0) in this study, the underestimation still seems to remain. The seasonal variability of anthropogenic CH<sub>4</sub> emissions in southern North America (including the contiguous United States and Mexico) from the GOSAT inversions (Fig. A10a) showed opposite patterns compared to InvSURF. The seasonal cycle in this region is likely to be associated with natural gas consumption (Zeng et al., 2023) and agriculture (Maasakkers et al., 2023). In contrast to results from Maasakkers et al. (2023) who argue that the seasonal pattern of natural gas consumption has strong interannual variations and is spatially inhomogeneous, our results from the GOSAT inversions were similar to those from Miller et al. (2013) which showed larger emissions during autumn and winter compared to summer.

In China, InvSURF and InvGTOT showed strong negative emission increments ~~around Beijing~~ (i.e. posterior emissions were lower than prior) around Beijing (Fig. 6), which is consistent with studies such as Lu et al. (2021) and Wang et al. (2022) who assimilated both surface and the GOSAT XCH<sub>4</sub> data. This seems to be a common feature found in other studies despite different GOSAT retrieval products, prior, transport and inverse models were used (Lu et al., 2021, and references therein). On the other hand, InvGLT and InvGLT\_land showed positive emission increments in eastern and southern China. This is closer to the findings from Chen et al. (2022) who assimilated the TROPOMI XCH<sub>4</sub> data. However, these results again contradict with Qu et al. (2021) who assimilated both GOSAT and TROPOMI XCH<sub>4</sub> data and argued that an inversion using only TROPOMI XCH<sub>4</sub> may be unreliable over China because the TROPOMI XCH<sub>4</sub> is strongly affected by cloud cover.

In central Africa and South Sudan wetland regions, our GOSAT inversions showed a large increase in total emissions (Fig. 6), which were primarily associated with wetland emissions. This is in line with findings from Lunt et al. (2019), who used the GOSAT XCH<sub>4</sub>, and Pandey et al. (2021), who used the TROPOMI XCH<sub>4</sub> in their inversions, although our posterior estimates were considerably lower. For instance, we found monthly maxima below 1 Tg CH<sub>4</sub> month<sup>-1</sup> around South Sudan, compared to Lunt et al. (2019) estimates of 7 Tg CH<sub>4</sub> month<sup>-1</sup> and Pandey et al. (2021) estimates of seasonal cycle amplitude of about 2–3 Tg CH<sub>4</sub> month<sup>-1</sup>. Nevertheless, the seasonal cycle in this region from the GOSAT inversions (Fig. A10b) showed significantly lower emissions in June–July and later maxima in August–September compared to prior and InvSURF. This corresponds slightly better to dry and wet seasons and is in line with Lunt et al. (2019) and Pandey et al. (2021), although we could not reproduce high emissions in late months.

The magnitude of high northern latitude (NHL) wetlands remained uncertain. Previous studies showed that in some cases, NHL emissions from the inversion based on the GOSAT data were lower compared to those based on surface data (Pandey et al., 2016; Feng et al., 2017), while others showed the opposite (Alexe et al., 2015; Baray et al., 2021). Our analysis showed that the wetland emissions for 60° N–90° N were lower in the GOSAT inversions regardless of the assimilated data type (total or lower tropospheric partial column) compared to InvSURF. This is in line with our previous studies assimilating TROPOMI XCH<sub>4</sub> data (Tsuruta et al., 2023), and using other wetland priors, such as JSBACH-HIMMELI, where emissions were relatively larger among process-based models ~~(Aalto et al., 2024; Tenkanen et al., 2024)~~ (Aalto et al., 2024; Tenkanen et al., 2025). Tsuruta et al. (2023) assumed that the lower NHL wetland emissions in the TROPOMI inversions were due to latitudinal

biases associated with model-retrieval differences (Lindqvist et al., 2024). However, as shown in this study, the inversion using  
490 lower tropospheric partial column data also resulted in lower NHL wetland emissions compared to InvSURF, indicating that  
there may be fundamental discrepancies between satellite and surface inversions. The differences were partly due to spatial  
coverages and temporal distributions of the observations and uncertainties associated with transport models, observations, prior  
emissions and possible biases in the satellite data, but further study is needed to find the exact cause of these discrepancies and  
to obtain more robust the emission estimates.

## 495 5 Conclusions

This study presented the advantages of JAXA/GOSAT lower tropospheric partial column retrievals in estimating global and  
regional CH<sub>4</sub> budgets using the CTE-CH<sub>4</sub> atmospheric inverse model. Our findings showed that assimilating the lower tropo-  
spheric partial column data led to posterior CH<sub>4</sub> fluxes and atmospheric CH<sub>4</sub> mole fractions that were more consistent with  
the inversion estimates using surface data compared to total column retrievals. In addition, partial column retrievals ~~better-con-~~  
500 strained CH<sub>4</sub> fluxes ~~in low latitudes~~ better than the total column retrievals globally and better than surface data in low latitudes  
with sparse observation network. This is a considerable advantage to the atmospheric inverse modelling community, and the  
partial column product could potentially be a better product than total column for estimation of CH<sub>4</sub> fluxes.

In addition, we found that lower tropospheric partial column data possibly reduce global emission uncertainty. ~~In addition~~ Furthermore,  
it was concluded that the lower tropospheric partial column ocean data have minimal influence on constraining CH<sub>4</sub> fluxes over  
505 land, suggesting that excluding ocean data could improve computational efficiency. However, further studies are needed to as-  
sess uncertainty in the partial column retrievals and transport model's ability to represent partial column mole fractions. Our  
results showed that the uncertainty reduction rates were low in North America and Europe in the GOSAT inversions, indicating  
the need to further investigate uncertainties in the JAXA/GOSAT data and the transport model, as well as the critical issue of  
lack of the satellite data in high latitudes during winter. This study also highlighted the importance of transport model resolu-  
510 tion in estimation of total and partial column data, indicating the need for high resolution transport models in satellite-driven  
inversions.

Our study was limited to retrievals from one satellite (GOSAT) assimilated to a single inverse model (CTE-CH<sub>4</sub>) for a  
relatively short period (four years). Future efforts should focus on exploiting the use of other satellite datasets and inverse  
models. JAXA/GOSAT provides partial column products since 2009 up to today, including those from GOSAT-2 since 2019,  
515 that will allow expanding the study period to perform trend analysis. In addition, in this study, ~~we did not take into account~~  
~~averaging kernels~~ averaging kernels were taken into account only in a test simulation with TM5 for a limited period and detailed  
retrieval uncertainty ~~, which is~~ were not taken into account. These are available in the newest version of JAXA/GOSAT partial  
column product (v3.0). ~~Including~~, and including this information will possibly result in more realistic estimates of CH<sub>4</sub> fluxes  
and their uncertainties. Lastly, JAXA/GOSAT provides GOSAT partial column products for upper layers (upper tropospheric  
520 and three stratospheric layers). The use of these products in inversions can provide useful information in identifying the causes  
of transport model biases in vertical layers.

*Code and data availability.* All the model results, inputs and code will be provided on request from the corresponding author (Aki Tsuruta, aki.tsuruta@fmi.fi).

*Author contributions.* The research was conceptualized by HS with contributions from AT and TA. AT developed model codes for assimilation of lower tropospheric partial column data, and carried out all inverse modelling simulations and visualization. AT did analysis with contributions from AK, KSh, FK, TA, MKT, LF, EK and HS. AK, KSh, FK, NK and HS provided the GOSAT data. EK calculated modelled XCH<sub>4</sub> values at TCCON sites. LF calculated TCCON tropospheric partial columns. KM provided ATom data. TCCON data were provided by OEG (Izaña), FH (Karlsruhe), RK (Sodankylä), IM (Tsukuba), HO (Rikubetsu), DFP (Lauder), KSh (Saga), KSt (Eureka), RS (Garmisch), MKS (Reunion Island), YT (Paris), VAV (Burgos), MV (Nicosia), TW (Orleans) and MZ (Xianghe). AT wrote the original draft of the manuscript with contributions from AK, LB, EK, MKT and HS. All authors have read and approved to publish the version of the manuscript.

*Competing interests.* The contact author has declared that none of the authors has any competing interests.

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## Appendix A: Appendix A

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**Table A1.** The ground-based in-situ measurement sites used in InvSURF and evaluation. Observation uncertainty (Obs. Unc.) is sum of measurement and transport model errors used in the observation covariance matrix. Data type is categorized into two: discrete (D) and continuous (C).

