Atmospheric data support a multi-decadal shift in the global methane budget towards natural tropical emissions

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Abstract. We use the GEOS-Chem global 3-D model and two inverse methods (the Maximum *A Posteriori* and Ensemble Kalman Filter) to infer regional methane (CH_4) emissions and the corresponding ~~carbon stable~~ stable carbon isotope source signatures, 2004–2020, across the globe using *in situ* and satellite remote sensing data. Over our 17-year study period, we find ~~consistent evidence from both atmospheric CH_4 datasets of~~ using the *in situ* data a progressive increase of CH_4 emissions ~~at tropical (30N to 30S)latitudes (+3.80~~ from tropical continental regions (3.6 Tg/yr/yr), including North Africa, tropical South America, and Tropical Asia. The second largest increase in CH_4 emissions over this period (1.6 Tg/yr/yr) ~~,~~ is from China. For Boreal regions we estimate a net decrease of -0.2 Tg/yr/yr and for northern and southern temperate regions we estimate trends of 0.03 Tg/yr/yr and 0.2 Tg/yr/yr, respectively. The increase in tropical CH_4 emissions is accompanied by a progressively isotopically lighter atmospheric δ^{13}C signature ~~, consistent with increasing biogenic wetland emissions. The highly-resolved satellite~~ from the tropics, particularly since 2012, which is consistent with an increased biogenic emission source. Over China, we find a weaker trend towards isotopically lighter δ^{13}C sources, suggesting that heavier isotopic source signatures play a larger contribution to this region. Satellite remote sensing data provide ~~evidence of~~ additional evidence of emission hotspots of CH_4 that are consistent with the location and seasonal timing of wetland emissions~~, limiting the hypothesis about the hydroxyl radical (OH) sink for CH_4 playing a significant role in observed global growth in atmospheric~~. The collective evidence suggests that increases in tropical CH_4 ~~. We find that since 2004, the largest growing regional contributions (2004–2020) are from North Africa (+19.9 Tg/yr), China (+21.6 Tg/yr), and Tropical South America (+14.2 Tg/yr). To quantify~~ emissions are from biogenic sources, with a significant fraction from wetlands. To understand the influence of our results to changes in ~~OH~~the oxidation of atmospheric CH_4 by the hydroxyl radical (OH), we also report regional CH_4 emission estimates using an alternative scenario of a 0.5%/yr decrease in OH since 2004, followed by a ~~5~~larger 1.5% drop in 2020 during the first COVID-19 lockdown. We find that our main findings are robust against those idealised year-to-year changes in OH~~.~~, although they are inconsistent with global-scale constraints on estimates of the global mean atmospheric growth of atmospheric CH_4.

1 Introduction

Changes in atmospheric methane (CH_4) over the last few decades have unfolded without clear explanation, exposing inadequacies in our measurement coverage and our ability to definitively attribute those changes to individual emissions and losses. The climatic importance of atmospheric CH_4 lies in its ability to absorb and emit infrared radiation at wavelengths that are relevant to outgoing terrestrial radiation and incoming shortwave radiation (Allen et al., 2023). Consequently, atmospheric CH_4 helps to maintain Earth's radiative balance and surface and atmospheric temperatures. Atmospheric CH_4 is derived from emissions due to thermogenic (organic matter broken down at high temperatures and pressures, mainly released during extraction and transport of fossil fuels), pyrogenic (through incomplete combustion of organic matter), and biogenic (microbial activity) based production pathways. The main loss process is from from the hydroxyl radical (OH), with minor losses from the reaction with chlorine, uptake from soils, and stratospheric loss. ~~CH_4~~ Methane is the second most abundant anthropogenic greenhouse gas ~~and has a GWP_{100} value of 28 (Masson-Delmotte et al., 2021), a widely used metric that estimates, for a given mass emission, the influence on capturing heat in the atmosphere over 100 years. A mass emission of CH_4 is therefore 28 times more powerful at capturing heat in the atmosphere than an equivalent mass emission of carbon dioxide~~ in terms of its anthropogenic radiative forcing. The global CH_4 growth rate was close to zero from 2000 to 2006 (Dlugokencky et al., 2020) but has since accelerated, with ~~global yearly~~ a global annual growth rate reported by NOAA exceeding 15 ppb for the first time in 2020 ~~(?)~~and more than 18 ppb in 2021 (Feng et al., 2023). Concurrently, we are witnessing a progressively isotopically lighter signature of global averaged CH_4 (more negative global average atmospheric $\delta^{13}C$ value)~~, possibly indicative of changing contributions of emissions sources , for example an increase in biogenic sources or a decrease in thermogenic sources (Lan et al., 2021~~. Analysis of CH_4 mole fraction and $\delta^{13}C$-CH_4 data suggest that thermogenic sources are unlikely to be the dominant driver of the post-2006 global mean increase in atmospheric CH_4 (Lan et al., 2021)). A growing body of work has proposed a range of hypotheses to explain short periods of observed global and regional variations in atmospheric CH_4 (Turner et al., 2019). In this study, we take a step back to look at observed CH_4 variations from 2004 to 2020, in order to capture the some of the zero-growth rate period and the subsequent increase in growth rate of CH_4 post-2007. We argue that monthly variations are part of a large-scale shift ~~of predominately thermogenic energy emissions from high northern latitudes~~ from predominately thermogenic emissions from northern midlatitudes to biogenic emissions from the tropics, driven by ~~Tropical North African and Tropical South American wetlands~~larger emissions over tropical North Africa and tropical South America.

The post-2007 increase in atmospheric CH_4 has been the focus of many studies and has been attributed to different plausible hypotheses associated with changes in various emissions sources, and the OH sink (Turner et al., 2019). These studies have reached their conclusions using *in situ* mole fraction observations alone or in combination with other observations, e.g. *in situ* $\delta^{13}C$ (Schaefer et al., 2016; Rice et al., 2016; Nisbet et al., 2016; Fujita et al., 2020; Lan et al., 2021; Basu et al., 2022), satellite observations (Worden et al., 2017; McNorton et al., 2018; Yin et al., 2021; Feng et al., 2022), or other trace gases, using a variety of analysis methods and computational models. Typical emissions sizes and uncertainty are indicated in Table 1, adapted from Saunois et al. (2020). Our approach is unique in that, for our $\delta^{13}C$ inversion, we are solving for the $\delta^{13}C$ isotopic source signature of a geographical region. From the isotopic source signature of a region, we can determine how the source

balance within a ~~particular~~ region has shifted over time~~(i.e., towards more pyrogenic, thermogenic or biogenic sources), and so~~, e.g. larger or smaller contributions from pyrogenic and biogenic sources, and consequently gain understanding of the geographical shifts in the CH$_4$ budget.

~~The reaction with~~ Methane oxidation by the OH radical in the troposphere is ~~the largest sink of CH$_4$,~~ responsible for 80% of the total CH$_4$ sink globally. Changes in OH may have played a role in recent changes in atmospheric CH$_4$ (Rigby et al., 2017; Turner et al., 2017) but the magnitude of this influence is uncertain (its short atmospheric lifetime of <1 s makes direct measurement of global variability very difficult). Reducing values of OH effectively increases atmospheric CH$_4$ and therefore has the same effect as increasing emissions of CH$_4$. Chemical reactions responsible for removing CH$_4$ from the atmosphere are faster for lighter isotopologues of CH$_4$. This isotopic fractionation therefore leads to an atmosphere enriched in heavier isotopes relative to the globally emitted CH$_4$. Lan et al. (2021) simulated CH$_4$ and δ^{13}C in a 3-D chemistry transport model covering the period 1984-2016, and found that changes in OH proposed by ~~(Turner et al., 2017)~~ Turner et al. (2017) are not consistent with the trend of increasingly isotopically light δ^{13}C observed in the atmospheric record. We explore the impact of reducing OH in a sensitivity study~~. The first COVID-19 lockdown in~~, taking into account a larger OH decrease during 2020 ~~corresponded to an unexpected large increase in atmospheric CH$_4$. Studies have suggested this could be partly explained by a 3-5% reduction in OH (Miyazaki et al., 2021; Laughner et al., 2021) resulting from a large-scale reduced emissions of nitrogen oxides associated with industry. This has yet to be corroborated by satellite data that provide complementary constraints on the key emitting regions over the tropics (Feng et al., 2022).~~ (Peng et al., 2022; Feng et al., 2023) that was associated with widespread redutions in nitrogen oxide emissions (Cooper et al., 2022).

Here, we calculate trends in regional CH$_4$ emissions and isotopic δ^{13}C source signatures across the world, ~~2004-2020~~2004–2020, using *in situ* mole fraction and δ^{13}C data, and satellite column-averaged dry-air mole fraction data. This is achieved by using three sets of inversions: two Maximum A-Posteriori inversions using ground-based data (solving separately for regional emissions and isotopic sources signatures), and an Ensemble Kalman Filter inversion using GOSAT data (solving for regional CH$_4$ emissions).

In the next section, we describe the data and methods we use to quantify changes in regional CH$_4$ emissions and the corresponding regional stable isotope source signatures. In section 3, we report our results of *a posteriori* regional CH$_4$ fluxes and regional δ^{13}C isotopic signatures, including analysis of sensitivity calculations that involve different assumptions about year to year changes in the OH sink. We conclude the paper in section 4.

2 Data and Methods

2.1 *In Situ* and Satellite Remote Measurements of Atmospheric Methane

We use surface-level flask data as constraints on both regional CH$_4$ emissions and δ^{13}C regional CH$_4$ emissions isotopic source signatures. The CH$_4$ mole fraction data are taken from 31 National Oceanic and Atmosphere Administration – Global Monitoring Laboratory (NOAA-GML) sites around the world (Figure 1), version 2020-07 (Dlugokencky et al., 2020). The data are monthly mean values, averaged from discrete data as collected at each site, analysed at NOAA-ESRL in Boulder, Colorado,

and recorded to the NOAA 2004A standard scale (Dlugokencky et al., 2005). Up to August 2019, the analysis was performed
using gas chromatography (Steele et al., 1987, Dlugokency et al., 1994; Dlugokencky et al., 2005) and since August 2019,
cavity ringdown spectroscopy has been used (Dlugokencky et al., 2020). We also include data from a site in Siberia, Karasevoe
(KRS), which is monitored by the National Institute for Environment Studies (NIES). This site was included to maximise
geographical coverage of *in situ* data. The CH_4 mole fraction measurements from this site are continuous, measuring from
65 m height, covering the period ~~2004-2020~~ 2004–2020 (Sasakawa et al., 2010). A scale factor of 0.997 is applied to the NIES
data in order to bring it into line with the NOAA 2004A scale (Zhou et al., 2009). The site constitutes part of the Japan-Russia
Siberia Tall Tower Inland Observation Network (JR-STATION).

$\delta^{13}C$ data are similarly monthly mean values, calculated from discrete flask samples at NOAA network sites, reported on
the international carbon isotope scale VPDB (Vienna Pee Dee Belemnite). Isotope ratio 'delta' values represent the excess
of a heavy, less abundant stable isotope (for $\delta^{13}C$ values, carbon-13) over the light, most abundant stable isotope (carbon-
12) in a sample, when compared to a standard. These measurements are useful as they are indicative of the source of the
CH_4: biogenic sources are dominated by isotopically lighter signatures and thermogenic sources are dominated by isotopically
heavier signatures. For the NOAA network, isotopic analysis of $\delta^{13}C$ was performed at the University of Colorado Institute
of Arctic and Alpine Research Stable Isotope Laboratory (CU-INSTAAR). They follow an isotope ratio mass spectrometry
approach (Miller, 2002; Vaughn et al., 2004). The geographical locations of *in situ* measurement sites are shown in Figure 1.
These sites are a subset of the entire NOAA network's capacity for measuring CH_4 mole fractions. The sites included in the
inversion (both for CH_4 and $\delta^{13}C$) are those that cover the entire period of the inversion (2004-2020) without significant periods
of measurement breaks to ensure a consistent interpretation of trends without consideration of possible biases introduced
through the inclusion or exclusion of specific sites.

We also estimate CH_4 fluxes for 2010-2020 from the Japanese Greenhouse gases Observing SATellite (GOSAT) that was
launched in 2009. GOSAT is in a sun-synchronous orbit with an equatorial local overpass time of 13:30. Since launch, it has
provided continuous global observations of dry-air atmospheric column-averaged CO_2 (XCO_2) and CH_4 (XCH_4), retrieved
from shortwave infrared wavelengths that are most sensitive to changes in CH_4 and CO_2 in the lower troposphere (Parker
et al., 2020). We use the latest (v9) proxy XCH_4:XCO_2 retrievals that use spectral absorption features around the wavelength
of 1.6 μm (Parker et al., 2020, Palmer et al., 2021), because of the smaller bias and better global coverage than those provided
by the full physics retrievals. Analyses show the precision of single proxy retrieval is about 0.72%, with a global bias of
0.2% (Parker et al., 2011, 2015, 2020). In our calculations, we assume a higher observation uncertainty of 1.2%, and deduct
a globally uniform bias of 0.3% to obtain better *a posteriori* agreement with the independent ground-based XCH_4 data by
the Total Carbon Column Observing Network (TCCON). These uncertainties are detailed in Feng et al. (2022). To anchor the
constraints from the proxy XCH_4:XCO_2 ratio (Fraser et al., 2014; Feng et al., 2017), we also assimilate the GLOBALVIEW
CH_4 and CO_2 data (Schuldt et al., 2021), with assumed uncertainties of 0.5 ppm and 8 ppb for *in situ* measurements of CO_2 and
CH_4, respectively. GLOBALVIEW constitutes a combination of CH_4 data from ground-based data (both flask and continuous)
and aircraft data, from 54 different laboratories, combined and published by NOAA-GML (Schuldt et al., 2021). Locations of
the assimilated GLOBALVIEW CH_4 (sub) dataset are shown in Feng et al., 2022.

