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Regional trends and drivers of the global methane budget

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Abstract

The ongoing development of the Global Carbon Project (GCP) global methane (CH₄) budget shows a continuation of increasing CH₄ emissions and CH₄ accumulation in the atmosphere over

2000-2017. Here we decompose the global budget into 19 regions (18 land and one oceanic) and five key source sectors to spatially attribute the observed global trends. A comparison of top-down (atmospheric and transport model-based) and bottom-up (inventory- and process model-based) CH₄ emissions estimates demonstrates robust temporal trends with CH₄ emissions increasing in 16 of the 19 regions. Five regions - China, Southeast Asia, USA, South Asia, and Brazil - account for > 40% of the global total emissions (their anthropogenic and natural sources together totalling > 270 Tg CH₄ yr⁻¹ in 2008-2017). Two of these regions, China and South Asia, emit predominantly anthropogenic emissions (> 75%) and together emit more than 25% of global anthropogenic emissions. China and the Middle East show the largest increases in total emission rates over the 2000 to 2017 period with regional emissions increasing by > 20%. In contrast, Europe and Korea & Japan, show a steady decline in CH₄ emission rates, with total emissions decreasing by ~10% between 2000 and 2017. Coal mining, waste (predominantly solid waste disposal) and livestock (especially enteric fermentation) are dominant drivers of observed emissions increases while declines appear driven by a combination of waste and fossil emission reductions. As such, together these sectors present the greatest risks of further increasing the atmospheric CH₄ burden and the greatest opportunities for greenhouse gas abatement.

Keywords

Methane emissions, source sectors, anthropogenic emissions, natural emissions, top-down, bottom-up, regional

1 Introduction

Methane (CH₄) has a large contribution to the global radiative budget and is responsible for about 0.5°C of present global warming over the period 1850-1900 (IPCC, 2021). Methane has a relatively short perturbation lifetime (12.4 years (Balcombe, Speirs, Brandon, & Hawkes, 2018)) and high global warming potential (28-36 times that of CO₂ over a 100-year period (IPCC, 2021)). As such, a decline in CH₄ emissions will rapidly reduce global CH₄ concentrations and mitigate the impact of climate change at decadal time scales (United Nations Environment Programme & Climate and Clean Air Coalition, 2021). However, any efforts to target CH₄ emissions reductions require a thorough understanding of the dominant CH₄ sources and sinks and their temporal and regional distribution and trends.

Methane is produced in three ways – pyrogenically, thermogenically or biogenically – from both anthropogenic and natural processes. Pyrogenic sources of CH₄ include biofuel combustion (e.g. wood burning for heating and cooking) and biomass burning (e.g. wildfires and peat fires). All pyrogenic sources produce CH₄ from the incomplete combustion of organic matter. Thermogenic CH₄ is produced from the breakdown of organic matter buried deep within the Earth's crust at high pressure and temperature. While geological CH₄ is released naturally into the atmosphere through gas seeps, most is released through activities related to the exploration, mining and transport of fossil fuels (Hmiel et al., 2020; Janssens-Maenhout et al., 2019; Petrenko et al., 2017). The majority of biogenic CH₄ is produced in anaerobic environments by the microbial mediated breakdown of organic matter. These environments include wetlands, inland waters, marine sediments, ruminants such as cattle, rice paddies, manure management and wastewater and landfill systems. Small quantities of CH₄ are also produced from the aerobic bacterial metabolisation of methylated compounds (e.g. Florez-Leiva, Damm, & Farías, 2013) and even photochemically (Li, Fichot, Geng, Scarratt, & Xie, 2020).

Counter-balancing these CH₄ sources are three chemically-driven atmospheric sinks of CH₄. The first two reactions with tropospheric OH radicals and tropospheric atomic chlorine account for ~88% (476 - 677 Tg CH₄ yr⁻¹) and ~2% (1-35 Tg CH₄ yr⁻¹) of the total sink, respectively, with a third stratospheric sink (e.g. reaction with O(¹D), Cl and OH in the stratosphere) accounting for a further ~5% (12-37 Tg CH₄ yr⁻¹) (Saunois et al., 2020). However, due to their highly reactive nature the key reactants are inherently difficult to quantify, driving a significant level of uncertainty in the spatial and temporal distribution of atmospheric sink estimates (Zhao et al., 2019). Many fundamental aspects of the spatial distribution of OH are currently unresolved, for example, estimates of the interhemispheric gradient can vary from 0.85 to 1.4 (NH/SH) depending on the methodology (See Patra et al., 2014 and references therein). The large-scale geographical breadth of many of the chemical and physical drivers in OH spatial variability associated with intra and inter hemispheric transport of CH₄ over one year (Anderson et al., 2021) make the OH sink incompatible with a geo-politically defined surface-region-based study (the focus of this paper). As a result, only sources of CH₄ are discussed at regional scale in this study.