ID	Sites	Country/Territory	Laboratory	Longitude [deg. E]	Latitude [deg. N]	Altitude [m a.s.l.]	Obs. Unc. [ppb]	Data type
ABT	Abbotsford, British Columbia	Canada	ECCC	-122.34	49.01	93	30	C
ALT	Alert, Nunavut	Canada	NOAA	-62.51	82.45	190	15	D
ALT	Alert, Nunavut	Canada	ECCC	-62.51	82.45	195	15	C
AMY	Anmyeon-do	Republic of Korea	NOAA	126.33	36.54	87	30	D
ASC	Ascension Island	United Kingdom	NOAA	-14.4	-7.97	90	15	D
ASK	Assekrem	Algeria	NOAA	5.63	23.26	2715	25	D
AZR	Terceira Island, Azores	Portugal	NOAA	-27.36	38.76	24	15	D
AZV	Azovo	Russian Federation	NIES	73.03	54.71	190	30	C
BAR	Baranova	Russian Federation	FMI	101.62	79.28	30	4.5	C
BCK	Behchoko, Northwest Territories	Canada	ECCC	-115.92	62.8	220	15	C
BHD	Baring Head Station	New Zealand	NOAA	174.87	-41.41	90	4.5	D
BKT	Bukit Kototabang	Indonesia	NOAA	100.31	-0.2	875	75	D
BLK	Baker Lake, Nunavut	Canada	ECCC	-96.01	64.33	61	15	C
BMW	Tudor Hill, Bermuda	United Kingdom	NOAA	-64.88	32.26	33	15	D
BRA	Bratt's Lake Saskatchewan	Canada	ECCC	-104.71	50.2	630	75	C
BRW	Barrow Atmospheric Baseline Observatory	United States	NOAA	-156.61	71.32	27	15	C
BRW	Barrow Atmospheric Baseline Observatory	United States	NOAA	-156.58	71.32	16	15	D
BRZ	Berezorechka	Russian Federation	NIES	84.33	56.15	248	75	C
BSD	Bilsdale	United Kingdom	UNIVBRIS	-1.15	54.36	628	30	C
CBA	Cold Bay, Alaska	United States	NOAA	-162.71	55.21	25	15	D
CBY	Cambridge Bay, Nunavut Territory	Canada	ECCC	-105.06	69.13	47	15	C
CFA	Cape Ferguson	Australia	CSIRO	147.06	-19.28	5	25	D
CGO	Cape Grim, Tasmania	Australia	NOAA	144.68	-40.68	164	4.5	D
CGO	Cape Grim	Australia	CSIRO	144.68	-40.68	94	15	C
CGR	Charles Point, Darwin	Australia	CSIRO	12.65	37.67	9	25	C
CHL	Churchill, Manitoba	Canada	ECCC	-93.82	58.74	89	15	C
CHR	Christmas Island	Republic of Kiribati	NOAA	-157.15	1.7	5	15	D
CMN	Mt. Cimone Station	Italy	ICOS-ATC,CNR-ISAC	10.7	44.19	2173	15	C
CPS	Chapais,Quebec	Canada	ECCC	-74.98	49.82	431	15	C
CPT	Cape Point	South Africa	NOAA	18.49	-34.35	260	25	D
CRV	Carbon in Arctic Reservoirs Vulnerability Expe...	United States	NOAA	-147.6	64.99	643	15	C
CRZ	Crozet Island	France	NOAA	51.85	-46.43	202	4.5	D
CUR	Monte Curcio	Italy	IIA	16.42	39.32	1801	15	C
CYA	Casey Station, Antarctica	Australia	CSIRO	110.52	-66.28	55	4.5	D
DEM	Demyanskoe	Russian Federation	NIES	70.87	59.79	155	30	C
DRP	Drake Passage	Drake Passage	NOAA	-61.68	-59.07	10	4.5	D
DSI	Dongsha Island	Taiwan	NOAA	116.73	20.7	8	15	D
DVV	Danville, Virginia	United States	PSU	-79.44	36.71	492	15	C
EGB	Egbert, Ontario	Canada	ECCC	-79.78	44.23	276	25	C
EIC	Easter Island	Chile	NOAA	-109.45	-27.13	72	4.5	D
ENA	Eastern North Atlantic, Graciosa, Azores	Portugal	LBNL-ARM	-28.03	39.09	40	25	C



**Table A1.** Continuation to Table A1.

ESP	Estevan Point, British Columbia	Canada	ECCC	-126.54	49.38	47	25	C
EST	Esther, Alberta	Canada	ECCC	-110.21	51.67	757	30	C
ETL	East Trout Lake, Saskatchewan	Canada	ECCC	-104.99	54.35	598	30	C
FNE	Fort Nelson, British Columbia	Canada	ECCC	-122.57	58.84	376	30	C
FSD	Fraserdale	Canada	ECCC	-81.57	49.88	250	30	C
GAT	Gartow	Germany	ICOS-ATC,HPB	11.44	53.07	411	25	C
GCI	Millerville, AL	United States	PSU	-85.89	33.18	428	25	C
GMI	Mariana Islands	Guam	NOAA	144.66	13.39	8	15	D
GPA	Gunn Point	Australia	CSIRO	131.04	-12.25	37	75	D
HBA	Halley Station, Antarctica	United Kingdom	NOAA	-26.21	-75.61	35	4.5	D
HNP	Hanlan's Point, Ontario	Canada	ECCC	-79.39	43.61	97	25	C
HPB	Hohenpeissenberg	Germany	ICOS-ATC,HPB	11.02	47.8	1065	25	C
HSU	Humboldt State University	United States	NOAA	-124.44	41.57	8	30	D
HTM	Hyltemossa	Sweden	ICOS-ATC,LUND-CEC	13.42	56.1	265	25	C
ICE	Storhofdi, Vestmannaeyjar	Iceland	NOAA	-20.29	63.4	122	15	D
INU	Inuvik, Northwest Territories	Canada	ECCC	-133.53	68.32	123	15	C
IPR	Ispra	Italy	ICOS-ATC,JRC	8.64	45.81	310	30	C
IZO	Izana, Tenerife, Canary Islands	Spain	NOAA	-16.48	28.3	2378	25	D
JFJ	Jungfraujoch	Switzerland	ICOS-ATC,HFSJG	7.99	46.55	3585	15	C
KEY	Key Biscayne, Florida	United States	NOAA	-80.2	25.67	6	25	D
KIT	Karlsruhe	Germany	ICOS-ATC,HPB	8.42	49.09	310	30	C
KJN	Kjølnes	Norway	Univ. Exeter	29.23	70.85	20	15	C
KMP	Kumpula	Finland	FMI	24.96	60.2	53	30	C
KRE	Kresin u Pacova	Czech Republic	ICOS	15.08	49.57	784	25	C
KRS	Karasevoe	Russian Federation	NIES	82.42	58.25	156	30	C
KUM	Cape Kumukahi, Hawaii	United States	NOAA	-155.01	19.51	3	15	D
LEF	Park Falls, Wisconsin	United States	NOAA	-90.27	45.95	868	30	C
LIN	Lindenberg	Germany	ICOS-ATC,HPB	14.12	52.17	171	30	C
LLB	Lac La Biche, Alberta	Canada	ECCC	-112.47	54.95	590	30	C
LLN	Lulin	Taiwan	NOAA	120.86	23.47	2867	25	D
LMP	Lampedusa	Italy	ICOS-ATC,ENEA	12.63	35.52	53	25	C
LMT	Lamezia Terme	Italy	ISAC	16.23	38.88	14	30	C
LUT	Lutjewad	Netherlands	ICOS-ATC,RUG	6.35	53.4	61	25	C
MAA	Mawson, Antarctica	Australia	CSIRO	62.87	-67.62	32	4.5	D
MEX	High Altitude Global Climate Observation Center	Mexico	NOAA	-97.31	18.98	4469	15	D
MID	Sand Island, Midway	United States	NOAA	-177.38	28.21	8	15	D
MLO	Mauna Loa, Hawaii	United States	NOAA	-155.58	19.54	3437	15	C/D
MNM	Minamitorishima	Japan	JMA	153.98	24.29	27	15	C
MQA	Macquarie Island	Australia	CSIRO	158.97	-54.48	13	4.5	D
MRC	Marcellus Pennsylvania	United States	PSU	-76.42	41.47	652	75	C

**Table A1.** Continuation of Table A1.

NAT	Farol De Mae Luiza Lighthouse	Brazil	NOAA	-35.19	-5.51	20	15	D
NMB	Gobabeb	Namibia	NOAA	15.01	-23.58	461	25	D
NOR	Norunda	Sweden	ICOS-ATC,LUND-CEC	17.48	60.09	146	15	C
NOY	Noyabrsk	Russian Federation	NIES	75.78	63.43	188	30	C
NWR	Niwot Ridge, Colorado	United States	NOAA	-105.57	40.05	3526	15	D
OPE	Observatoire perenne de l'environnement	France	ICOS-ATC,LSCE	5.5	48.56	510	30	C
OXK	Ochsenkopf	Germany	ICOS-ATC,CAL-FCL	11.81	50.03	1185	30	C
PAL	Pallas-Sammaltunturi, GAW Station	Finland	ICOS-ATC,FMI	24.12	67.97	577	15	C
PDM	Pic du Midi	France	LSCE	0.14	42.94	2887	15	D
POC	Pacific Ocean	Pacific Ocean	NOAA	-130.75	0.12	20	15	D
PSA	Palmer Station, Antarctica	United States	NOAA	-64.05	-64.77	15	4.5	D
PUI	Puijo	Finland	ICOS-ATC,UEF	27.66	62.91	84	30	C
PUY	Puy de Dome	France	ICOS-ATC,LSCE	2.97	45.77	1475	15	C
RPB	Ragged Point	Barbados	NOAA	-59.43	13.16	20	15	D
RUN	La Réunion	France	ICOS-ATC,LSCE	55.38	-21.08	2160	15	C
RYO	Ryori	Japan	JMA	141.82	39.03	280	15	C
SAC	Saclay	France	ICOS-ATC,CEA	2.14	48.72	260	75	C
SCT	Beech Island, South Carolina	United States	NOAA	-81.83	33.41	420	75	C
SDZ	Shangdianzi	China	NOAA	117.12	40.65	298	15	D
SEY	Mahe Island	Seychelles	NOAA	55.53	-4.68	7	15	D
SGP	Southern Great Plains, Oklahoma	United States	NOAA	-97.5	36.62	339	75	D
SGP	Southern Great Plains, Oklahoma	United States	LBNL-ARM	-97.49	36.61	374	75	C
SHM	Shemya Island, Alaska	United States	NOAA	174.08	52.72	28	25	D
SMO	Tutuila	American Samoa	NOAA	-170.56	-14.23	60	15	D
SMR	Hyytiala	Finland	ICOS-ATC,UHELS	24.29	61.85	306	25	C
SNB	Sonnblick	Austria	EAA	47.05	12.96	3111	15	C
SOD	Sodanyklä	Finland	FMI	26.64	67.36	227	25	C
SPO	South Pole, Antarctica	United States	NOAA	-24.8	-89.96	2821	4.5	D
STE	Steinkimmen	Germany	ICOS-ATC,HPB	8.46	53.04	281	75	C
SUM	Summit	Greenland	NOAA	-38.42	72.6	3215	15	D
SVB	Svartberget	Sweden	ICOS-ATC,SLU	19.77	64.26	419	25	C
SYO	Syowa Station, Antarctica	Japan	NOAA	39.59	-69	16	4.5	D
TAC	Tacolneston	United Kingdom	NOAA	1.14	52.52	236	25	D
TAP	Tae-ahn Peninsula	Republic of Korea	NOAA	126.13	36.73	21	75	D
THD	Trinidad Head, California	United States	NOAA	-124.15	41.05	112	25	D
TIK	Hydrometeorological Observatory of Tiksi	Russia	NOAA	128.89	71.6	29	15	D
TIK	Tiksi	Russian Federation	FMI	128.89	71.6	29	15	C