2.2 GEOS-Chem Atmospheric Chemistry and Transport Model

125 To relate CH_4 emissions to atmospheric CH_4 concentrations, we use v12.1 of the GEOS-Chem 3-D global chemical transport model (CTM) (Bey et al., 2001) at a horizontal resolution of 2° (latitude) by 2.5° (longitude) with 47 vertical levels from the surface to 80 km height, with meteorological data from the MERRA-2 meteorological reanalyses (Gelaro et al., 2017) from the NASA Global Modeling and Assimilation Office (GMAO).

Our *a priori* emissions include: 1) monthly EDGAR ~~v4.3.2 anthropogenic emissions (Janssens-Maenhout et al., 2019)~~ v6
130 anthropogenic emissions (Crippa et al., 2021) that accounts for emissions from oil and gas, coal, livestock, landfills, wastewater, rice, and other anthropogenic sources (including biofuel) from 2004 to 2018; 2) monthly GFED-4 biomass burning emissions (version 4.1; Randerson et al., 2017); and 3) monthly v1.0 WetCHARTs wetland emissions (Bloom et al., 2017). The Harvard-NASA Emissions COmponent (HEMCO) software within GEOS-Chem converts the emission inventories at their native horizontal resolution to the GEOS-Chem 2° × 2.5° resolution. Beyond the end of the emissions inventory, emissions are
135 repeated yearly in *a priori* simulation.

Table 1 shows the $\delta^{13}C$ ~~isotopic source~~ signatures for the source types included in our simulations. These are extracted as mean global values from Sherwood et al. (2017), which provide a database of global isotopic source signatures that are broken down into the same sectors as we employed in our simulations. However, individual source types show a wide range of source signatures, and this uncertainty is reflected in the assigned uncertainty given to the *a priori* source signatures in inversion
140 (Section 2.3). ~~We~~ In the inversion, we differentiate between Arctic and tropical wetlands by applying a 10‰ isotopically lighter source signature to the Arctic source (Table 1), following Ganesan et al. (2018) who produced a global wetland source signature map based ~~upon~~ on published $\delta^{13}C$ data. In GEOS-Chem, we simulate isotopologues separately (i.e. for $\delta^{13}C$, $^{12}CH_4$ and $^{13}CH_4$), and then calculate $\delta^{13}C$ values. The arithmetic underlying the conversion of isotope ratios to isotopologue emissions for input to the model are detailed in Appendix A.

145 We include the loss of atmospheric CH_4 from reaction with chlorine, soil uptake, and from oxidation by OH. We use monthly 3-D fields of OH, calculated using the full-chemistry version of GEOS-Chem, and monthly 3-D field of atomic chlorine (Sherwen et al., 2016). Stratospheric loss frequency fields are determined using the NASA GMI stratospheric model (Duncan et al., 2007). Estimates of the microbial consumption of CH_4 in soils is determined from Fung et al. (1991). The resulting atmospheric lifetime of ~~CH_4~~ CH4 against OH is ~~9.73 years, consistent with the observed methyl chloroformlifetime~~
150 ~~of 5.39 years. This~~ 9.77 years. The corresponding lifetime for methyl chloroform is 5.41 years, which is consistent with atmospheric observation of methyl chloroform. This lifetime also compares well with ~~multi-model simulations~~ multi-mode (Voulgarakis et al., 2013; Morgenstern et al., 2017) that reported global mean lifetimes of CH_4 that range 7.2-~~10.1~~ –10.1 yrs. In our default model configuration, none of these loss processes include interannual variations.

To account for isotopic fractionation due to loss of CH_4 in the troposphere and stratosphere, we use published kinetic
155 isotope effect values (KIEs). These values are employed to scale the reaction rate constants used in the simulations for $^{12}CH_4$ and $^{13}CH_4$ (Table A1). The OH and Cl sinks are handled in the hard coding of the model, whereas the soil sink is handled as a

negative emission in the HEMCO file. Therefore, for the soil sink, the KIE is directly applied as a scale factor in the HEMCO configuration file (Snover and Quay, 2000; Burkholder et al., 2019).

~~'Spinning up' is an important aspect of atmospheric modelling in order for simulated mole fractions to reach equilibrium.~~ ~~We spin-up the model by scaling a~~ We created the initial conditions for atmospheric CH_4 by first scaling a standard CH_4 ~~restart~~ GEOS-Chem restart file (a file ~~from a previous GEOS-Chem model run. The restart file is at 4x5resolution, and originally represented 2012, but is spun up to represent 2004. The $\delta^{13}C$ inversion uses a posterioriregional emissions from the~~ containing a default realistic distribution of CH_4 ~~inversion as a starting point, with sectoral emissions scaled as detailed in Appendix A. We then run the model over the year 2004 sixty times using the~~ across the atmosphere) to conditions near representative of the start of our analysis in January 2004. We then ran the model sixty times with repeating 2004 MERRA-2 meteorology and emissions ~~,~~ (corresponding to approximately six ~~times the chemical lifetime of~~ e-folding lifetimes for CH_4~~. We find this is sufficient to allow mole fractions and isotope ratios to equilibrate (not shown)~~) to improve as far as possible the simulation of atmospheric gradients in CH4 in the initial conditions. We then ~~run~~ ran a single-year inversion for 2004 to optimise the $\delta^{13}C$ isotope ratios and CH_4 ~~values~~ concentrations relative to ground-based observations, following the inverse method detailed below. The $\delta^{13}C$ inversion used the regional emissions estimate provided by the posteriori from the CH_4 inversion as a starting point, with sectoral emissions scaled as detailed in Appendix A. The output of this ~~short inversion is improved estimates of initial conditions for $\delta^{13}C$ and CH_4~~ 2004 inversion is a final step in creation of the initial conditions, which serve as a starting point for the longer inversion that we report here (~~2004-2020). This restart file originally represented 2012, however it is spun up over 60 years to the point where it no longer resembles 2012 and will be representative of 2004, the starting point of the simulation.~~ 2004- 2020).

For all our calculations, we sample GEOS-Chem at the grid box and local time that corresponds to the *in situ* and satellite remote sensing data. For the satellite data, we also apply scene-dependent averaging kernels to account for vertical structure. This approach allows us to directly compare the model with measurements. Regional trends are calculated by examining the grid boxes ~~which correspond to~~ encompassed by a given region on the global grid.

2.3 Inverse Methods

We use two inverse methods that reflect the volume and simplicity of the data being used. For *in situ* data we use the Maximum *A Posteriori* (MAP) inverse methods and for the more voluminous satellite data we use an ensemble Kalman filter (EnKF). For brevity, we include only the essential details about either method and refer the reader to dedicated papers.

We use the Siegel linear non-parametric estimates (Siegel, 1980) to fit a line to our *a posteriori* CH_4 emissions. This method is less sensitive to outliers, e.g. El Niño, that would otherwise compromise the linear trend estimate (Palmer et al., 2021), and the resulting linear trend estiate has lower variables that simpler methods. We find Siegel trend estimates are similar to those estimated by the Theil-Sen estimator.

2.3.1 Maximum *A Posteriori*

To infer regional *a posteriori* CH$_4$ fluxes and regional δ^{13}C emissions source signatures from the atmospheric measurements of CH$_4$, we use the Maximum *A Posteriori* solution (MAP) inverse method (Rodgers, 2000). We solve for CH$_4$ fluxes and δ^{13}C emissions signatures from 14 geographical regions (Figure 1). This method combines *a priori* knowledge and its uncertainty with the measurements and their uncertainties, and has been used in a number of studies, e.g., Fraser et al. (2014); McNorton et al. (2018).

The MAP solution and the associated *a posteriori* uncertainty is described as, respectively:

$$\mathbf{x}^a = \mathbf{x}^b + (\mathbf{H}^T\mathbf{B}^{-1}\mathbf{H} + \mathbf{R}^{-1})^{-1}\mathbf{H}^T\mathbf{B}^{-1}(\mathbf{y} - \mathbf{H}\mathbf{x}^b), \tag{1}$$

$$\mathbf{A} = (\mathbf{H}^T\mathbf{B}^{-1}\mathbf{H} + \mathbf{R}^{-1})^{-1}, \tag{2}$$

using the ~~conventional~~ convention that lower-case and upper-case variables denote vectors and matrices, ~~where~~ respectively. The $\mathbf{x}$ denotes the state vector that describes the estimated quantities, which in this study includes monthly CH$_4$ fluxes and δ^{13}C source signatures from regions across the world (Figure 1). Subscripts 'a' and 'b' denote *a posteriori* and *a priori* CH$_4$ fluxes, respectively, and superscripts '-1' and 'T' denote matrix inverse and transpose operations, respectively. The measurement vector $\mathbf{y}$ includes CH$_4$ mole fraction or δ^{13}C data. The matrices $\mathbf{B}$, $\mathbf{A}$, and $\mathbf{R}$ denote the error covariances matrices for the *a priori*, *a posteriori*, and measurements, respectively. $\mathbf{B}$ and $\mathbf{R}$ are diagonal matrices. For $\mathbf{B}$ we assume uncertainties of 50% of the regional CH$_4$ fluxes and 15‰ for the δ^{13}C values, and for $\mathbf{R}$ we assume 10 ppb for the mole fraction data and 0.1‰ for the isotope data. These uncertainties were based ~~upon~~ on similar studies (Fraser et al., 2014; McNorton et al., 2016). We assume a model transport error of 12 ppb, following Feng et al. (2022).

The Jacobian matrix $\mathbf{H}$ describes the sensitivity of the measurements to changes in the state vector, i.e. $\partial\mathbf{y}/\partial\mathbf{x}$. For the mole fraction CH$_4$ inversion, the Jacobian matrix describes the sensitivity of mole fractions in the model to changes in regional CH$_4$ emissions. We construct the matrix using a series of GEOS-Chem model runs. We systematically let each individual emitting region (described by the state vector) emit for one month while all other regions are emitting as normal. The individual regional source is then switched off (emissions set to zero) and the effect of this on the 3-D atmospheric distribution of CH$_4$ mole fractions is recorded over the following three months. The result of this test is recorded at the grid boxes that correspond to the location of the measurement sites. The resulting mole fractions therefore describe the sensitivity of a particular measurement site to changes in a specific regional source up to three months after emission. This is repeated for every month within the inversion timescale, for every region described in the state vector.

For the δ^{13}C inversions, the ~~a priorisimulation uses a posterioriregional emissions from the CH$_4$ inversion as a starting point. The Jacobian matrix in this case~~ Jacobian matrix describes the sensitivity of modelled δ^{13}C to changes in the regional isotopic source signatures. We construct the Jacobian as the difference between a control model calculation (using the CH$_4$ *a posteriori* regional emissions and mean source signature values from Sherwood et al. (2017)) and perturbed source signature model calculation for the whole study period (2004-2020). For the perturbed model calculation, we systematically perturb the isotopic source signature of each region (all of the sectors that are ~~containing~~ contained geographically within a region) isotopically heavier by 20‰ for the period 2004-2020. The difference between the control and perturbed run in δ^{13}C value

at the location of each measurement site is then divided by the value of $\delta^{13}C$ perturbation for the region source signature, to understand the effect of changing a regions source signature ~~upon~~ on the $\delta^{13}C$ value recorded at each measurement site location. ~~Each individual regions' model calculation is spun up separately from the control model calculation in order to account for lagging in the model.~~

The output from the inversion are improved estimates of regional CH_4 fluxes and $\delta^{13}C$ source signatures. The model simulates the global atmosphere on a $2° \times 2.5°$ horizontal grid. The *a posteriori* regional CH_4 fluxes and isotopic source signatures are applied to the grid boxes in the model which correspond to a given region in an *a posteriori* simulation. ~~The a posteriorisimulation is then used to compare with the in situdata to ensure the quality of the a posteriorifit.~~

2.3.2 Ensemble Kalman Filter

We use an Ensemble Kalman Filter (EnKF) approach in performing the inversion using satellite data, because we cannot easily evaluate the necessary matrix operations associated with an analytic inversion. Here we use an ensemble of flux perturbation pulses to represent uncertainty in our *a priori* estimate for regional monthly CH_4 fluxes. We subsequently use a global chemistry transport model (i.e., the GEOS-Chem v12) to track the transport and chemistry processes of the tagged emission pulses in the atmosphere, to project their spreads to the observation space. With the ensemble of *a priori* flux perturbations, and the simulated observation impacts, we use the Ensemble Transform Kalman Filter (ETKF) algorithm to numerically estimate the *a posteriori* CH_4 fluxes and the associated uncertainties by optimally comparing the model simulation with observations (see Feng et al., 2017 for more details). To reduce the computational costs, mainly from tracking tagged emission pulses, we introduce a 4-month moving lag window for each assimilation step, because any observation has limited ability to distinguish between the signals emitted long (>4 months) before, from variations in the ambient background atmosphere (Feng et al., 2017). As a result, we are able to include a larger state vector, consisting of monthly scaling factors for 487 (476 land regions and 11 oceanic regions) regional CH_4 (and CO_2) pulse-like basis functions (Figure S1 in ~~(Feng et al., 2022)~~Feng et al. (2022)). We define these land sub-regions by dividing the 11 TransCom-3 (Gurney et al., 2002) land regions into 42 to 56 nearly equal sub-regions, and use the 11 oceanic regions defined by the TransCom-3 experiment. Because of their smaller sizes, we have assumed a higher uncertainty percentage (60%) for *a priori* emissions than the MAP approach described above. We also include spatial correlation with a correlation length of ~~500km~~500 km between the sub-regions.