A fourth sink is the biological consumption of CH₄ by methanotrophic microbes in soils and other environments. It is estimated to be responsible for 5% of the global total CH₄ sink (Saunois et al., 2020). This process occurs in aerobic soils and inland waters where methanotrophic bacteria consume CH₄ and produce CO₂ (Le Mer & Roger, 2001). The oxidation of CH₄ can occur at the same site as the production of CH₄ either concurrently (e.g. aerobic soils capping landfills) or temporally shifted (e.g. sediments shifting from anoxic to oxic with seasonal waterflow patterns).

As such, the oxidative process can play a role in controlling CH₄ emissions to the atmosphere. In this paper, rather than separating production and consumption we focus on the policy relevant net surface emissions and exclude discussion of some spatially diffuse non-anthropogenic fluxes (e.g. upland soil sinks).

This paper analyses the recently published global CH₄ budget Saunois et al. (2020) and conducts an in-depth examination of CH₄ (total and sectoral) emission trends in 19 regions. Here we examine regions defined using geo-political boundaries and focus on net surface emissions (rather than separating production and consumption) because they are more directly relevant to policy analyses and the development of effective regional mitigation strategies. We use, as in Saunois et al, 2020, multiple emissions estimates from Top-Down (TD) approaches, based on atmospheric constraints, and Bottom-Up (BU) methods, which utilise extensive data inventories, terrestrial biospheric modelling and the statistical upscaling of empirical data.

In presenting data from a range of BU and TD methods this paper reflects the current state of science in relation to global-scale estimates of regional CH₄ emissions. By also clearly and concisely articulating their current limitations we build the groundwork for future research which will incorporate improvements in TD and BU methodologies. Using multiple methodologies allows us to not only estimate the uncertainty of these regional emissions (using the spread in emissions estimates) but also the uncertainty in the regional trends (using the spread in emissions trends). While not totally independent of BU methods (see Section 2.2), TD methods rely on CH₄ atmospheric mole fraction measurements which are an independent observational constraint. As such, commonalities between multiple BU and TD estimates can identify robust trends and patterns in regional CH₄ emissions and differences suggest areas requiring further investigation.

A regional decomposition (rather than latitudinal as in Saunois et al. (2020)) is essential as the drivers of CH₄ emissions vary widely in their spatial distribution and relative importance with climate, ecosystem type, anthropogenic activities and environmental policies. We also examine both decadal and annual data for the 2000-2017 period allowing the identification of trends and policy impacts.

Here we aggregate CH₄ emission types into five (three anthropogenic and two natural) key source sectors: agriculture and waste (*Ag&Waste*), fossil fuel (*Fossil*), biomass burning and biofuels (*BB&F*), wetlands (*Wetl*) and other-natural non-wetland (*NonWetl*).

We then examine four key questions pertinent to the design and implementation of emissions reductions strategies:

1. Which regions are the largest contributors to global CH₄ emissions?

2. What activities or sources are driving these emissions? – natural vs. anthropogenic sources and specific source sector types.
3. Which regions are the dominant contributors to the increasing trend in global CH₄ emissions and which sectors are driving this rise?
4. Are any regions seeing a decline in CH₄ emissions and which sectors are responsible for this decline?

We also examine key differences in TD and BU estimates, using them to identify methodological limitations and to highlight uncertainties and areas requiring further investigation.