**Table A1.** Continuation of Table A1.

TOH	Torfhaus	Germany	ICOS-ATC,HPB	10.53	51.81	948	25	C
TPD	Turkey Point, Ontario	Canada	ECCC	-80.56	42.64	266	25	C
TRN	Trainou	France	ICOS-ATC,LSCE	2.11	47.96	311	25	C
USH	Ushuaia	Argentina	NOAA	-68.31	-54.85	32	4.5	D
UTA	Wendover, Utah	United States	NOAA	-113.72	39.9	1332	25	D
UTO	Uto	Finland	ICOS-ATC,FMI	21.37	59.78	65	25	C
UUM	Ulaan Uul	Mongolia	NOAA	111.1	44.45	1012	25	D
VGN	Vaganovo	Russian Federation	NIES	62.32	54.5	277	30	C
WIS	Weizmann Institute of Science at the Arava Ins...	Israel	NOAA	35.06	29.96	482	25	D
WLG	Mt. Waliguan	Peoples Republic of China	NOAA	100.9	36.27	3890	15	D
WSA	Sable Island, Nova Scotia	Canada	ECCC	-60.01	43.93	8	25	C
YON	Yonagunijima	Japan	JMA	123.01	24.47	50	30	C
ZEP	Ny-Alesund, Svalbard	Norway and Sweden	ICOS-ATC,NILU	11.89	78.91	489	15	C
ZOT	Zotino	Russian Federation	MPIBGC	89.21	60.48	415	25	C

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**Table A2.** List of the ICOS sites, data categories, PIs and references for the sites used in InvSURF and evaluation.

Site code	Data category	Authors	References
CMN	ICOS ATC NRT CH4 growing time series	Cristofanelli, Paolo and Trisolino, Pamela	Cristofanelli and Trisolino (2022)
CMN	ICOS ATC CH4 Release	Cristofanelli, Paolo and Trisolino, Pamela	Cristofanelli and Trisolino (2021)
GAT	ICOS ATC NRT CH4 growing time series	Kubistin, Dagmar and Plaß-Dülmer, Christian and Kneuer, Tobias and Lindauer, Matthias and Müller-Williams, Jennifer	Kubistin et al. (2022a)
GAT	ICOS ATC CH4 Release	Kubistin, Dagmar and Plaß-Dülmer, Christian and Arnold, Sabrina and Lindauer, Matthias and Müller-Williams, Jennifer and Schumacher, Marcus	Kubistin et al. (2021c)
HPB	ICOS ATC NRT CH4 growing time series	Kubistin, Dagmar and Plaß-Dülmer, Christian and Kneuer, Tobias and Lindauer, Matthias and Müller-Williams, Jennifer	Kubistin et al. (2022b)
HPB	ICOS ATC CH4 Release	Kubistin, Dagmar and Plaß-Dülmer, Christian and Arnold, Sabrina and Lindauer, Matthias and Müller-Williams, Jennifer and Schumacher, Marcus	Kubistin et al. (2021d)
HTM	ICOS ATC NRT CH4 growing time series	Heliasz, Michal and Biermann, Tobias	Heliasz and Biermann (2022)
HTM	ICOS ATC CH4 Release	Heliasz, Michal and Biermann, Tobias	Heliasz and Biermann (2021)
IPR	ICOS ATC NRT CH4 growing time series	Manca, Giovanni	Manca (2022)
IPR	ICOS ATC CH4 Release	Bergamaschi, Peter and Manca, Giovanni	Bergamaschi and Manca (2021)
JFJ	ICOS ATC NRT CH4 growing time series	Emmenegger, Lukas and Leuenberger, Markus and Steinbacher, Martin	Emmenegger et al. (2022)
JFJ	ICOS ATC CH4 Release	Emmenegger, Lukas and Leuenberger, Markus and Steinbacher, Martin	Emmenegger et al. (2021)
KIT	ICOS ATC NRT CH4 growing time series	Kubistin, Dagmar and Plaß-Dülmer, Christian and Kneuer, Tobias and Lindauer, Matthias and Müller-Williams, Jennifer	Kubistin et al. (2022c)
KIT	ICOS ATC CH4 Release	Kubistin, Dagmar and Plaß-Dülmer, Christian and Arnold, Sabrina and Lindauer, Matthias and Müller-Williams, Jennifer and Schumacher, Marcus	Kubistin et al. (2021e)
KRE	ICOS ATC NRT CH4 growing time series	Marek, Michal V. and Vítková, Gabriela and Komínková, Kateřina	Marek et al. (2022)
KRE	ICOS ATC CH4 Release	Marek, Michal V. and Vítková, Gabriela and Komínková, Kateřina	Marek et al. (2021)
LIN	ICOS ATC NRT CH4 growing time series	Kubistin, Dagmar and Plaß-Dülmer, Christian and Kneuer, Tobias and Lindauer, Matthias and Müller-Williams, Jennifer	Kubistin et al. (2022d)
LIN	ICOS ATC CH4 Release	Kubistin, Dagmar and Plaß-Dülmer, Christian and Arnold, Sabrina and Lindauer, Matthias and Müller-Williams, Jennifer and Schumacher, Marcus	Kubistin et al. (2021f)
LMP	ICOS ATC NRT CH4 growing time series	di Sarra, Alcide and Piacentino, Salvatore	di Sarra and Piacentino (2022)
LMP	ICOS ATC CH4 Release	di Sarra, Alcide and Piacentino, Salvatore	di Sarra and Piacentino (2021)
LUT	ICOS ATC NRT CH4 growing time series	Chen, Huilin and Scheeren, Bert	Chen and Scheeren (2022)
LUT	ICOS ATC CH4 Release	Chen, Huilin and Scheeren, Bert	Chen and Scheeren (2021)
NOR	ICOS ATC NRT CH4 growing time series	Lehner, Irene and Mölder, Meelis	Lehner and Mölder (2022)
NOR	ICOS ATC CH4 Release	Lehner, Irene and Mölder, Meelis	Lehner and Mölder (2021)
OPE	ICOS ATC NRT CH4 growing time series	Ramonet, Michel and Conil, Sébastien and Delmotte, Marc and Laurent, Olivier and Lopez, Morgan	Ramonet et al. (2022a)
OPE	ICOS ATC CH4 Release	Ramonet, Michel and Conil, Sébastien and Delmotte, Marc and Laurent, Olivier	Ramonet et al. (2021a)
OXK	ICOS ATC NRT CH4 growing time series	Kubistin, Dagmar and Plaß-Dülmer, Christian and Kneuer, Tobias and Lindauer, Matthias and Müller-Williams, Jennifer	Kubistin et al. (2022e)
OXK	ICOS ATC CH4 Release	Kubistin, Dagmar and Plaß-Dülmer, Christian and Arnold, Sabrina and Lindauer, Matthias and Müller-Williams, Jennifer	Kubistin et al. (2021a)
PAL	ICOS ATC NRT CH4 growing time series	Hatakka, Juha	Hatakka (2022)
PAL	ICOS ATC CH4 Release	Hatakka, Juha	Hatakka (2021)
PUI	ICOS ATC NRT CH4 growing time series	Lehtinen, Kari and Leskinen, Ari	Lehtinen and Leskinen (2022)
PUY	ICOS ATC NRT CH4 growing time series	Colomb, Aurélie and Ramonet, Michel and Yver-Kwok, Camille and Delmotte, Marc and Lopez, Morgan and Pichon, Jean-Marc	Colomb et al. (2022)
PUY	ICOS ATC CH4 Release	Colomb, Aurélie and Ramonet, Michel and Yver-Kwok, Camille and Delmotte, Marc and Pichon, Jean-Marc	Colomb et al. (2021)
RUN	ICOS ATC NRT CH4 growing time series	De Mazière, Martine and Sha, Mahesh Kumar and Ramonet, Michel	De Mazière et al. (2022b)
RUN	ICOS ATC CH4 Release	De Mazière, Martine and Sha, Mahesh Kumar and Ramonet, Michel	De Mazière et al. (2021)
SAC	ICOS ATC NRT CH4 growing time series	Ramonet, Michel and Delmotte, Marc and Lopez, Morgan	Ramonet et al. (2022b)
SAC	ICOS ATC CH4 Release	Ramonet, Michel and Delmotte, Marc	Ramonet and Delmotte (2021)
SMR	ICOS ATC NRT CH4 growing time series	Mammarella, Ivan	Mammarella (2022)
SMR	ICOS ATC CH4 Release	Levula, Janne and Mammarella, Ivan	Levula and Mammarella (2021)
STE	ICOS ATC NRT CH4 growing time series	Kubistin, Dagmar and Plaß-Dülmer, Christian and Kneuer, Tobias and Lindauer, Matthias and Müller-Williams, Jennifer	Kubistin et al. (2022f)
STE	ICOS ATC CH4 Release	Kubistin, Dagmar and Plaß-Dülmer, Christian and Arnold, Sabrina and Lindauer, Matthias and Müller-Williams, Jennifer	Kubistin et al. (2021b)
SVB	ICOS ATC NRT CH4 growing time series	Smith, Paul and Marklund, Per	Smith and Marklund (2022)
SVB	ICOS ATC CH4 Release	Marklund, Per and Ottosson-Löfvenius, Mikael and Smith, Paul	Marklund et al. (2021)
TOH	ICOS ATC NRT CH4 growing time series	Kubistin, Dagmar and Plaß-Dülmer, Christian and Kneuer, Tobias and Lindauer, Matthias and Müller-Williams, Jennifer	Kubistin et al. (2022g)
TOH	ICOS ATC CH4 Release	Kubistin, Dagmar and Plaß-Dülmer, Christian and Arnold, Sabrina and Lindauer, Matthias and Müller-Williams, Jennifer and Schumacher, Marcus	Kubistin et al. (2021g)
TRN	ICOS ATC NRT CH4 growing time series	Ramonet, Michel and Lopez, Morgan and Delmotte, Marc	Ramonet et al. (2022c)
TRN	ICOS ATC CH4 Release	Ramonet, Michel and Lopez, Morgan and Delmotte, Marc	Ramonet et al. (2021b)
UTO	ICOS ATC NRT CH4 growing time series	Hatakka, Juha and Laurila, Tuomas	Hatakka and Laurila (2022)
UTO	ICOS ATC CH4 Release	Laurila, Tuomas	Laurila (2021)
ZEP	ICOS ATC NRT CH4 growing time series	Lund Myhre, Cathrine and Platt, Stephen Matthew and Hermansen, Ove and Lunder, Chris	Lund Myhre et al. (2022)
ZEP	ICOS ATC CH4 Release	Lund Myhre, Cathrine and Platt, Stephen Matthew and Hermansen, Ove and Lunder, Chris	Lund Myhre et al. (2021)