2.4 Sensitivity of Results to Changes in Assumed OH Distributions

To examine the sensitivity of our results to changes in ~~assumed OHdistributions~~the magnitude of OH, we run a single sensitivity run that is made up of two parts. First, we imposed a 0.5%/yr uniform decrease to our 3-D OH field from 2004 to 2019, ~~following similar trends proposed by Turner et al. (2017) (who proposed a~~consistent with the 7% reduction ~~in OH, 2003-2016) and second, we uniformly decrease OH by 5~~over 2003–2016 proposed by Turner et al. (2017). Second, we imposed a larger global-scale OH reduction of 1.5% in 2020 ~~to describe estimated changes due to a global-scale reduction in emissions of~~based on recent studies (Miyazaki et al., 2021; Laughner et al., 2021) to describe a more abrupt change due to widespread reductions in nitrogen oxides (NO_x) associated with ~~the first COVID-19 lockdown Miyazaki et al. (2021); Laughner et al. (2021). It has~~

also been suggested that OH mole fractions may have actually increased during 2000–2016 due to increasing water vapour and NO$_x$ in the tropics (Zhao et al., 2019), however considering the scenario under COVID-19 lockdowns in closing down manufacturing during the first Covid-19 lockdown (Cooper et al., 2022). Newer studies have suggested the OH reduction in 2020 , a decreasing trend over the previous years is only considered here. A similar approach to this was followed by Feng et al. (2022) for which there is some opposition that suggests the change in OH during COVID-19 should be larger, although there is no empirical determination of this change. We then recalculate was closer to 1% (Peng et al., 2022; Feng et al., 2023) , but these estimates are also subject to uncertainties. The purpose of this numerical experiment is to determine the sensitivity of *a posteriori* CH$_4$ flux estimates to changes in assumed variations in OH and not to determine a distribution of OH that simultaneously fits observed changes in CH$_4$ and δ^{13}C-CH$_4$.

3 Results

Here, we report *a posteriori* estimates for total CH$_4$ emissions inferred from the ground-based data . *in situ* and GOSAT data and then the corresponding *a posteriori* isotopic source signatures for δ^{13}C. We draw comparisons with previous studies throughout this section.

4 ~~Results~~

A posteriori emission estimates of total CH$_4$

Figure 2 shows the annual mean differences in regions between *a priori* emission estimates and *a posteriori* emission estimates for both ground-based (2004–2020) and GOSAT results (absolute 2009–2020). Absolute emissions values are plotted in Figure 3). This indicates the changes from the for completeness.

On a global scale, terrestrial *a prioriposteriori* emissions and allows comparison of the two independent approaches. The inferred from *in situ* and GOSAT data have progressively increased relative to *a priori* emissions are constructed as detailed in Section 2.2. Focusing on values since about 2014. The peak difference is in 2020 (due to interest in emissions changes as a result of the COVID-19 lockdowns) on a global scale, when we find increased emissions relative to *a priori* emissions of 72.0 68.5 ± 35.5 61.5 Tg/yr in 2020 for the *in situ* inversion and 61.5 ± 37.3 Tg/yr higher emissions for the GOSAT inversion. The Global ocean CH$_4$ emissions inferred from *in situ* inversion results indicate that this difference originates from tropical regionssuch as Tropical South America (+13.5 ± 1.9 and GOSAT data support a small negative bias (1-2 Tg/yr) in the *a priori*, which we do not discuss further.

As a zeroth order check of our *a posteriori* emission estimate of total CH$_4$, we compare in Figure 4 the published NOAA atmospheric growth rate of CH$_4$ with our corresponding *a posteriori* atmospheric mole fractions. Generally, we find the *a posteriori* values inferred from *in situ* and GOSAT data are consistent with the overall trend of the changes in the growth rate, with large year-to-year changes that we explain now in terms of regional emission changes.

Changes in the global terrestrial emissions reflects changes from different geographical regions. Differences between *a posteriori* emission estimates inferred from *in situ* and GOSAT data are partly due to differences in the geographic coverage of the datasets. Ground-based data have poorer geographic coverage, particularly over the tropics and the southern hemisphere and satellite data are currently available at most once per day in cloud-free conditions. Using the *in situ* data, we find that the largest *a posteriori* emission increases over the 2004 to 2020, determined by the Siegel linear estimator, are over the tropics (3.6 Tg/yr~~in 2020), North Africa (+15.1 ± 6.8~~ /yr, comprising North Africa, tropical South America, and Tropical Asia), followed by China (1.6 Tg/yr~~, 2020) and China (+17.3 ± 4.4~~ /yr) then by small contributions (individually <0.2 Tg/yr~~, 2020).~~ ~~There are decreases relative to~~) from elsewhere. Table 1 provides an overview our annual mean sector-based *a ~~priori~~posteriori* emissions ~~in 2020 in Temperate North America (-13.3 ± 3.4~~ for 2004–2020. Generally, our values are close to the reported median values and within the range of values reported by Saunois et al. (2020).

Over the tropics, there is broad consistency between GOSAT and *in situ* data (Figure 2) that highlights the negative bias in the *a priori* over Northern Africa (bias of -8.6 Tg/yr), ~~Southern Africa (-5.6 ± 2.1~~ Tropical Asia (bias of -7.2 Tg/yr), ~~Temperate and Tropical~~ South America (~~-4.1 ± 4.0~~ bias of -11.63 Tg/yr)~~and Boreal Eurasia (-2.3 ± 3.9~~. The *in situ* and GOSAT data for China support a small, steady increase in emissions from 2009 to 2020 (1.0 Tg/yr)~~. According to these results, mid-latitudinal emissions are being overestimated and tropical emissions underestimated.~~

~~Likewise, the GOSAT-based inversion results indicate~~, with emissions inferred from the GOSAT data generally smaller than *a ~~posteriori~~priori* ~~emissions increases from the~~ values throughout the period (Figure 2). Data over India have a small mean annual trend (0.33 Tg/yr) over the same period. *a ~~priori~~In situ* ~~emissions are centred around tropical regions. In 2020, there are emissions increases from~~ and GOSAT data are more consistent in sign (but not magnitude) at temperate latitudes (Figure 2). *A posteriori* emissions from *in situ* and GOSAT data are generally lower than the *a priori* ~~estimates in Tropical South America~~ ~~(+20.3 ± 1.9~~ by more than 12.0 Tg/yr ~~) and North Africa (+13.1 ± 6.8~~ and 5.6 Tg/yr~~). Similar to the in situresults, there are decreases in some mid-latitudinal regions, specifically~~, respectively, over Temperate North America~~(-3.9 ± 1.8~~, and higher by more than 13.0 Tg/yr ~~) and Temperate South America (-6.4 ± 7.1~~ and 7.0 Tg/yr, respectively, over Temperate Eurasia, with the smallest discrepancies relative to the *a priori* before 2009. *A posteriori* emissions from boreal regions appear to be larger than *a priori* values by 4.2 Tg/yr ~~).~~

~~The increase in tropical emissions has been highlighted by previous studies, whether using GOSAT data or~~ before 2009 (Figure 2). After 2009, *in situ* data ~~as constraints in a3-D CTM inversion (McNorton et al., 2016 and Fujita et al., 2020, examining (2003-2015) and (1995-2013) respectively). The increase in North Africa is especially noteworthy in 2020, where emissions have been attributed to increased wetland emissions by previous studies (Lunt et al., 2019, 2021; Pandey et al., 2021; Feng et al.,).~~

~~There are some differences between the two inversion results, likely due to the differing geographic coverage of the different datasets (ground-based data has poorer geographic coverage, and satellite data are available only once per day in cloud-free conditions). Specifically, emissions from Boreal North America and China are lower than the~~ become progressively more consistent with the *a priori* over North America and is typically smaller than *a priori* values over Eurasia by 2.6 Tg/yr. For GOSAT, we find the converse situation: after 2009, data are lower than *a priori* ~~emissions for the GOSAT-based inversion~~

320 (-4.6 ± 1.1 and -5.1 ± 3.8 values by 4.4 Tg/yr in 2020 respectively), but increase for the over North America and comparable with *a priori* values over Eurasia. In the southern hemisphere, *in situ* data closely *a priori* values, as expected, since there are fewer places where *in situ* inversion (+4.4 ± 3.6 and +17.3 ± 4.4 data are collected. GOSAT data show a small but persistent increase in emissions with time over Southern Africa (0.41 Tg/yr in 2020 respectively). It is noteworthy that, despite differences in the absolute annual emissions estimates, both the GOSAT-based inversion and), highlighting the negative bias in *a priori*

325 emissions over Australia and over Temperature South America.

We use the *a posteriori* error covariance matrix from our MAP inversion (**A**, equation 2) to determine our ability to independently estimate CH_4 emissions from our geographical regions. Figure A1 shows no significant *a posteriori* correlations between neighbouring geographical regions in our state vector. This is consistent with the *in situ* based inversion indicate a gradual emissions increase in China from ~2012. Sheng et al. (2021) find data being able to estimate independently regional

330 emission estimates in our state vector.

Our *a posteriori* emission estimates are broadly consistent with previous studies. For example, the increase in tropical emissions has been reported using GOSAT data or *in situ* data within a 3-D CTM inversion (McNorton et al., 2016; Fujita et al., 2020) , which examined shorter time periods of 2003–2015 and 1995–2013, respectively. The increase over Eastern Africa (that lies within our North Africa region) has been reported by several studies (Lunt et al., 2019, 2021; Pandey et al., 2021; Feng et al., 2022, 2023)

335 . Sheng et al. (2021) reported using GOSAT data that CH_4 emissions from China increasing increased by 0.36 Tg/yr , from 2012 to 2017 using GOSAT data. Comparing 2017. Over the same time period, we find estimate an increase of Chinese emissions of 0.63 0.64 Tg/yr inferred from the ground-based and 0.50 Tg/yr inferred from *in situ* data and increase of 0.50 Tg/yr inferred from the GOSAT data. We find that China has an isotopically lighter δ^{13}C source signature from 2012 (Figure 5), similar to the other regions we study, however the isotopic shift in the latitude band that corresponds to China is not as large as

340 in others (Figure ??), which suggests that heavier isotopic signature sources (such as coal mines) could be part of the emissions makeup here.

We see improved emissions estimates for and GOSAT data, respectively.

Figure A2 shows observed CH_4 timeseries at ground-based sites that we use to determine the corresponding GEOS-Chem *a posteriori versus a priori* simulations. Figure A2 shows mole fraction estimates and *a posteriori* mole fraction estimates

345 inferred from ground-based data at site locations. We find smaller residuals between simulated mole fractions using the fractions. *a posteriori A priori* emissions and the measurements values already show excellent agreement with observations (mean residual 9.01 ppb ; of 14.1 ppb and root-mean-square error (RMSE) 11.94 ppb) than between the of 18.3 ppb), but this is generally improved after the model is fitted to the *a priori in situ* values and the measurements (mean residual 13.06 ppb; RMSE 17.13 data, with smaller mean residuals (12.5 ppb) and RMSE (17.0 ppb). This compares favourably with is consistent previous

350 studies such as McNorton et al. (2018) , with that reported *a posteriori* RMSE of 12.30 ppb. Likewise, we see agreement of mole fraction estimates using GOSAT data (Figure A4; mean residual 41.72 ppb, RMSE 51.57 ppb). There are no significant values of 12.3 ppb. Figure A3 shows *a posteriori* correlations between neighbouring regions (Figure A1), determined by the CH_4 mole fractions at NOAA sites that we do not include the inversion. This provides an additional and independent test of our ability to describe atmospheric CH_4 using a subset of NOAA data that we use in our inversion (Table A2). Generally,

355 our *a posteriori* ~~error covariance matrix, A, meaning that~~ estimates agree with these independent data, but for some sites the model has difficulty reproducing the data, e.g. AMY (western S. Korea), KZD (Kazakhstan), and SDZ (mainland China). This is because some sites are influenced by local sources that are not representative of the spatial scale of our transport model ($\simeq$50,000 km^2). Similarly, we find agreement using *~~a posteriori regional emissions estimates are independent of one another. Independent validation of the~~ a posteriori* mole fractions ~~are provided by comparison with NOAA network sites not included~~

360 ~~in the inversion~~ using GOSAT data (Figure ~~A3~~A4; mean residual of 29.1 ppb and RMSE 35.1 ppb).

A posteriori source signatures of δ^{13}C

Figure 5 shows *a posteriori* regional δ^{13}C emissions source signatures inferred from ground-based *in situ* data. ~~The results are grouped~~ We group our results into approximately three-year bands, as a residual from the ~~2004-2007~~ 2004–2007 mean value, to show how the regional isotopic source signatures change across the time series.

365 Relative to *a priori* emissions (Figure A5), *a posteriori* values from Northern Boreal regions (Boreal North America and Eurasia) have isotopically lighter signatures (-62‰) ~~, consistent with the dominance of biogenic emissions~~ due to a larger contribution from isotopically lighter biogenic emissions and/or a smaller contribution from isotopically heavier thermogenic or pyrogenic emissions (Figure A5). Conversely, *a posteriori* values from regions such as Temperate Eurasia, Australia and Southern Africa have isotopically heavier source signatures (approximately -40‰) ~~, suggesting a greater~~ due to a larger pro-

370 portion of thermogenic or pyrogenic emissions ~~.~~ and/or a smaller contribution from isotopically light biogenic emissions.