2 Methods and materials

The spatial distribution of sources and sinks of CH₄ can be quantified using TD or BU approaches. Bottom-up approaches (BU) include process-based models, inventories, satellite-based products and other datasets which collate and scale-up local (or regional) direct flux measurements (e.g. flux towers and chamber-based approaches). Most commonly, BU-based estimates of anthropogenic CH₄ emissions are determined using inventory methods (e.g. EDGAR v4.3.2, Janssens-Maenhout et al., 2019). Here, known regionally distributed activity information and statistics are combined with technology-specific emission factors and abatement factors which account for recapture and emissions mitigation technology. Satellite-derived biomass burning products combine estimates of the area burned, biomass loading, combustion completeness and biome-specific emission factors to estimate CH₄ emissions (e.g. van der Werf et al. (2017)). Process based models, for example wetland models (e.g. Poulter et al. (2017)), can also be used to estimate sector-specific regionally distributed CH₄ emissions. Lastly, a variety of methods have been used to statistically scale up small scale or sparse empirical measurements of CH₄ flux to regional and global totals (e.g. Etiope, Ciotoli, Schwietzke, & Schoell, 2019).

Regional studies, which typically use region specific datasets and methodologies, are not included in this analysis as they may bias cross-region comparisons. Instead, where appropriate, regional studies are used to inform the discussion of trends identified in the global-scale regionally distributed datasets.

Top-down approaches take observed atmospheric mole fractions of CH₄ and combine them with global (or regional) transport models and an inversion framework to calculate an optimum set of spatially and temporally distributed source and sink fluxes. Studies using this approach are plentiful and use a variety of transport models, inversion methods, atmospheric datasets (in situ, flask, total column ground-based and satellite) and prior flux estimates. Examples include

Bergamaschi et al. (2018), Maasackers et al. (2019) and Yin et al. (2021). Some methods aim to separate certain emission sectors based on differences in their spatial and temporal distributions (e.g. Bergamaschi et al. (2013)), while others only solve for net emissions at the surface (Chandra et al., 2021). Then the partitioning of TD posterior (output) fluxes between specific source sectors (e.g. *Fossil* vs. *BB&F*) is carried out with various degrees of uncertainty depending on the methods and the degree of refinement of sectors.

Here we use a subset of the ensemble of TD and BU datasets gathered in Sauniois et al. (2020) (See Section 2.1 and 3.2.1) and a newly developed inland water emissions estimate (See Section 2.1.4 and S2). This approach means that the sum of the regional (and sectoral) budgets presented here, while very similar, are not equivalent to the global budget presented in Sauniois et al. (2020) (See Section 3.2.1).

2.1 Bottom-up data

2.1.1 Anthropogenic

A summary of the key global anthropogenic BU assessments used in this paper is given in Table 1, with further details found in Sauniois et al. (2020) and the references therein. Here we focus on the four global gridded anthropogenic datasets that encompass the sectors of interest, namely EDGAR v4.3.2. (Janssens-Maenhout et al., 2019), GAINS ECLIPSE v6 (Höglund-Isaksson, Gómez-Sanabria, Klimont, Rafaj, & Schöpp, 2020), USEPA 2012 (USEPA, 2012) and CEDS (Hoesly et al., 2018). For ease of reading these will henceforth be referred to as EDGAR, GAINS, USEPA and CEDS. It is important to note that while included in the analysis, the CEDS inventory (Hoesly et al., 2018) is based partly on an earlier dataset (EDGARv4.2) which is known to overestimate Chinese coal emissions (Peng et al., 2016). As FAO does not provide *Waste*, *Fossil* and *Biofuels* emissions estimates we, unlike Sauniois et al. (2020), do not include these in our analysis.

2.1.2 Biomass burning

Six biomass burning emissions estimates - FAO (FAO, 2019b), FINNv1.5 (Wiedinmyer et al., 2011), GFASv1.3 (Kaiser et al., 2012), GFEDv4.1s (van der Werf et al., 2017), QFEDv2.5r1 (Darmenov, 2015) and the USEPA emissions inventory (USEPA, 2012) - were discussed in Sauniois et al. (2020). The FAO data (FAO, 2021) were updated in Dec 2020 after the publication of Sauniois (2020) and both the original and updated versions are included in Fig. S1. Key

differences among biomass burning estimates include the lack of inter-annual variability (IAV) in the USEPA estimate and the unusually high (by a factor of 2-10, e.g. Fig. S1 USA and Russia) regional emissions of the original FAO estimates. Here, unlike Saunois et al. (2020), we use the mean of the updated FAO (FAO, 2019b), FINNv1.5 (Wiedinmyer et al., 2011), GFASv1.3 (Kaiser et al., 2012), GFEDv4.1s (van der Werf et al., 2017) and QFEDv2.5r1 (Darmenov, 2015) estimates of biomass burning.