**Table A3.** The TCCON sites used for evaluation.

ID	Station Name, Location	Latitude	Longitude	References
br	Bremen, Germany	53.1°N	8.85°E	Notholt et al. (2022)
bu	Burgos, Philippines	18.53°N	120.65°E	Morino et al. (2022c)
ci	California Institute of Technology, USA	34.14°N	118.13°W	Wennberg et al. (2022b)
db	Darwin, Australia	12.46°S	130.89°E	Deutscher et al. (2023b)
df	Armstrong Flight Research Center, USA	34.96°N	117.88°W	Iraci et al. (2022)
et	East Trout Lake, Canada	54.36°N	104.99°W	Wunch et al. (2002)
eu	Eureka, Canada	80.05°N	86.42°W	Strong et al. (2022)
gm	Garmisch, Germany	47.48°N	11.06°E	Sussmann and Rettinger (2017)
hf	Hefei, China	31.9°N	117.17°E	Liu et al. (2023)
iz	Izaña, Spain	28.3°N	16.5°W	Blumenstock et al. (2017)
js	Saga, Japan	33.24°N	130.29°E	Shiomi et al. (2022)
ka	Karlsruhe, Germany	49.1°N	8.44°E	Hase et al. (2022)
ll	Lauder, New Zealand	45.04°S	169.68°E	Sherlock et al. (2022)
ni	Nicosia, Cyprus	35.14°N	33.38°E	Petri et al. (2023)
ny	Ny Ålesund, Norway	78.92°N	11.92°E	Buschmann et al. (2022)
oc	Lamont, USA	36.6°N	97.49°W	Wennberg et al. (2022c)
or	Orleans, France	47.97°N	2.11°E	Warneke et al. (2022)
pa	Park Falls, USA	45.94°N	90.27°W	Wennberg et al. (2022a)
pr	Paris, France	48.85°N	2.36°E	Te et al. (2022)
ra	Reunion Island, France	20.9°S	55.49°E	De Mazière et al. (2022a)
rj	Rikubetsu, Japan	43.46°N	143.77°E	Morino et al. (2022a)
so	Sodankylä, Finland	67.37°N	26.63°E	Kivi et al. ((2022)
tk	Tsukuba, Japan	36.05°N	140.12°E	Morino et al. (2022b)
wg	Wollongong, Australia	34.41°S	150.88°E	Deutscher et al. (2023a)
xh	Xianghe, China	39.8°N	116.96°E	Zhou et al. (2022)

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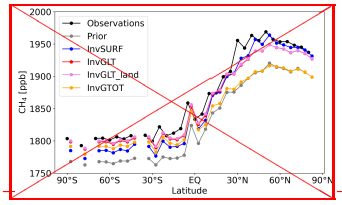


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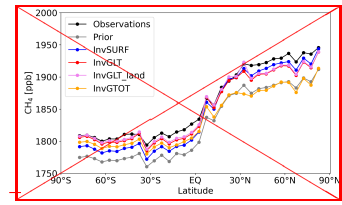
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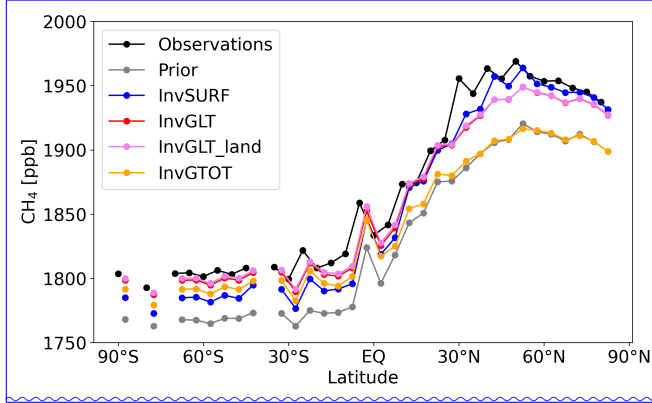
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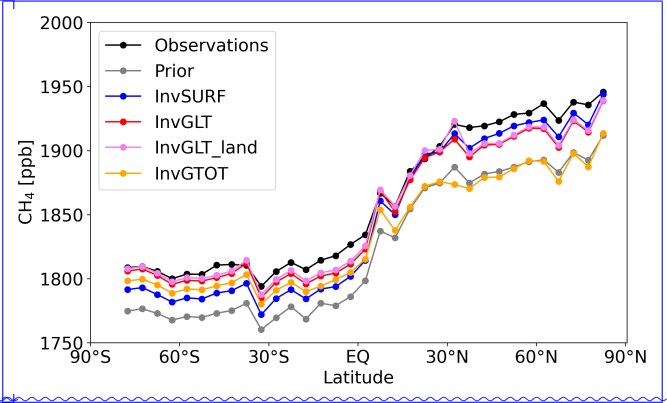
(a) Surface stations



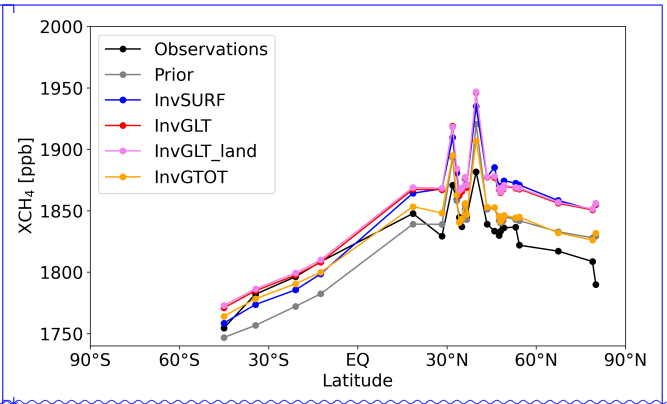
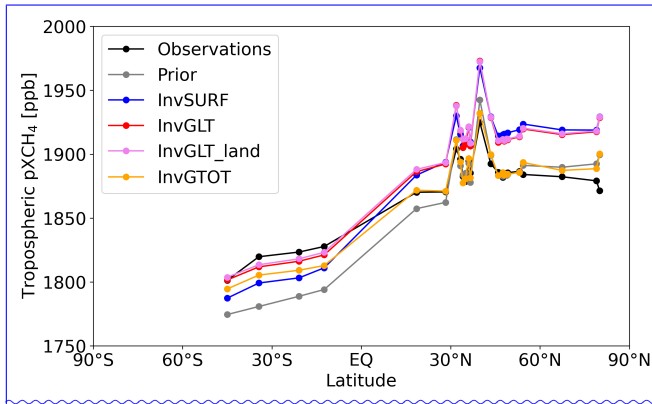
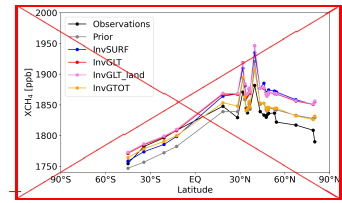
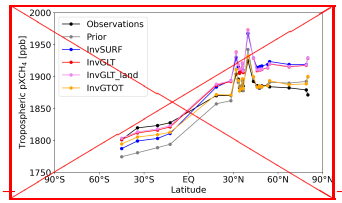
(b) ATom, 0–2000 m