Figure 5 shows a general trend towards isotopically lighter regional source signatures of δ^{13}C across the time series. ~~This~~ Our analysis suggests this trend has been ongoing since 2012 and is observed in all regions worldwide, ~~however is strongest as~~ but is strongest (compared with *a priori* estimates ~~in~~) over Tropical and Southern Hemispheric regions~~such as~~. For example, Tropical South America and Southern Africa ~~(1.8‰ and 2.1~~ are 1.2‰ and 0.9‰ isotopically lighter than *a priori* values for

375 for 2019 and 2020, respectively~~). Emissions from these tropical regions have a strong δ^{13}C seasonal cycle (Figure A5), with isotopically lighter values during summer months, driven by a greater proportion of biogenic emissions at this time. It therefore follows that less significant seasonality is indicative of thermogenic or pyrogenic emissions making up a significant part of the emissions mix (observed for example in China and Temperate Eurasia). There is also evidence to suggest a period around 2012~~

~~.~~

380 Our analysis also highlights a period 2007-2012 when regional source signatures, particularly northern hemisphere regions, become isotopically heavier ~~(by approximately 1.0‰~~ compared with *a priori* source signatures ~~), especially in the Northern Hemisphere, before becoming isotopically lighter~~again. ~~This suggests a~~(by 1.0‰ 2007–2009; by 0.8‰ 2010–2012; and 0.3‰ 2013–2015). After 2012, regional source signatures of δ^{13}C generally become isotopically lighter. This result is suggestive that 2012 was a period when there was a change in the ~~sources of~~ balance of global sources that determine changes in atmospheric

385 CH$_4$~~dominating CH$_4$ emissions during this brief period~~. These isotopic shifts in 2008 and 2012 are noted by Nisbet et al. (2016), who ~~use~~ used a box model and examine data from sites measured by NOAA and Royal Holloway, University of London (RHUL). They found that changes in removal rates ~~would~~ could not explain these anomalies ~~; the events are therefore~~ so that these events were attributed to changing emissions.

~~We find some significant~~ We find that China experiences a weaker shift in 2012 to a (0.1‰) isotopically lighter δ^{13}C source signature compared to *a ~~posteriori~~priori* ~~correlations between neighbouring regions~~for these values (Figure 5) and compared to other temperate regions. This suggests that heavier isotopic source signatures (~~Figure A6) , determined by~~ such as coal mines) play a larger contribution to this region.

Unlike the *a posteriori* ~~error covariance matrix,~~ total CH_4 emission estimates, we find significant ~~A~~a *posteriori* ~~, which indicates that we cannot differentiate between the isotopic source signatures of neighbouring regions (such as~~ correlations between neighbouring regions for δ^{13}C source signatures (Figure A6). For example, there is a correlation of 0.95 between estimates for Southern Africa and Temperate South America ~~). This~~so these cannot be considered as independent estimates. This result aligns with Basu et al. (2022) ~~, who used both~~ who used CH_4 mole fraction and δ^{13}C measurements to determine that tropical biogenic sources are driving CH_4 growth~~, however acknowledge~~. They acknowledged that measurement coverage ~~limits possible conclusions based upon~~ limited conclusions based exclusively on isotope ratio measurements. Nevertheless, ~~the~~ they found a clear trend of stronger emissions of isotopically lighter CH_4~~is clear, indicating~~, indicative of an increased role ~~in~~ for biogenic emissions in the global source makeup.

~~The corresponding~~ We find that *a posteriori* regional δ^{13}C source signatures ~~produce an atmospheric time series~~ result in a time series of δ^{13}C that is more consistent with ~~measurements~~observations than *a priori* values (Figure A7), ~~particularly during 2008-2018~~ as expected. This particularly affects the period 2008–2018 when *a priori* emissions source signatures are ~~significantly~~isotopically lighter. ~~Figure A7 shows δ^{13}C~~ Our *a ~~prior~~and a* posteriori ~~values at site locations. The a posteriori~~source signatures result in ~~smaller residuals between the a posteriori simulation and measurements (mean residual~~a mean observed-model residual and RMSE of 0.11‰ ~~, RMSE 0.15‰), than from the prior (mean residual 0.19‰, RMSE 0.23‰). This result compares well to McNorton et al. (2018) (~~and 0.14‰, respectively. These are smaller than those corresponding to *a ~~posteriori~~a priori* values for the observed-model residual (0.37‰) and RMSE (0.41‰). Our comparison is consistent with McNorton et al. (2018) (RMSE 0.1‰) and Fujita et al. (2020) (~~a posteriori~~RMSE 0.08-0.25‰).

~~In Figure ??, we combine this information into a zonal plot, reported approximately every 30latitude, for CH_4 emissions and the corresponding changes in regional isotopic source signatures of δ^{13}C. We find consistency between the magnitude of the changes in CH_4 inferred from ground-based and GOSAT data, particularly in the low latitudes. The plot also shows there has been a progressive increase in emissions from tropical latitudes (between 60-80 Tg~~

Sensitivity to assumptions about OH

Figure 6 shows the result from our sensitivity test that assumes 0.5%/yr ~~in 2019-2020) and a decrease at northern midlatitudes (up to -10 Tg/yr). This suggests that emissions have shifted from northern midlatitudes towards tropical emissions. We also find a move towards isotopically lighter regional source signatures of δ^{13}C across all latitudinal bands, with a change of approximately -2‰ in the tropics. Comparing Figures 5 and ??, we see similar trends across latitudinal bands and the regions within them, for example trends in European and Chinese source signatures align with the 30-60N latitudinal band. Our results compare well with Nisbet et al. (2019), who use a box model to fit emissionsscenarios to in situmeasurements, examining~~

2000-2018. They show strongest emissions increases from the tropics (approximately +20 Tg/yr, Figure 5). They likewise show consistently isotopically lighter atmospheric δ^{13}C across the time series, by approximately 0.03‰/yr.

Figure 4 compares our calculated atmospheric growth rate from the model simulations with the growth rates calculated from the in situ observations alone. We applied the same technique as NOAA follow to compare their published growth rate to the a posteriori mole fractions of our inversion. The general trend in increasing growth rate is evident in both measurement and model datasets with inter-annual discrepancies explained through model measurement mismatch at specific sites uniform decrease to our 3-D OH field from 2004 to 2019, followed by a more abrupt decrease of -1.5% in 2020 to describe the widespread reduction in nitrogen oxide emissions. This is an idealised sensitivity test that is inconsistent with global-scale constraints on estimates of the global mean atmospheric growth of atmospheric CH_4, i.e., most of the observed global growth in atmospheric CH_4 can be explained by the changes in OH. Nevertheless this test provides us with some idea of the robustness of our results against changes in OH.

Figure 6 shows that the 0.5% negative trend in We find that this alternative assumption about OH does not make a significant difference to our significantly affect our results until much later in the timeseries (2017–2019), reflecting our large *a posteriori* estimates (emissions change is not larger than uncertainties. We find a similar quality of fit of the *a posteriori* uncertainty) until later in the timeseries (2017-2019), reflecting our large model to the data with or without considering the OH trend (not shown). This does not preclude a role for changes in OH but the concurrent *a posteriori* uncertainties. However, we find that a sudden 5% decrease in OH during shifts in CH_4 emissions and regional isotopic source signatures of δ^{13}C are consistent with decreasing OH playing a smaller role than increasing emissions of isotopically light δ^{13}C source signatures in determining observed changes in atmospheric CH_4 (Lan et al., 2021).

The larger, abrupt change in 2020 results in a marked reduction (approximately 9%, 50 6%, 40 Tg/yr) in the emissions necessary to explain the increase in atmospheric CH_4. This reduction in emissions increases particularly affects high-emitting regions such as China and Tropical Asia. Despite this, the regional results are generally within the There is still debate about the impact of *a posteriori* uncertainties of our control calculation, which does not include a year-to-year change in OH. On balance, given the large-scale, unprecedented changes in atmospheric chemistry during 2020 it is likely that OH has played a role in the global atmospheric growth rate of CH_4, but changes in emissions likely overwhelm the impact from reduced OH methane emissions. Peng et al. (2022) used *in situ* data and concluded that the increase in atmospheric CH_4 in 2020 could be attributed approximately equally between a decrease in OH and an increase in OH. Analysis of GOSAT suggest that increased emissions play a larger role (Qu et al., 2022; Feng et al., 2023). We find a similar fit of the model to data with or without considering the OH trend (not shown).

4   Conclusions

We estimated regional CH_4 emissions and δ^{13}C source signatures for the period 2004-2020 2004–2020, inclusively, by fitting the GEOS-Chem 3-D atmospheric chemistry transport model to surface mole fraction data and GOSAT atmospheric column data using Bayesian inverse methods. Collectively, our results indicate that the post-2007 increases in CH_4 emissions are best

455   explained by a progressive latitudinal shift in emissions from the northern midlatitudes to tropical latitudes. *A posteriori* CH_4 emission estimates inferred from the ground-based and GOSAT data show larger tropical emissions, particularly over North Africa, Tropical Asia, and Tropical South America, and over China and at the same time as mid-latitudinal emission ~~proportion~~ proportions decreases. Source signature estimates inferred from the $\delta^{13}C$ measurements over the same time period indicate that the latitudinal shift in CH_4 emissions is due to larger proportion of ~~biogenic sources.~~ sources with a lighter atmospheric

460     $\delta^{13}C$ signature (e.g., biogenic source such as wetlands) and/or a smaller proportion of sources with a heavier atmospheric $\delta^{13}C$ signature (e.g. thermogenic or pyrogenic sources). Our results are broadly consistent with previous studies that focus on shorter, contributing periods (McNorton et al., 2018; Nisbet et al., 2019; Fujita et al., 2020; Yin et al., 2021; Lan et al., 2021; Basu et al., 2022)).

~~Our control calculations used monthly 3-D distributions of OH without any year-to-year variation. To explore how changes~~

465   ~~in OH might affect our results, we ran a sensitivity experiment for which the monthly 3-D OH fields was decreased~~ We find that our main results are robust against assuming a 0.5%/yr OH decrease from 2004 to 2019, ~~inclusively, based on values proposed by previous studies (Turner et al., 2017). For this sensitivity experiment, we find our results are within a posteriori uncertainty of the control calculations for most of the time series, and therefore steadily decreasing OH concentrations are not responsible for observed changes in the distribution of CH_4. We also considered how a proposed larger 5% change in~~ consistent with

470   Turner et al. (2017), followed by an abrupt 1.5% OH drop in 2020 ~~(Miyazaki et al., 2021; Laughner et al., 2021), due to widespread COVID-19 related emission reductions in nitrogen oxides, affected our results. We find smaller CH_4 emissions increases during 2020, as expected, but for most regions they are still within our control a posteriori emissions estimates for 2020. A much larger reduction in OH would be necessary to describe exclusively observed changes in atmospheric CH_4, which would consequently affect regional isotope signatures and observed variations of many atmospheric trace gases in a manner that has yet to be~~

475   ~~reported~~ that reflects the widespread decrease in nitrogen oxide emissions from shutting down manufacturing during the first Covid-19 lockdown. This is an idealised sensitivity test but nevertheless provides us with some idea of the robustness of our results against changes in OH.

    Sparse geographic coverage of ground-based data results in larger uncertainties for regional emission estimates that are ~~poorly covered~~ informed by fewer data, i.e. ~~,~~ high and low latitudes in both hemispheres. For CH_4, this deficiency can be partly

480   addressed using the satellite data, but isotope ratios cannot usefully be retrieved from Earth observation satellite instruments. ~~In this study,~~ We use only three measurement sites for $\delta^{13}C$ in the Southern Hemisphere, which have a continuous record over the period of study. A consequence of this data ~~sparseness~~ sparcity is strong correlations between source signatures from neighbouring regions (Figure A6). ~~We further limited our study by picking measurements sites for which data are available over our study period (Figure 1). Sectoral~~ We assume mean sectoral $\delta^{13}C$ source signatures ~~are taken as mean values~~ from

485   Sherwood et al. (2017). These values are highly uncertain, as different sectors produce a range of possible $\delta^{13}C$ values, and there are significant overlaps between recorded source signatures (Douglas et al., 2017), but the values chosen represent our current best knowledge of mean values. These data have greater value when they are used in a broader context with other data, as we have described in this study. We have used satellite observations to help identify that large-scale emission changes over regions that coincide with wetlands. ~~The collective evidence demonstrates that increasing tropical wetland emissions play~~

490   Collectively, empirical evidence, including *in situ* and GOSAT observation of CH_4 and *in situ* $\delta^{13}C$ data, points to an increasing biogenic source originating from the tropics. While we cannot definitely attribute these changes to increasing wetland emissions, there is sufficient contextual evidence, building on previous studies, to suggest that wetlands are playing a significant role in ~~the observed atmospheric growth~~ recent growth of atmospheric CH_4. First, large changes in OH that would needed to explain this atmospheric growth are inconsistent with increasingly isotopically light $\delta^{13}C$ observations in

495   the atmospheric record (Lan et al., 2021). Second, we know from *in situ* data the broad geographical regions responsible for increasing CH_4 emissions and isotopically lighter $\delta^{13}C$ source signature, where the seasonal cycles are consistent with biogenic emissions peaking outside the burning season. Third, GOSAT provide us with additional information about the geographical distribution of CH_4 ~~.~~emissions: tropical emission hotspots are colocated with known wetland regions (Lunt et al., 2019, 2021; Pandey et al., . Finally, we also have evidence from other satellite data, e.g., hydrology, that help explain the growth of wetland emissions

500   in the last decade (Lunt et al., 2019; Feng et al., 2022). Greater confidence in source attribution of changes in atmospheric CH_4 may come from collecting and interpreting δD and multiply-substituted 'clumped' isotopes (Douglas et al., 2017; Chung and Arnold, 2021), alongside $\delta^{13}C$. This needs to be accompanied by field measurements of these isotope ratios to improve delineation between different sectors.

~~Our work is also consistent with recent studies that have reported anomalous large CH_4 emissions over Eastern Africa (East~~
505   ~~Africa and the Horn of Africa) due to elevated rainfall over upstream catchment areas (Lunt et al., 2019, 2021; Pandey et al., 2021)~~ ~~. These large-scale precipitation changes have been linked with the positive phase of the Indian Ocean Dipole (Feng et al., 2022)~~ ~~, which describes a sea-surface temperature gradient over the Indian Ocean. Similarly, increased CH_4 emissions over the~~ ~~Amazon basin (Wilson et al., 2021) are linked with large-scale changes in climate (Feng et al., 2022). These substantial increases~~ The evidence presented here is consistent with a growing body of work that points to a substantial increase in biogenic CH_4

510   emissions from the tropics. This increase will likely have major implications for our achieving the goals of the Paris Agreement (Nisbet et al., 2019). Nature does not care about the origin of atmospheric CH_4 so that increasing biogenic emissions will require larger emission reductions from anthropogenic sectors, placing additional pressure on citizens to reduce their carbon footprints.