2.1.3 Wetlands

Regional BU *Wetl* estimates were examined using 13 models from the wetland model intercomparison conducted as part of the 2020 GCP CH₄ budget project (GCP, 2021). These model runs use a common wetland extent map (diagnostic) (Z. Zhang et al., 2021) while nine prognostic runs (which were not used for regional estimates but do inform the discussion) used a range of internally generated wetland extent maps (see supplement Section 4 of Saunois et al. (2020)). Mean regional BU *Wetl* estimates were calculated using the data from the diagnostic wetland model runs.

2.1.4 Non-wetland sources including oceanic

Four BU estimates of total other-natural non-wetland (*NonWetl*) sources were calculated in this study. They ranged between 177-186 Tg CH₄ yr⁻¹ and were determined as the sum of the three main sources: aquatic biogenic (including inland waters, oceanic, estuaries and blue carbon, i.e. mangroves, seagrasses and marsh systems), geological (land and ocean) and termite.

Here we use a new estimate of global inland water and estuarine fluxes (Section S2). It is the first global-scale regionally distributed data set of this nature and was calculated as the sum of lake and reservoir, river and stream and estuarine fluxes. It was developed specifically for this study and not available at the time of publication for Saunois et al. (2020).

The biogenic oceanic estimate calculated by Saunois et al. (2020), 6 Tg CH₄ yr⁻¹ (range 4-10 Tg CH₄ yr⁻¹), included oceanic, estuarine and blue carbon fluxes. But estuarine and blue carbon fluxes occur at the interface between land and marine waters (i.e. straddling both land and ocean regions). In this study we treat the global oceans as a single region, hence in order to spatially distribute the estuarine and blue carbon fluxes they have been allocated to the adjacent land, rather than the ocean region. As such, the ocean biogenic component used in this study (3 Tg CH₄ yr⁻¹) was calculated as the difference between the Saunois et al. (2020) total oceanic

biogenic flux (6 Tg CH₄ yr⁻¹) and the total of the new independent regionally distributed estimate of estuarine flux (3 Tg CH₄ yr⁻¹).

Only a single spatially distributed estimate of geological CH₄ emissions, 37 Tg CH₄ yr⁻¹, is available (Etiopie et al., 2019). This estimate is lower than the mean global total reported by Sauniois et al. (2020) but well within their range, 18-65 Tg CH₄ yr⁻¹. Termite emissions are based on four distributions (one each from Kirschke et al. (2013); Sauniois et al. (2016) and two from Sauniois et al. (2020)) and range between 9-18 Tg CH₄ yr⁻¹.

It is important to note that while these four sectors represent the four largest natural non-wetland CH₄ emissions, other natural flux types, such as wild animals, are excluded as their regional distribution was either unknown or poorly characterised. Global estimates of these omitted fluxes are very small, ranging between 1 and 4 Tg CH₄ yr⁻¹, < 0.6% of total emissions (Sauniois et al., 2020). Considering the magnitude of the uncertainties in the inland waters, oceanic and termite estimates (> 100%), it is expected that this omission is easily encompassed in the inherent uncertainty of the *NonWetl* estimates.

2.2 Top-down data

As described in Sauniois et al. (2020, Section 4.2) a comparison of nine different inversions systems together resulting in a total of 22 inversion simulations was conducted. A set of common prior source and sink estimates were developed and these, along with a set of atmospheric observations (Fig. 1), were provided to the participants (Sauniois et al., 2020, Supplementary material). However, their use was not compulsory (See Sauniois et al. (2020) Table S6 for further information). Monthly gridded prior and posterior flux estimates for each of the five broad sub-sectors were submitted by participants for analysis at 1°x1° grid resolution. Further details including the theory of atmospheric inversions, the models and the atmospheric observations used in our study can be found in Sauniois et al. (2020).