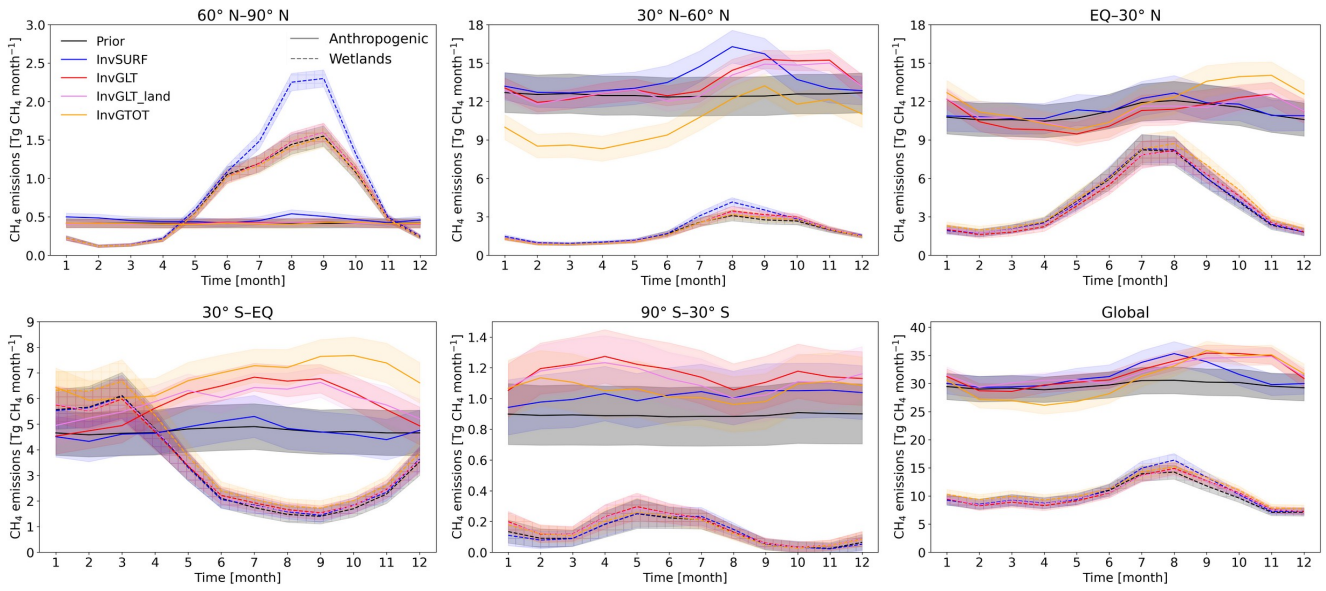
(c) TCCON, tropospheric partial column



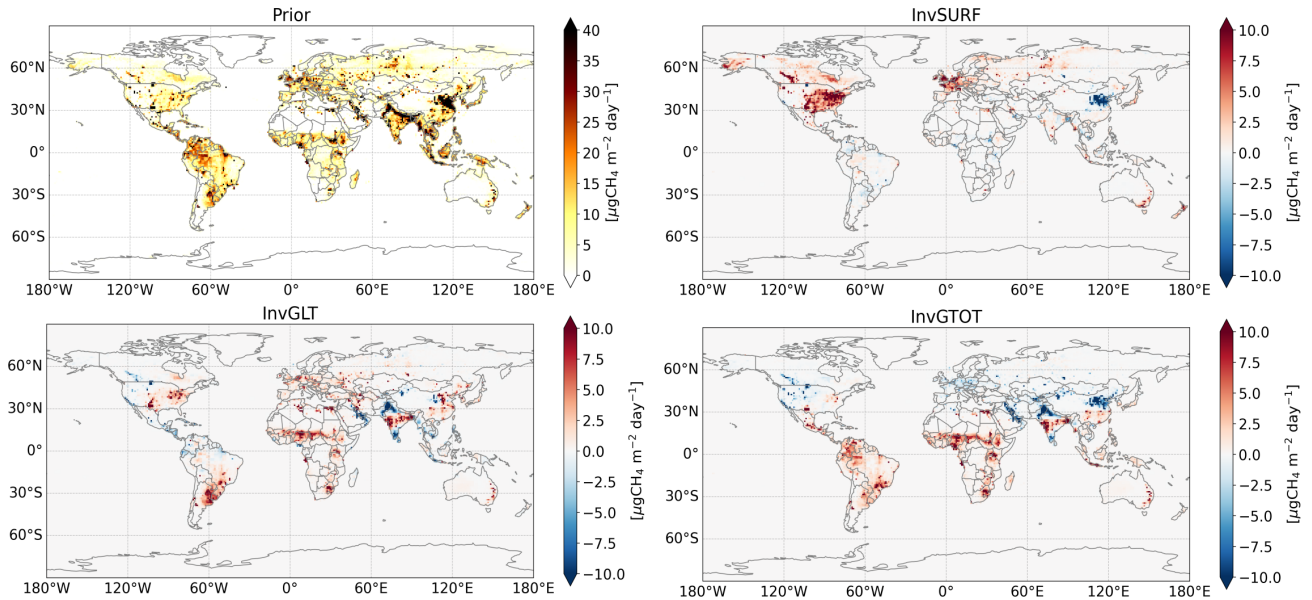
(d) TCCON, total column



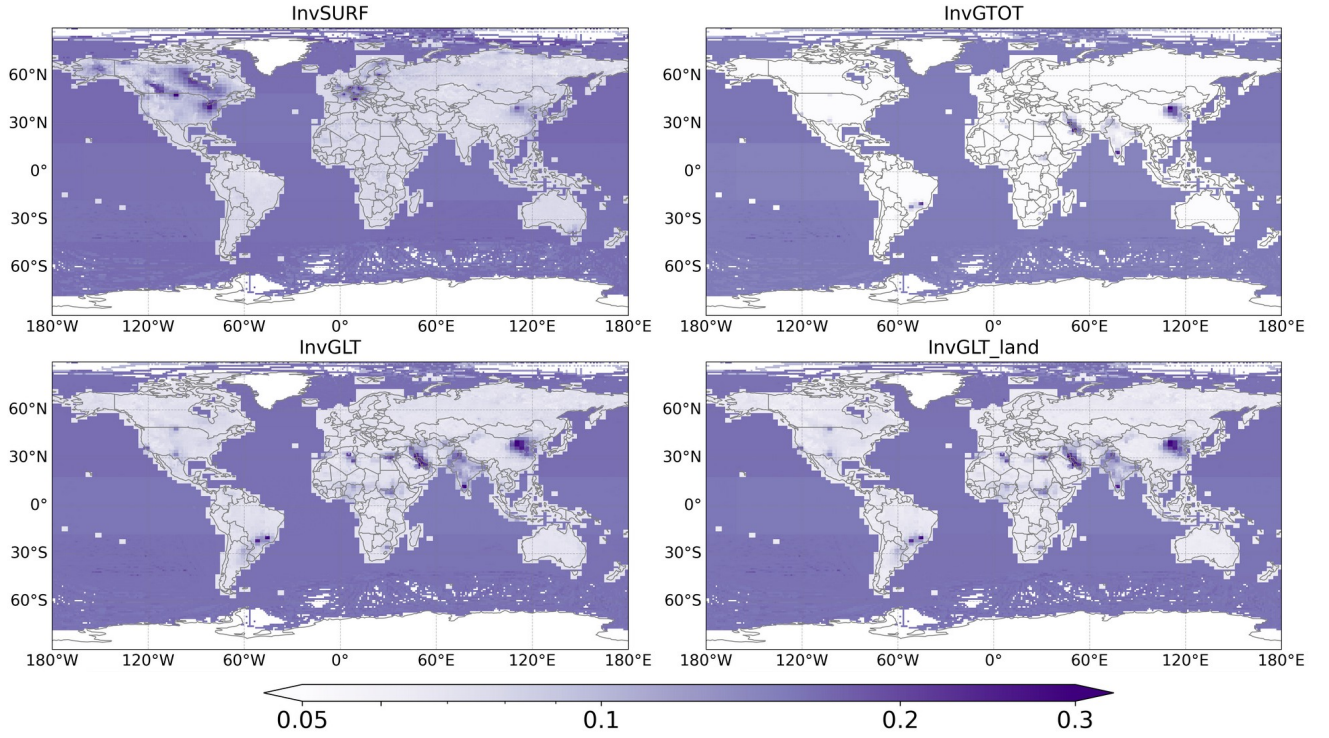
**Figure 4.** Mean atmospheric  $\text{CH}_4$ , averaged over 2016–2019 at (a) surface ground-based and shipboard stations, (b) ATom observation locations, and (c) and (d) TCCON sites. For (a), the data that were assimilated in InvSURF were used. For (a) and (b), mean over five-degree latitude bands are shown. For (c) and (d), no spatial averaging is applied. The coloured lines are the posterior estimates from each inversion, which is estimated by running TM5 forward using posterior fluxes of the corresponding inversion simulation.