5   Code and data availability

515   The community-led GEOS-Chem model of atmospheric chemistry and model is maintained centrally by Harvard University (http://geos-chem.seas.harvard.edu), and is available on request. The ensemble Kalman filter code is publicly available as PyOSSE (https://www.nceo.ac.uk/data-tools/atmospheric-tools/).

6   Data availability

All the data and materials used in this study are freely available. The NOAA-GML and CU-INSTAAR ground-based CH_4 and
520    $\delta^{13}C$ data are available from the NOAA GML FTP server (https://gml.noaa.gov/dv/data), subject to their fair use policies. Data

from JR-STATION network was provided with cooperation of NIES Japan. The University of Leicester GOSAT Proxy v9.0 XCH_4 data are available from the Centre for Environmental Data Analysis data repository at (https://doi.org/10.5285/18ef8247f52a4cb6a14013f8235cc1eb), and from the Copernicus Climate Data Store. EDGAR data is available at (https://edgar.jrc.ec.europa.eu/), GFED-4 data is available at (https://www.globalfiredata.org/data.html), WETCHARTS
525   data is available at (https://daac.ornl.gov/cgi-bin/dsviewer.pl?ds_id=1502).

Appendix A: Isotopologue Emissions

To simulate the atmospheric isotope ratio δ^{13}C the isotopologues $^{12}CH_4$ and $^{13}CH_4$ are considered separately in the model. To calculate the specific sectoral isotopologue emissions we use the emissions calculated from the mole fraction CH_4 simulation and the isotope ratios defined in Table 1. We consider the isotope ^{13}C relative to all isotopes in the sample (designated thereafter
530   as $13x$) using:

$$13x = \frac{^{13}C}{^{12}C + {^{13}C}} = \frac{^{13}C/^{12}C}{1 + (^{13}C/^{12}C)}, \tag{A1}$$

where $^{13}C/^{12}C$ is calculated from the δ^{13}C reported on the international carbon isotope scale VPDB (Vienna Pee Dee Belemnite). This is the proportional molar abundance of the isotopologues containing ^{13}C (dominated by $^{13}CH_4$) relative to the isotopologues containing ^{12}C (dominated by $^{12}CH_4$) . This value has to be adjusted before being applied in GEOS-Chem to
535   convert from isotope ratio values to kg values used by emission inventories:

$$SF13 = 13x \times \frac{M_{13}}{M_{tot}}, \tag{A2}$$

where $SF13$ is the scale factor applied to each emissions type for the $^{13}CH_4$ simulation, M_{13} is the molecular weight of $^{13}CH_4$ (17.035 g/mol) and M_{tot} is the molecular weight of CH_4 (16.04 g/mol).

For the $^{12}CH_4$ counterpart to $^{13}CH_4$, we use a similar approach. The ratio of ^{12}C compared with all isotopes in the sample
540   (designated as $12x$) is given by:

$$12x = \frac{^{12}C}{^{13}C + {^{12}C}}. \tag{A3}$$

This is similarly adjusted from molar to mass ratio; $SF12$ is the scale factor for each emissions type in the $^{12}CH_4$ simulations:

$$SF12 = 12x \times \frac{M_{12}}{M_{tot}}, \tag{A4}$$

545   where M_{12} is the molecular weight of $^{12}CH_4$ (16.03 g/mol). Since ^{13}C and ^{12}C are the only stable carbon isotopes of CH_4, $13x$ and $12x$ should sum to 1.

Competing interests. The authors declare that they have no competing interests.

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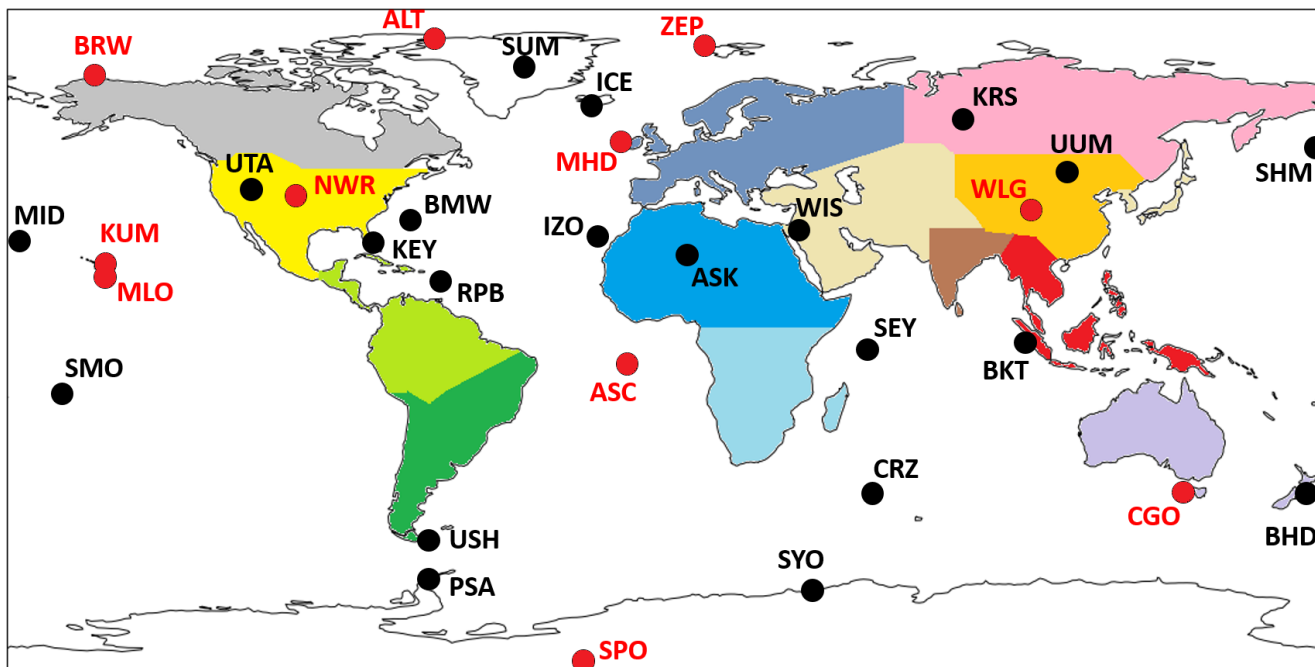
Figure 1. Map showing regions that are optimised in the CH$_4$ and δ^{13}C inversions, in different colours. Black dots and labels show the location of ground-based measurement sites that measure CH$_4$ mole fraction. Red dots and labels indicate both mole fraction CH$_4$ and δ^{13}C measuring sites. Regions are named as follows: Grey - North American Boreal; Yellow - North American Temperate; Light Green - South American Tropical; Dark Green - South American Temperate; Purple - Europe; Blue - North Africa; Light Blue - Southern Africa; Pink - Boreal Eurasia; Orange - China; Brown - India; Peach - Temperate Eurasia; Red - Tropical SE Asia; Lilac - Oceania; White - Oceans. Site identifiers are detailed in Table A2.
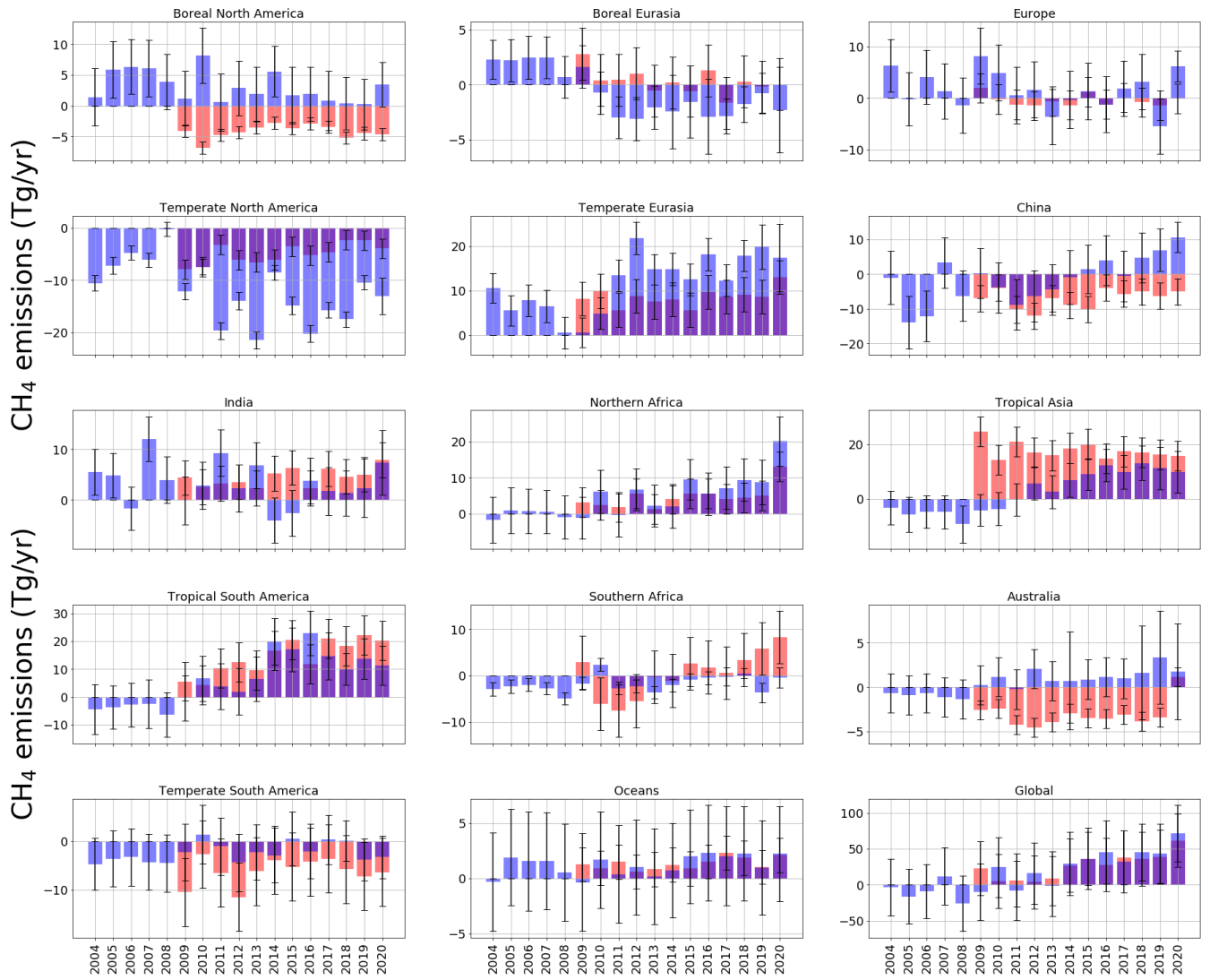
Figure 2. Annual mean CH$_4$ *a posteriori* emissions estimates as a residual value relative to *a priori* (Tg/yr) from each of the inversion regions in latitudinal order (geographic coverage indicated by Figure 1), for both ground-based and GOSAT inversion results. Uncertainties are indicated, as calculated from inversion calculations, with a *a priori* uncertainty of 50% for the ground-based results and 60% for the GOSAT results. The ground-based *a posteriori* is in blue; the GOSAT *a posteriori* are in red.
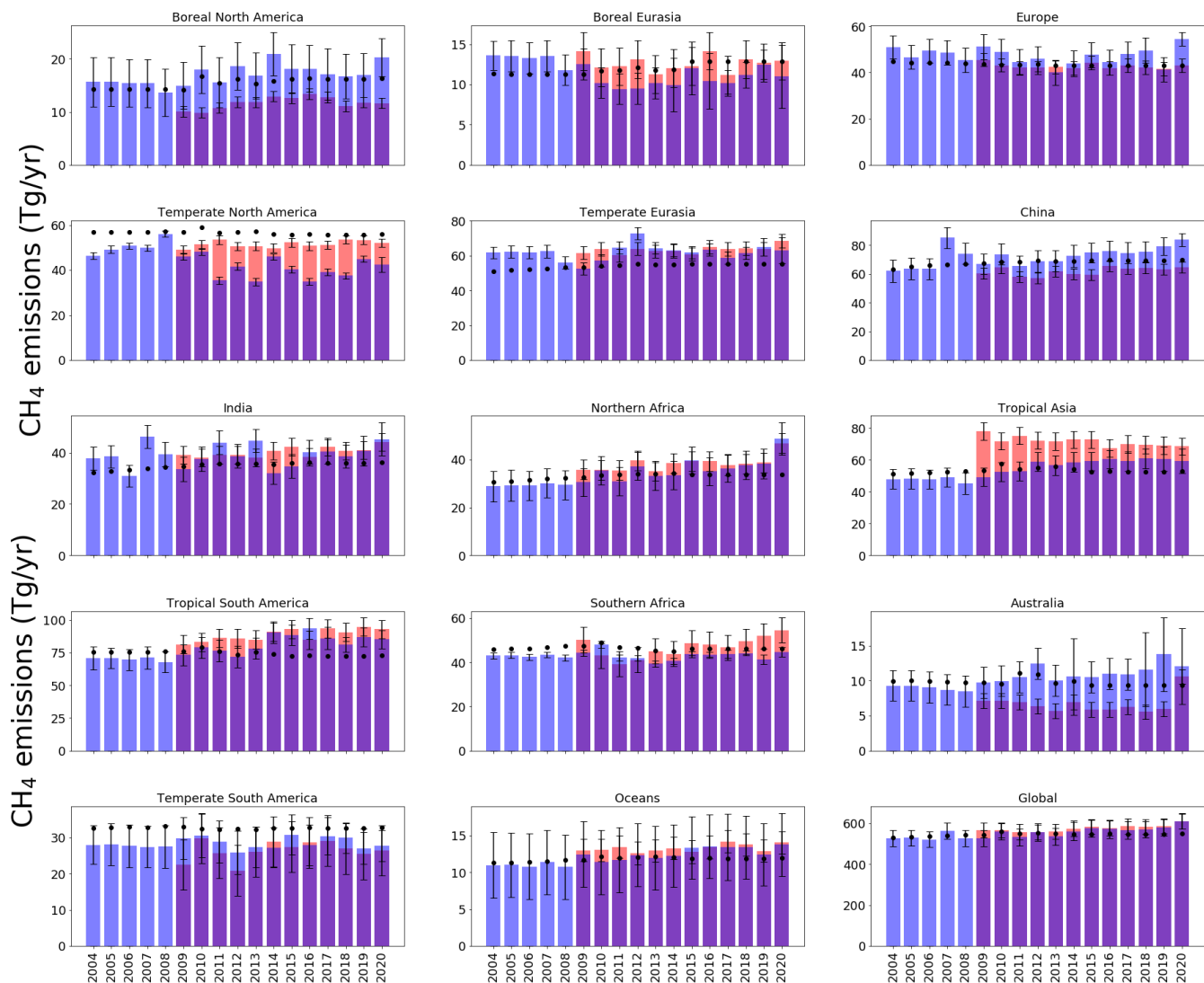
Figure 3. *A posteriori* emissions estimates (Tg/yr) inferred from ground-based *in situ* data (blue) and GOSAT data (red, with record starting in 2010) for the geographical regions shown by Figure 1. *A priori* emissions estimates are denoted by black dots and *a posteriori* uncertainties are denoted by whisker bars.
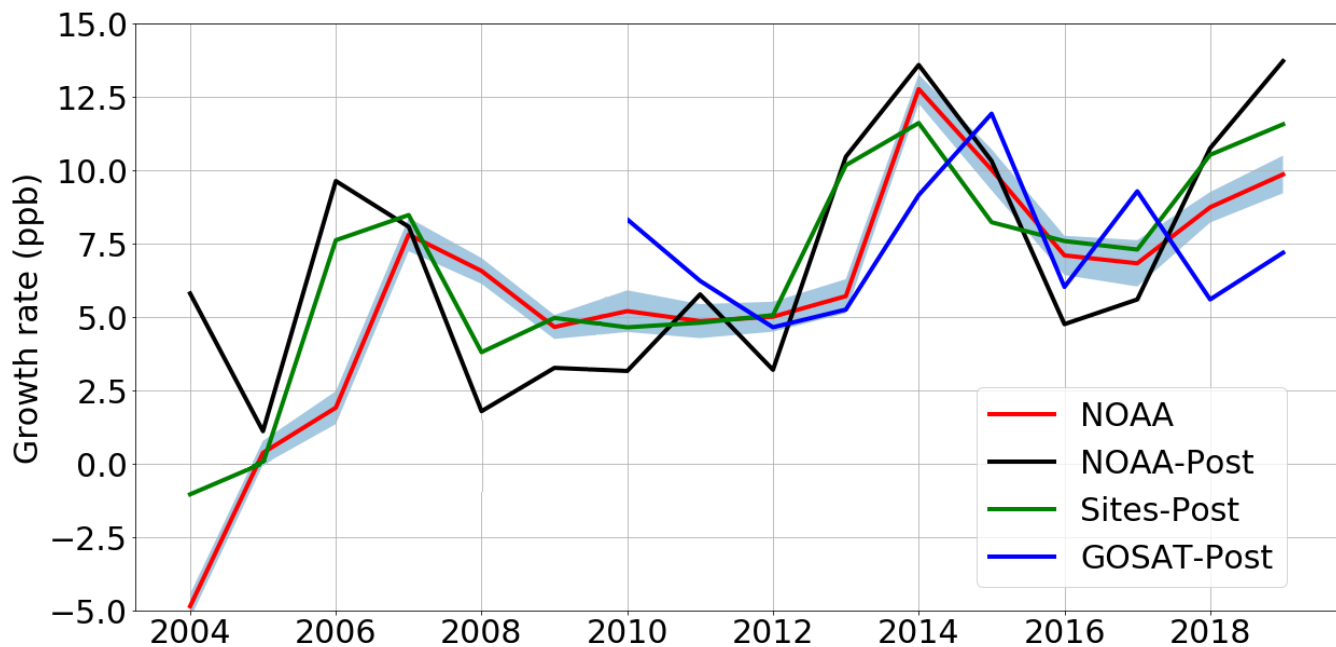
Figure 4. *A posteriori* annual mean atmospheric CH_4 growth rate inferred from *in situ* (black line) and GOSAT data (blue line) compared with the equivalent data as published by NOAA (red line, with uncertainty as blue surrounding field, Dlugokencky et al., 2020). The green line denotes the annual atmospheric growth rate determined using the *in situ* mole fraction data from the sites included in the inversion ('Sites-Post'). To calculate the atmospheric growth rates from model calculations (Ground-Post and GOSAT-post), we compare the average global CH_4 mole fraction in one year (the mean mole fraction of every grid box in every month of a year), with the mean value from the following year. The calculation is January-January, in order to remove the effects of the seasonal cycle, following the approach by NOAA (Dlugokencky et al., 2020).
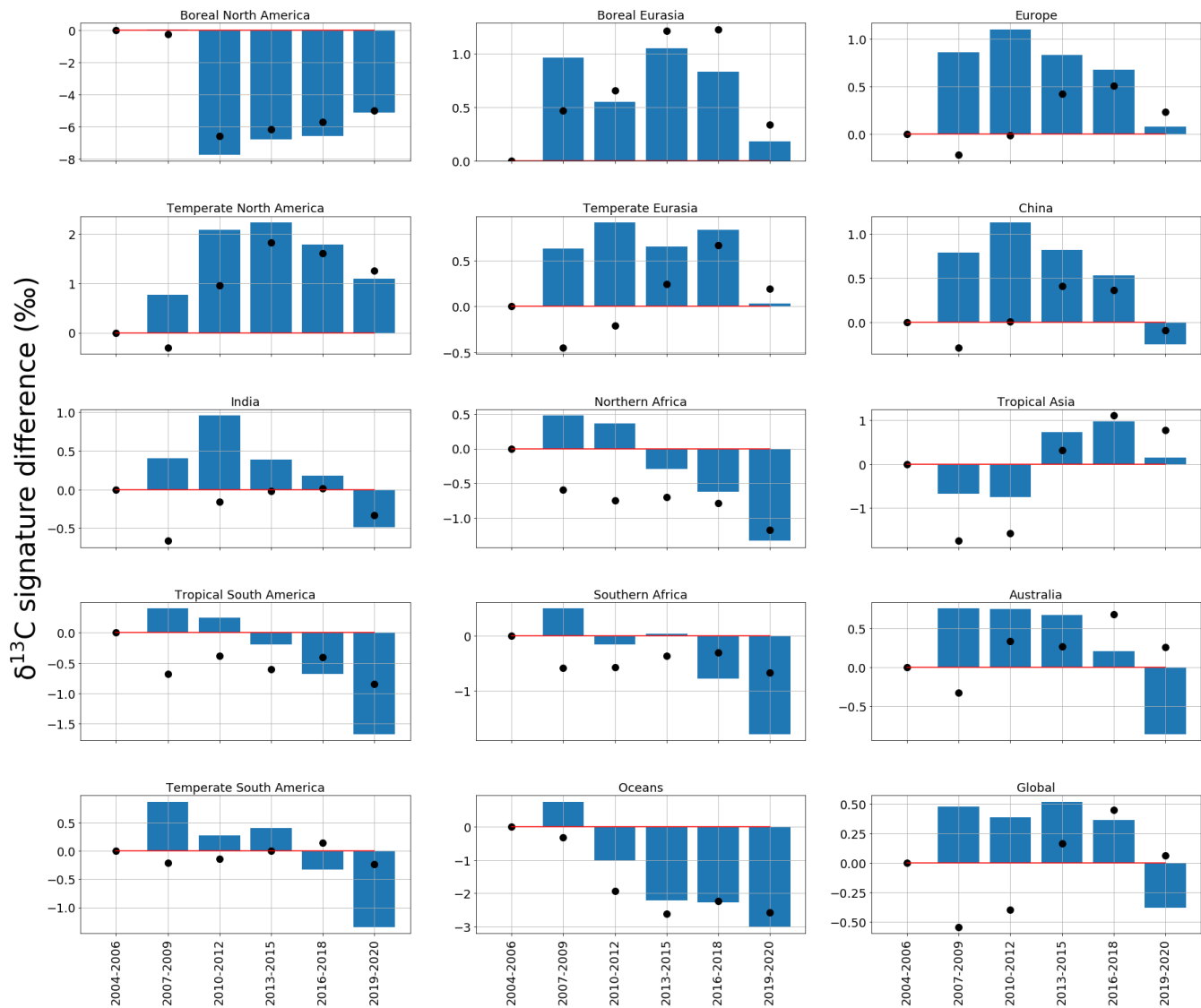
Figure 5. Regional and global *a posteriori* δ^{13}C emissions source signatures (‰), in three-yearly groups (2004-06, 2007-09, 2010-12, 2013-15, 2016-18, 2019-20) as a residual from the 2004-06 *a posteriori* regional emissions source signature value. The *a priori* equivalent is represented by black dots. The regions are those solved for in the CH_4 and δ^{13}C inversions and are indicated by Figure 1.
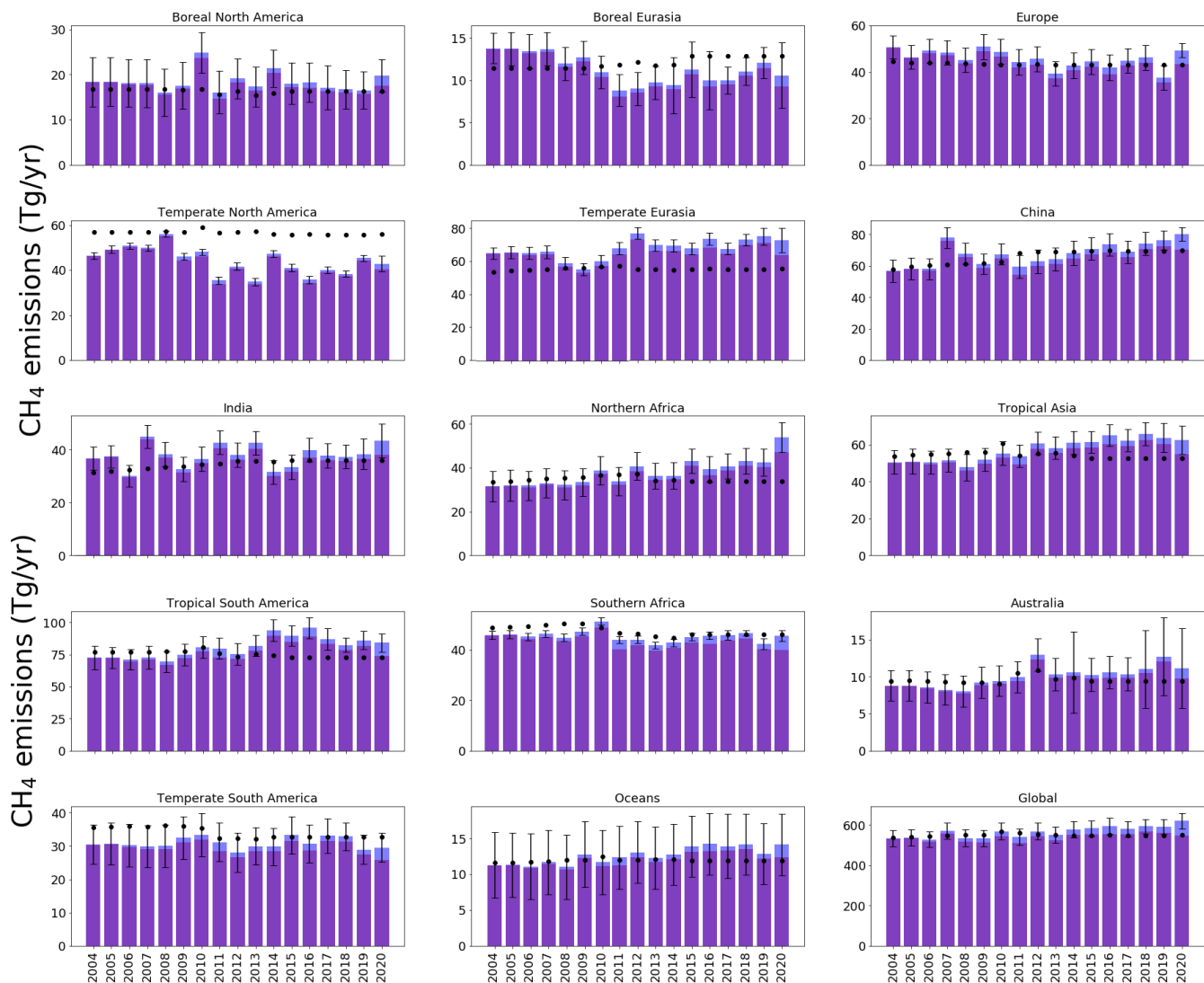
Figure 6. Annual mean CH$_4$ emissions (Tg/yr) for each region of the inversion (indicated by Figure 1) inferred from the ground-based data (dark blue) and the emissions estimates determined by a reduced OH values (described in the text, shown in red). *A priori* regional emissions estimates are indicated by black dots. Regional uncertainties for the *a posteriori* emissions are indicated.
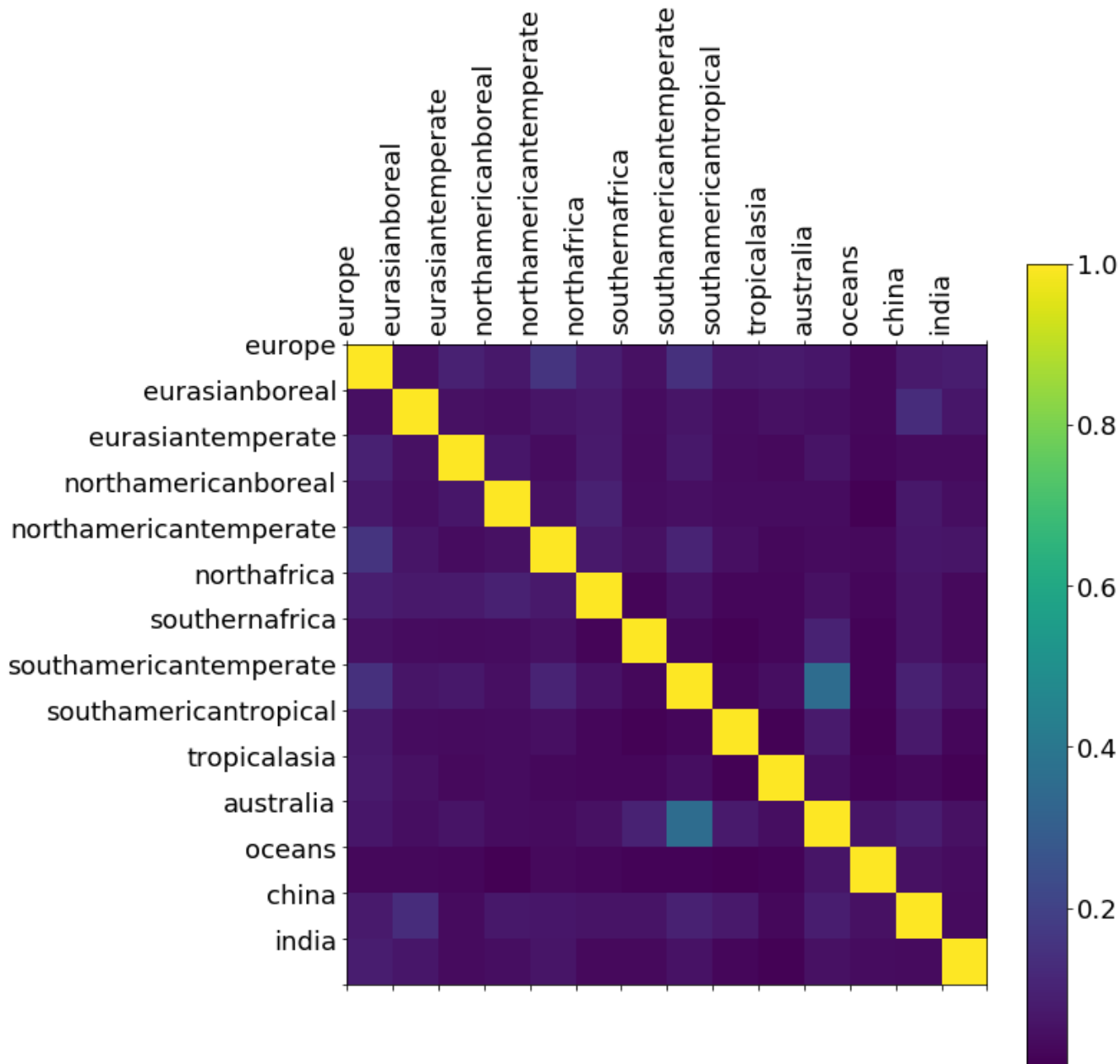
Figure A1. *A posteriori* correlations between CH₄ emissions from geographical regions inferred from ground-based CH₄ mole fraction data. These correlations are determined by normalising the diagonal elements of the *a posteriori* error covariance matrix (Eq. 2).
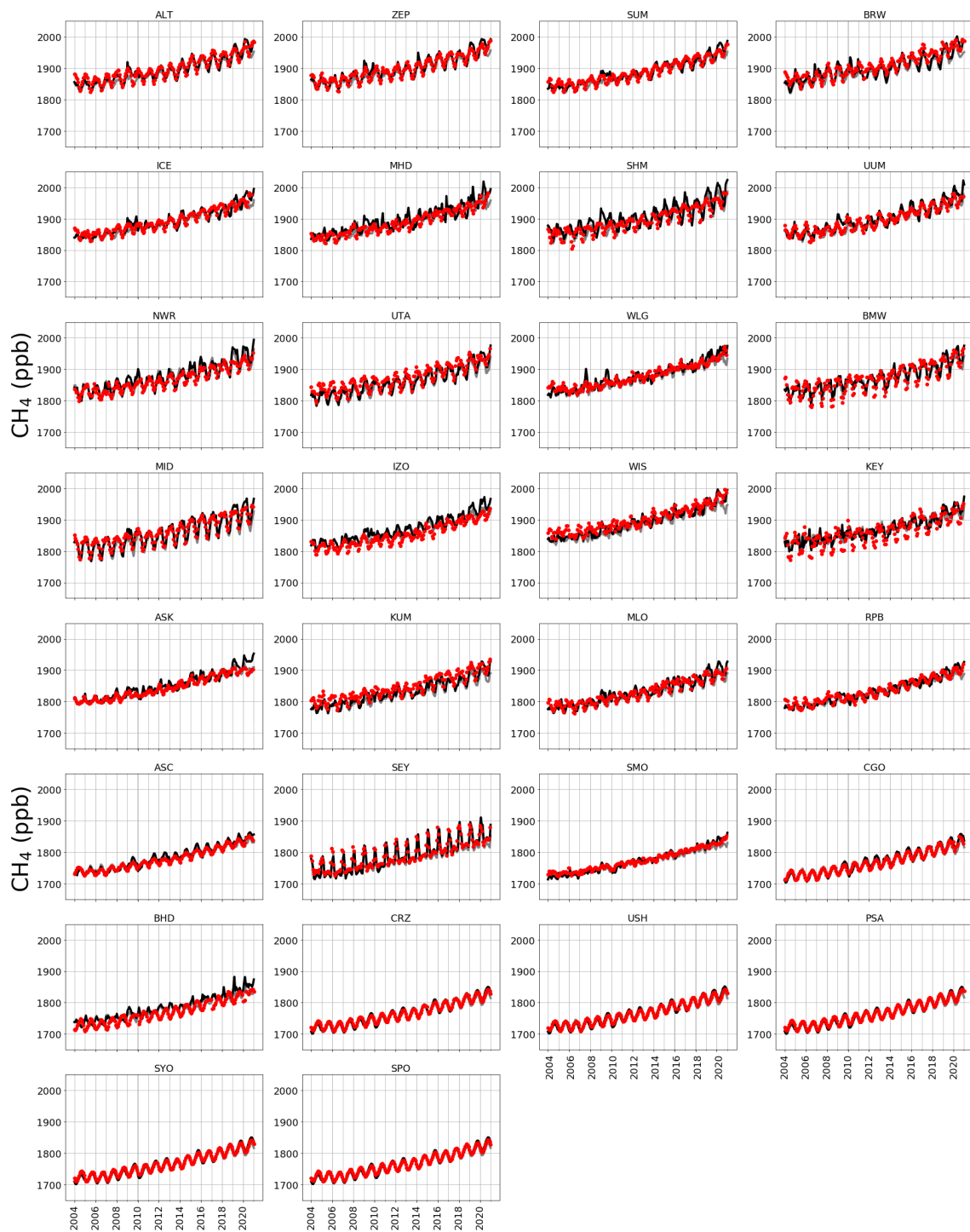
Figure A2. Observed (red dots), and *a priori* (grey), *a posteriori* (black) model atmospheric mole fractions at a series of NOAA sites (subplot titles denote site codes, Table A2), covering a range of latitudes.