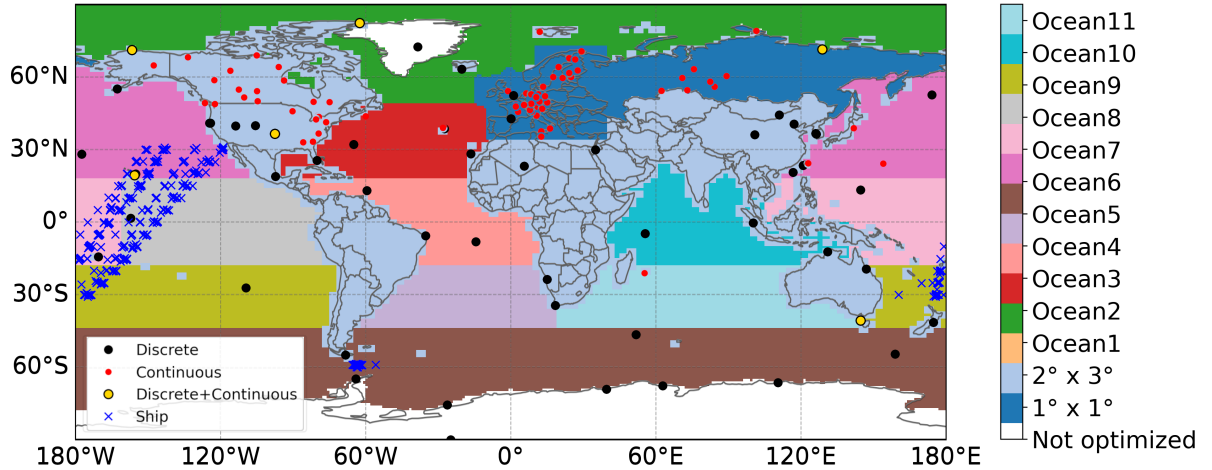
**Figure 5.** Monthly mean anthropogenic and wetland CH<sub>4</sub> emissions at 30° latitudinal bands and global totals. The means are taken from 2016–2019. Shaded areas illustrate uncertainty as standard deviations of 500 ensemble members.



**Figure 6.** Prior total CH<sub>4</sub> emissions (left top) and emission increments (posterior - prior), averaged over 2016–2019. Positive values indicate posterior emissions being higher than the prior.

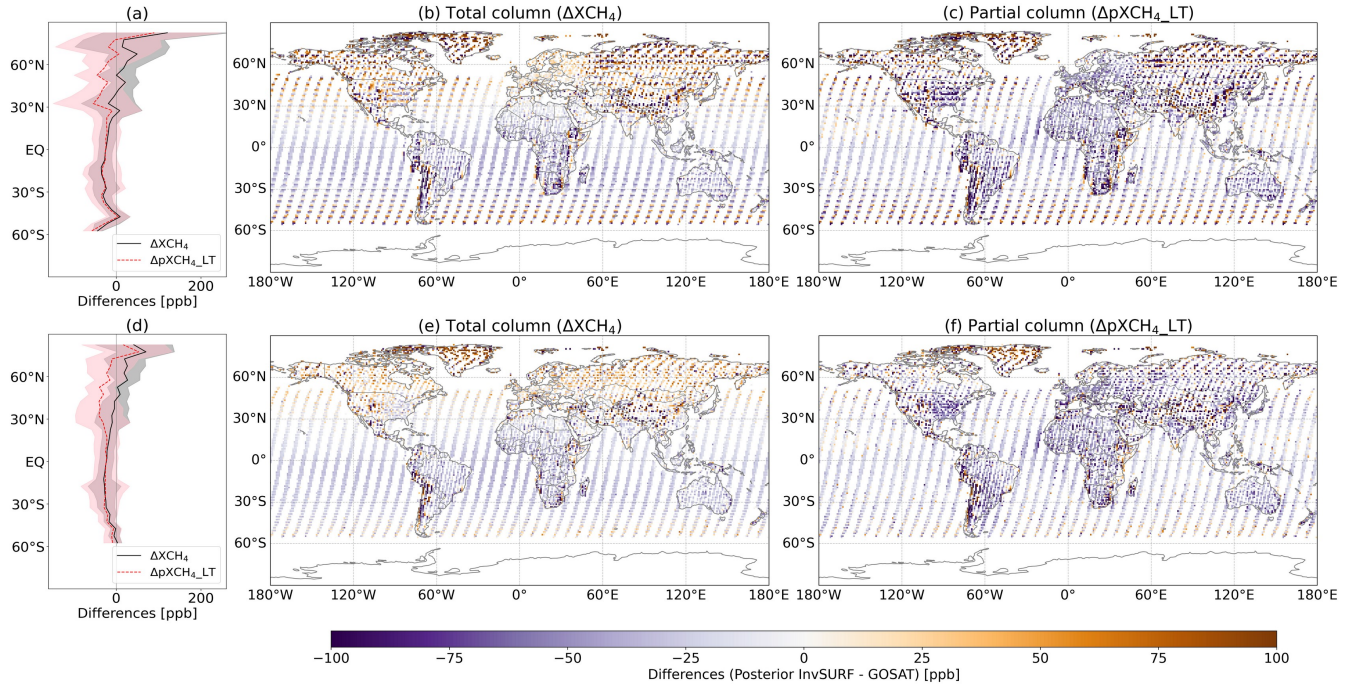


**Figure 7.** Annual mean uncertainty reduction rate  $(\sigma_{\text{prior}} - \sigma_{\text{posterior}}) / \sigma_{\text{prior}}$  of total fluxes, averaged over 2016–2019.  $\sigma$  were calculated as standard deviation from 500 ensemble members. Note the colour is in logarithmic scale.

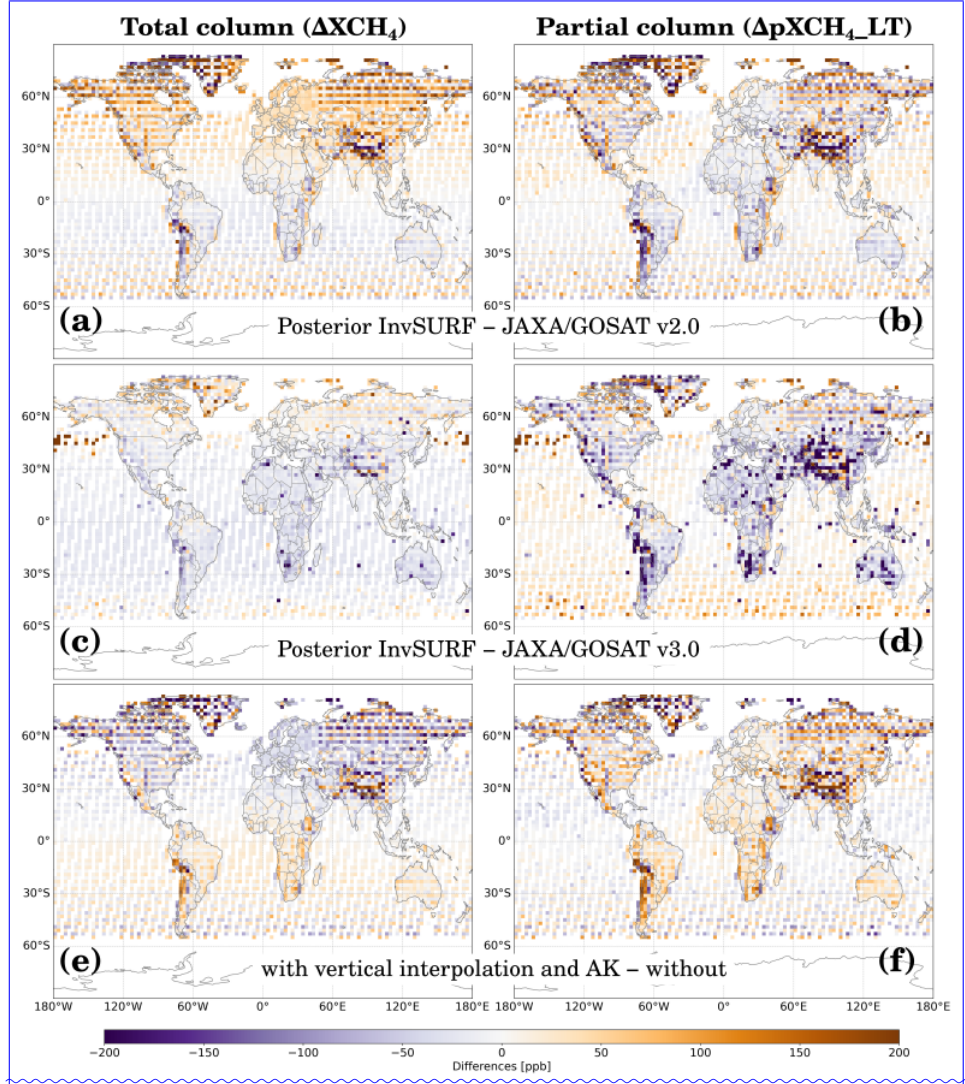


**Figure A1.** Location of ground-based surface observations assimilated in InvSURF (dots and x-marks) assimilated in the InvSURF inversion experiment (see section 2.4), and optimization regions used in CTE-CH<sub>4</sub> (background colours). Over land areas, the fluxes were optimized grid-wise, and ocean region-wise.

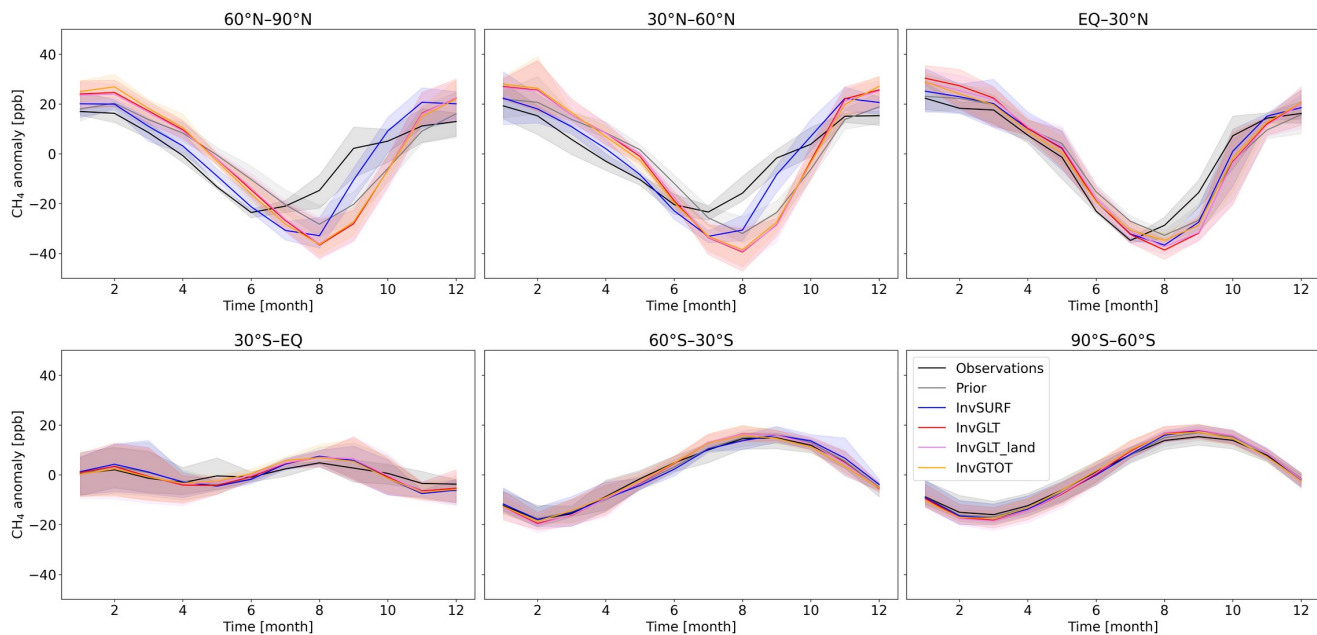




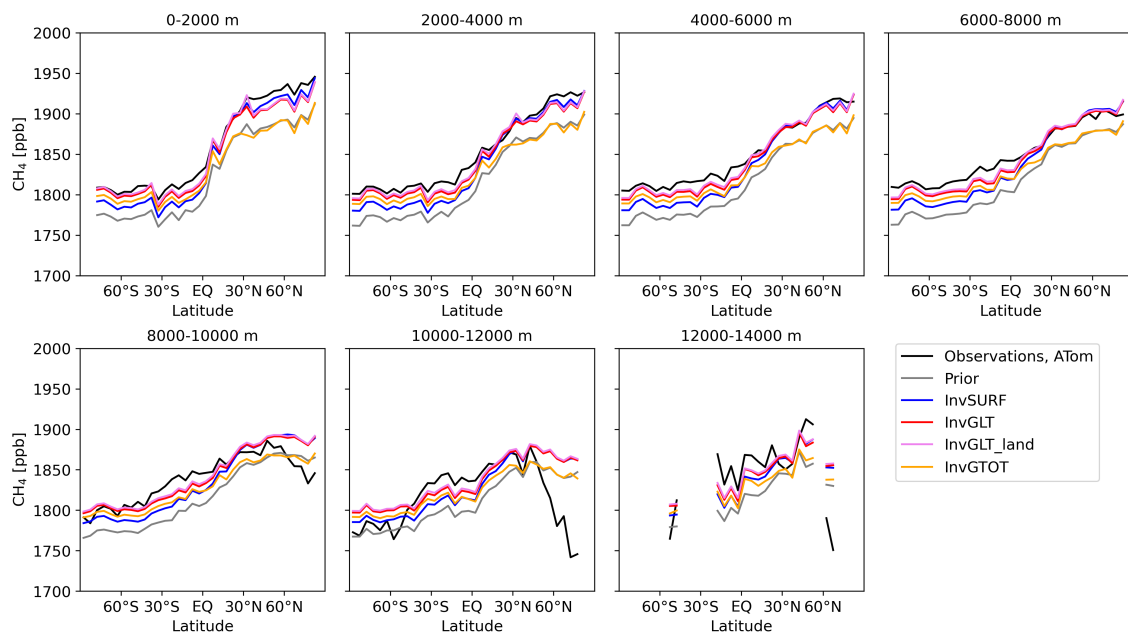
**Figure A2.** Mean differences between prior mole fractions and JAXA/GOSAT retrievals, averaged over 2016–2019. (a) and (d) five degree latitudinal means (solid line) with standard deviation (shaded area), and (b), (c), (e) and (f)  $1^\circ \times 1^\circ$  grid means. (a), (b) and (c) were calculated using TM5 with  $1^\circ \times 1^\circ$  zoom over Europe and  $6^\circ \times 4^\circ$  globally, and (d), (e) and (f) using  $2^\circ \times 3^\circ$  (latitude × longitude) globally. Positive values indicate posterior InvSURF mole fractions being higher than the GOSAT retrievals.



**Figure A3.** (a)-(d) Mean differences between posterior InvSURF and JAXA/GOSAT retrievals averaged over 2016 and at  $2^\circ \times 3^\circ$  grids. Positive values indicate posterior InvSURF mole fractions being higher than the JAXA/GOSAT retrievals. (a) and (b) compared to v2.0, without averaging kernels and vertical interpolation. (c) and (d) compared to v3.0 with vertical interpolation and averaging kernels applied. (e) and (f) are comparisons of modelled values between those calculated with vertical interpolation and AK (i.e. posterior InvSURF values from (c) and (d)) and those without interpolation and AK (i.e. posterior InvSURF values from (a) and (b)), illustrating the effect of interpolation and AK directly. Positive values indicate higher mole fraction values with interpolation and AK. (a), (c), (e) are comparisons of total columns and (b), (d), (f) are of lower tropospheric partial columns.

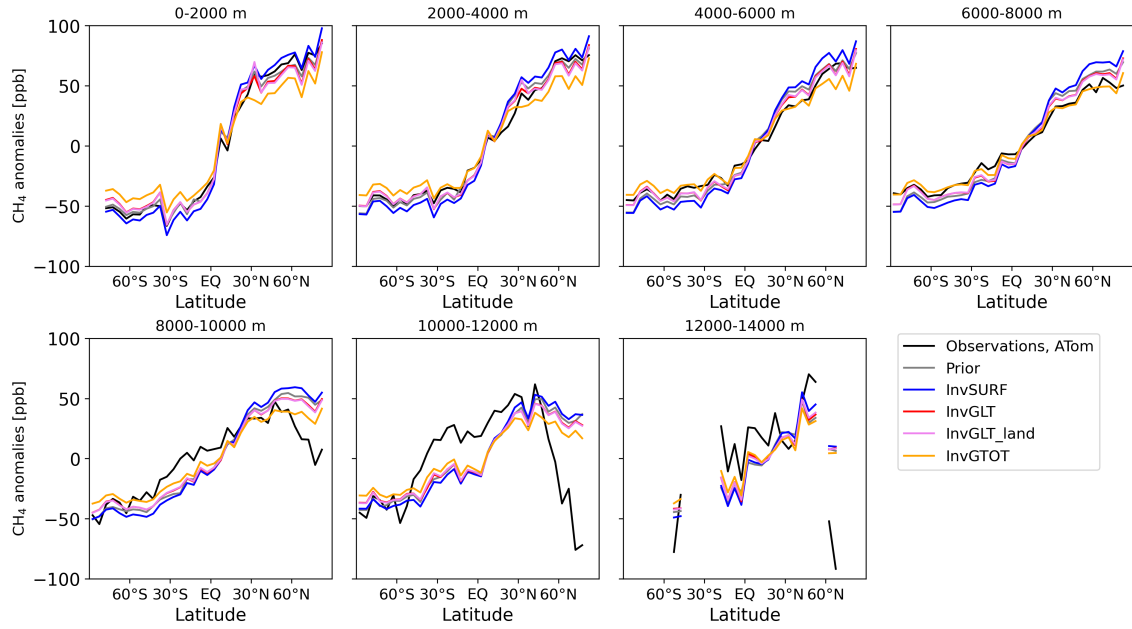


**Figure A4.** Detrended monthly average mole fractions at surface stations, assimilated in InvSURF. The data is averaged from 2016–2019 and at 30° latitudinal bands. Shaded areas are minimum and maximum of detrended monthly values within 2016–2019.

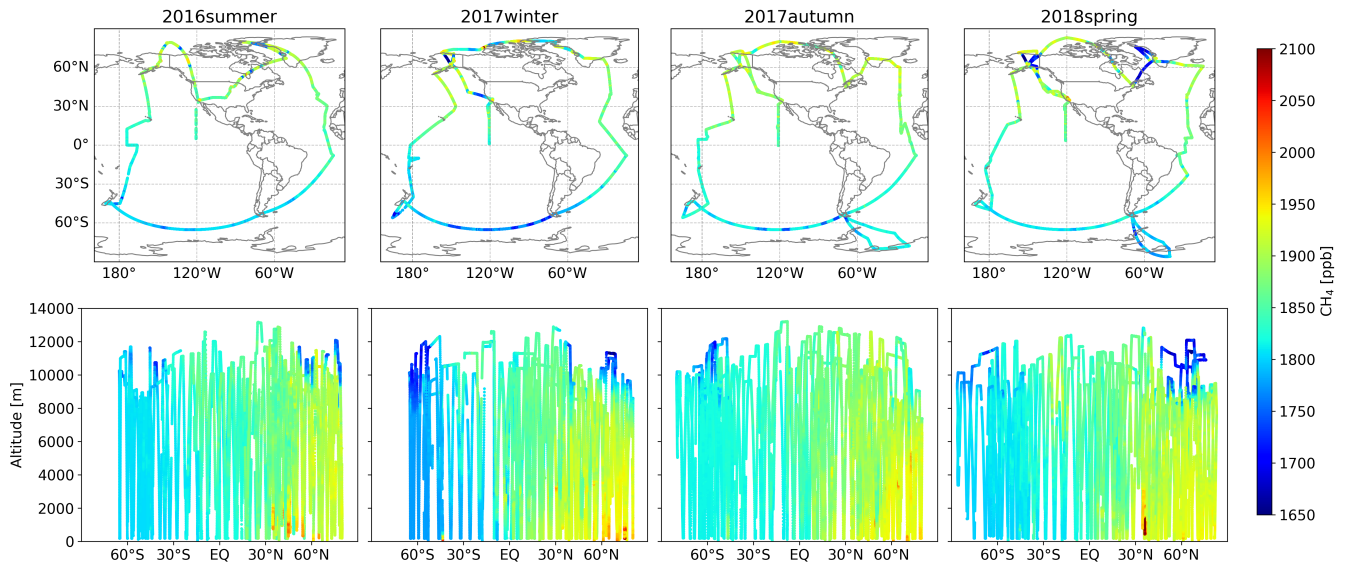


**Figure A5.** Mean atmospheric CH<sub>4</sub> at location and time of aircraft measurements from Atmospheric Tomography Mission, averaged over five degree latitude bands and 2000 m altitude bands above sea level during 2016–2019.