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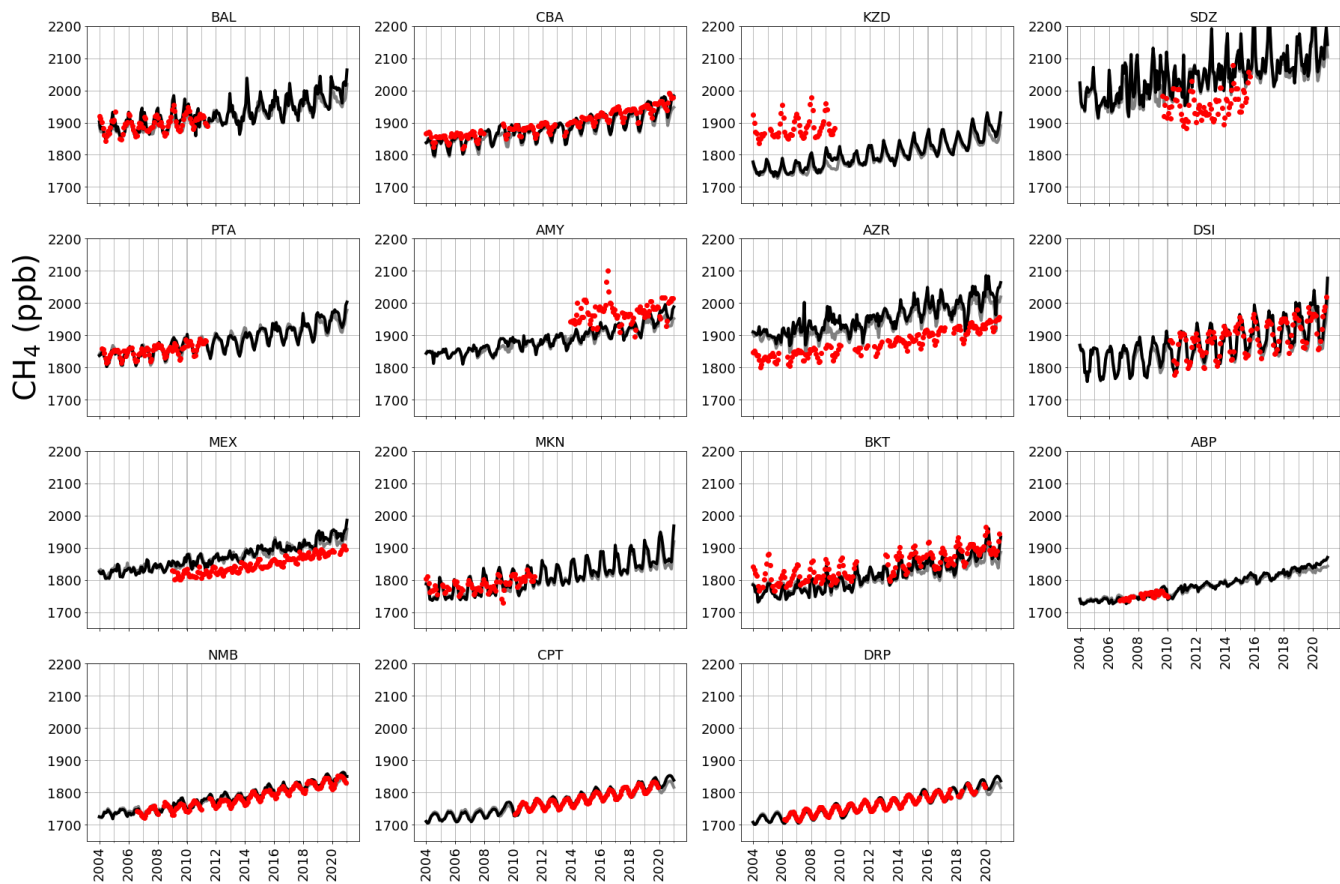
Figure A3. ~~Left: Triennial~~ *A posteriori* (black) monthly estimates of atmospheric CH$_4$, simulated at NOAA sites across latitudes. Red dots indicate monthly mean CH$_4$ ~~emission anomalies (Tg/yr)~~ data from ~~2004 to 2020 for ground-based~~ the NOAA network sites indicated. These sites were not included in the CH$_4$ inversion~~(denoted by shades~~, but are shown here to provide independent validation of ~~red~~ *a posteriori* emissions. The sites included are: Baltic Sea, Poland (55.35°N, 17.22°E)~~and from 2010 to 2020 for GOSAT inversion~~; Cold Bay, Alaska (~~denoted by shades of grey~~55.21°N, 162.72°W); Sary Taukum, ~~and triennial mean δ^{13}C emissions source signatures~~ Kazahkstan (~~‰,~~44.08°N, ~~denoted by shades of blue~~76.87°E)~~from 2004 to 2020~~; Shangdianzi, ~~all grouped every 30~~China (44.65°~~latitude. Right: The same results as left~~N, ~~defined as difference relative to the 2004–2006 mean values. The colours~~ 117.12°E); Point Arena, USA (38.95°N, 123.74°W); Anmyeon-do, Republic of ~~each x axis are indicative of the results to which they refer~~ Korea (~~i.e blue~~36.54°N, ~~upper axis for δ^{13}C results~~126.38°E); Terceira Island, ~~red~~Azores (38.77°N, ~~lower axis for CH$_4$ emissions results~~27.37°W); Dongsha Island, Taiwan (20.70°N, 116.73°E); High Altitude Global Climate Observation Center, Mexico (18.98°N, 97.31°W); Mt Kenya, Kenya (0.06°S, 37.29°E); Bukit Kototabang, Indonesia (0.20°S, 100.31°E); Arembepe, Brazil (12.77°S, 38.17°W); Gobabeb, Namibia (23.58°S, 15.03°E); Cape Point, South Africa (34.35°S, 18.49°E); and Drake Passage (59.00°S, 64.69°W).