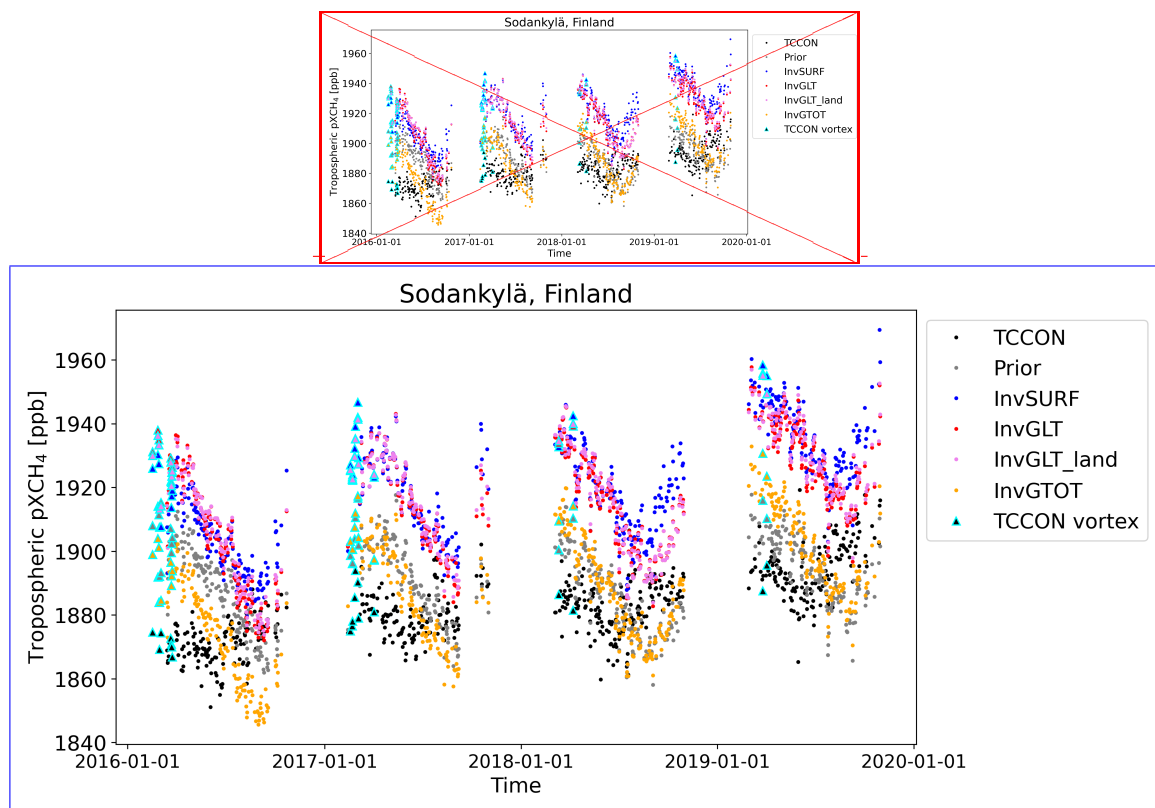




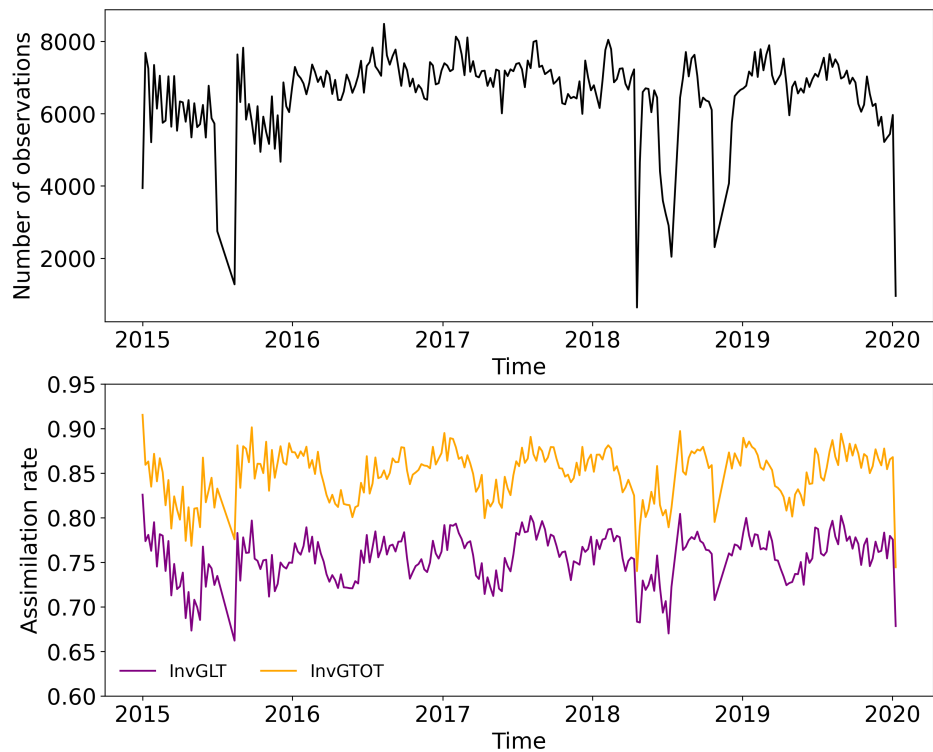
**Figure A6.** As in A5, but means were subtracted at each altitude bands.



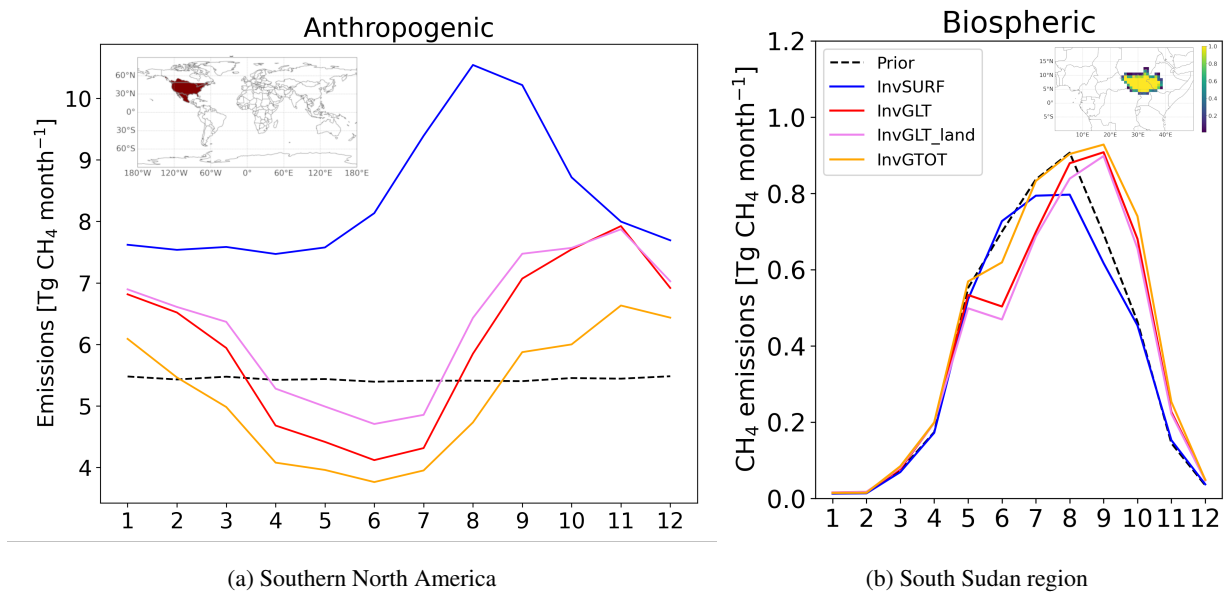
**Figure A7.** Observed  $\text{CH}_4$  mole fractions from aircraft measurements of Atmospheric Tomography Mission during the study period.



**Figure A8.** Daily averaged tropospheric partial columns at the Sodankylä TCCON station, Finland.



**Figure A9.** Global total number of JAXA/GOSAT observations per week and assimilation rates in GOSAT inversions. The x-axis is weeks of optimization steps, and not the actual time of observation.



**Figure A10.** Average monthly  $\text{CH}_4$  emissions in (a) southern North America and (b) South Sudan regions. The maps illustrate aggregated areas.