Figure A4. Observed (red dots), and three-hourly surface *a posteriori* CH₄ values inferred from GOSAT data (black) at the location of a number of NOAA sites (Table A2) 2010-2020.
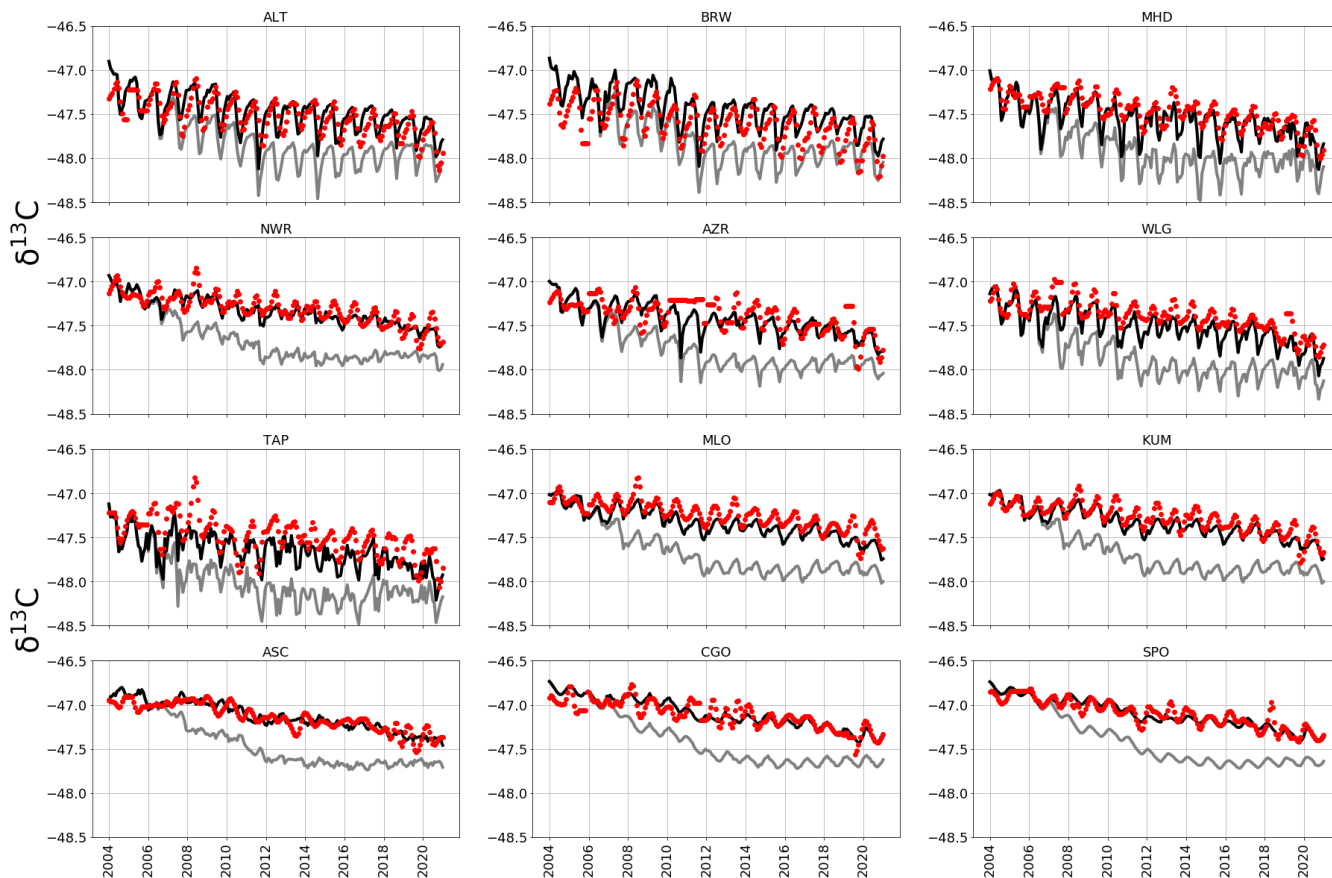
Figure A5. Monthly *a priori* (grey) and *a posteriori* (blue) regional δ^{13}C source signatures (‰). Values are produced using ground-based *in situ* δ^{13}C data. Uncertainties in source signatures are indicated as shaded envelopes, with *a priori* uncertainties of 15 ‰.
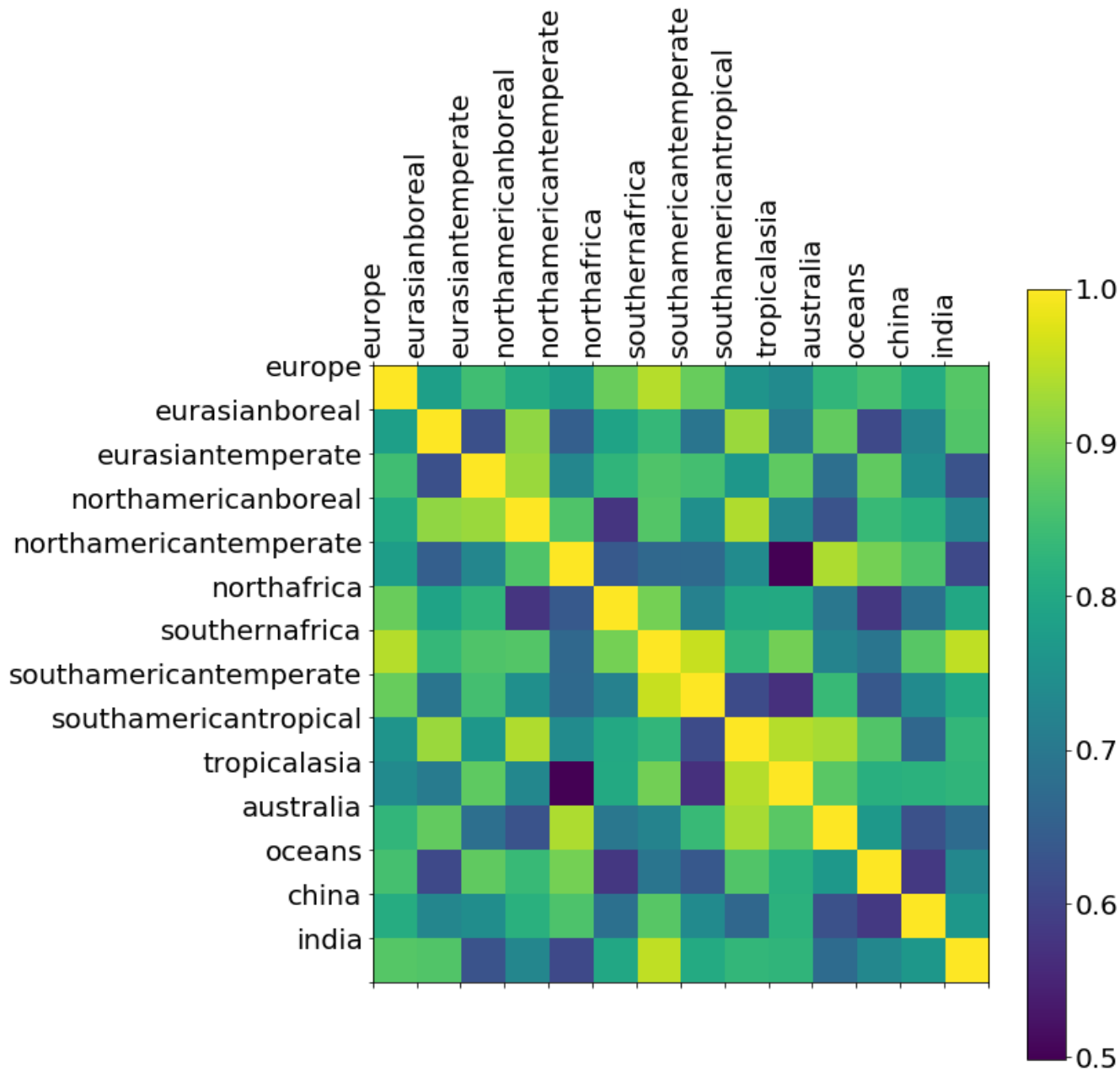
Figure A6. *A posteriori* correlations between δ^{13}C source signatures from geographical regions inferred from ground-based δ^{13}C data. These correlations are determined by normalising the diagonal elements of the *a posteriori* error covariance matrix (~~Equation~~ Eq. 2).

Figure A7. *A priori* (grey) and *a posteriori* (black) monthly estimates of atmospheric $\delta^{13}C$, simulated at NOAA sites across latitudes (site codes listed in Table A2). Red dots indicate monthly mean $\delta^{13}C$ data from CU-INSTAAR for the respective sites.

~~A posteriori(black) monthly estimates of atmospheric CH_4, simulated at NOAA sites across latitudes. Red dots indicate~~ ~~monthly mean CH_4 data from the NOAA network sites indicated. These sites were not included in the CH_4 inversion, but are~~ ~~shown here to provide independent validation of a posterioriemissions. The sites included are: Baltic Sea, Poland (55.35N,~~ ~~17.22E); Sary Taukum, Kazahkstan (44.08N, 76.87E); Shangdianzi, China (44.65N, 117.12E); Point Arena, USA (38.95N,~~ ~~123.74W); Dongsha Island, Taiwan (20.70N, 116.73E); Mt Kenya, Kenya (0.06S, 37.29E); Arembepe, Brazil (12.77S, 38.17W);~~ ~~Cape Point, South Africa (34.35S, 18.49E); Drake Passage (59.00S, 64.69W).~~

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Table 1. Global mean emissions of different CH$_4$ source types from bottom-up inventories (Saunois et al., 2020) and our *a posteriori* emission estimates, and the corresponding conventional isotope ratios signatures (Sherwood et al., 2017). Uncertainties are shown as max-min values in square brackets.

Source Type	Annual Mean Emission (Saunois) 2003-2012 (Tg/CH$_4$)	Annual Mean Emission (This Study) 2004-2020 (Tg/CH$_4$)	Isotopic Ratio δ^{13}C (‰)
Gas and Oil	80 [68-92]	82.0	-44.0 [$\pm$ 10.7]
Coal	42 [29-61]	53.7	-49.5 [$\pm$ 11.2]
Livestock	111 [106-116]	115.2	-65.4 [$\pm$ 6.7]
Waste	65 [60-69]	67.9	-56.0 [$\pm$ 7.6]
Biomass Burning	17 [14-26]	14.3	-26.2 [$\pm$ 4.8]
Termites	9 [3-15]	11.9	-63.4 [$\pm$ 6.4]
Wetlands	149 [102-182]	170.9	-61.5 [$\pm$ 5.4] (Tropical) -71.5 [$\pm$ 5.4] (Arctic)
Rice	30 [25-38]	30.7	-62.2 [$\pm$ 3.9]

Table A1. Kinetic Isotope Effects (KIEs) for different isotopologues reacting with the three main sinks of CH_4 (OH, Cl, soil) at 298 K. A KIE indicates relative reaction rate compared with $^{12}CH_4$; the reaction rate constant is applied to the OH and Cl sinks and is dependent ~~upon~~ on temperature (T); and the scaling factor is applied to the soil sink at each timestep (handled as a negative emission).

Isotopologue	Sink	KIE	Reaction Rate Constant	Scaling Factor	Literature Source
$^{12}CH_4$	OH	1	$2.45 \times 10^{-12} \times e^{\frac{-1775}{T}}$	n/a	Burkholder et al., 2019
$^{12}CH_4$	Cl	1	$9.600 \times 10^{-12} \times e^{\frac{-1360}{T}}$	n/a	Kirschke et al., 2013
$^{12}CH_4$	soil	n/a	n/a	1	Snover and Quay, 2000
$^{13}CH_4$	OH	1.0039	$2.44 \times 10^{-12} \times e^{\frac{-1775}{T}}$	n/a	Burkholder et al., 2019
$^{13}CH_4$	Cl	1.06	$9.057 \times 10^{-12} \times e^{\frac{-1360}{T}}$	n/a	Feilberg et al., 2005
$^{13}CH_4$	soil	n/a	n/a	1.0670	Snover and Quay, 2000

Table A2. Sites that are included in the *in situ* inversions. All sites are part of the NOAA network, other than KRS, which is part of the JR-STATION network, monitored by NIES Japan.

Code	Full Name	Latitude	Longitude
ALT	Alert Station	82.28	-62.30
ZEP	Ny-Alesund, Svalbard	78.90	11.89
SUM	Summit, Greenland	72.60	-38.42
BRW	Barrow Station	71.32	156.61
ICE	Storhofdi,Iceland	63.40	-20.29
KRS	Karasevoe, Siberia	58.14	82.25
MHD	Mace Head, Ireland	53.33	-9.90
SHM	Shemya Island, Alaska	52.71	174.12
UUM	Ulaan Uul, Mongolia	44.45	111.09
NWR	Niwot Ridge, Colorado	40.05	-105.59
UTA	Wendover, Utah	39.90	-113.72
WLG	Mt. Waliguan, China	36.29	100.90
BMW	Bermuda	32.26	-64.88
WIS	Ketura, Israel	29.96	35.06
IZO	Izana, Tenerife	28.31	-16.50
MID	Midway Islands	28.22	-177.37
KEY	Key Biscane, Florida	25.67	-80.16
ASK	Assekrem, Algeria	23.26	5.63
KUM	Cape Kumukahi, Hawaii	19.56	-154.89
MLO	Mauna Loa, Hawaii	19.54	-155.58
RPB	Ragged Point, Barbados	13.17	-59.43
SEY	Mahe Island, Seychelles	-4.68	55.53
ASC	Ascension Island	-7.97	-14.40
SMO	American Samoa	-14.25	-170.56
CGO	Cape Grim	-40.68	144.69
BHD	Baring Head	-41.40	174.87
CRZ	Crozet Island	-46.43	51.85
USH	Ushuaia, Argentina	-54.84	-68.31
PSA	Palmer Station, Antarctica	-64.77	-64.05
SYO	Syowa Station, Antarctica	-69.01	39.59
SPO	South Pole, Antarctica	-89.98	-24.8