There are three key details to note with TD inversions. Firstly, they fit to the observed atmospheric CH₄ mole fraction (i.e. the difference between CH₄ sources and sinks) rather than directly fitting to the individual sources or sinks. As part of this process all but two inverse models (TOMCAT and PYVARLMDz, Table S1) used a spatially varying but 12-month climatological prior estimate of the global CH₄ sink. This assumption is a significant limitation as this sink can vary interannually (Zhao et al., 2019). As such, for TD methods, some temporal variability in the chemical sink may instead be assigned to temporal variability in the source estimates. Similarly, variability in the spatial distribution of the chemical sink may also drive variability in the spatial

distribution of the TD derived regional CH₄ sources. However, as five estimates of the chemical sink spatial distribution were used in our suite of inversions this source of uncertainty should be reflected in the spread of the ensemble of the inversions.

Secondly, rather than optimising for individual source sectors (e.g. *Fossil* or *WetI* fluxes) some inversions optimise for the total flux for each region or pixel and then partition it to source sectors using a fixed regional ratio of sources calculated from prior flux information at pixel scale. This partitioning can lead to significant uncertainties if not all sources increase or change at the same rate in a given region or pixel. However, optimising by pixel rather than region does allow finer spatial scale adjustments which may reduce any aggregation errors. Similarly, uncertainties in the priors can drive uncertainties in the sectoral emissions estimates of systems which optimise for individual sectors as, when relying on CH₄ observations alone (as opposed to the use of CH₄, CH₄ isotopes and related tracers), it is the spatio-temporal distribution of the priors which drives the sectoral differentiation. Here seven of the nine inversion systems optimise for some source sectors (that can differ from our sectors) and the other two (MIRO4-ACTM and PYVARLMDz) solve for net total emissions (Table S1). Thirdly, TD methods generally use BU estimates as prior knowledge and as such BU and TD methods cannot be considered as truly independent.

2.3 Data processing

Datasets were provided on a variety of grids, at different temporal resolutions and over a range of time periods (Supplementary Section S1 and references within Table 1). These datasets were regridded, projected to a common grid and interpolated or extended to a common temporal resolution and time period (Supplementary Section S1). Regions (18 land and 1 ocean) were constructed using geopolitical boundaries and chosen on the basis of size, geopolitical importance and vegetation type (Fig. 4 and Saunio et al. (2020) Table S1). Box plots of emissions estimates were calculated for the period 2008-2017 (Supplementary Section S3).

Rates of change in regional and global emissions estimates were calculated over the 2000-2017 period as described in the Supplementary (Section S4). Comparisons of TD and BU regional emissions were made for the total regional CH₄ emission, the total natural and total anthropogenic emissions and sectoral emissions.

2.3.1 Sector definitions

Methane emission types were aggregated into five key source sectors (see also supplementary S5):

1. Agriculture and waste (*Ag&Waste*) – net emissions related to agriculture, including rice cultivation and ruminant farming, and waste management activities, except agricultural waste burning
2. Fossil fuel (*Fossil*) – emissions related to fossil fuel exploration, extraction, transport, distribution, production and use, including fugitive and industrial emissions
3. Biomass burning and biofuels (*BB&F*) – emissions related to biomass burning (including agricultural waste burning) and biofuel usage
4. Wetlands (*Wetl*) – Net wetland emissions
5. Other natural non-wetlands (*NonWetl*) – The sum of net inland water, net oceanic, geological (land and ocean) and termite emissions

2.3.2 Bottom-up estimates - total anthropogenic and natural

Total BU anthropogenic emissions were calculated as the sum of the *Fossil*, *BB&F* and *Ag&Waste* emissions for each of the emissions inventories. Five BU total anthropogenic emissions estimates were calculated, one for each of the four major anthropogenic emissions inventories and a mean of the four. Total BU natural emissions were calculated as the sum of the mean *NonWetl* emissions estimate and each of the 13 wetland process-based models using the diagnostic wetland area resulting in 13 estimates of total natural emissions.

In an effort to reduce the number of BU total emissions estimates, the mean of the *NonWetl* estimate (i.e. the sum of inland waters, termites, oceans and geological fluxes) and the mean *Wetl* flux was combined with the five anthropogenic estimates to produce five BU estimates of total emissions. However, comparisons between the TD and BU estimates of *Wetl* fluxes were made using the full suite of 13 BU estimates and the mean. Similarly, TD and BU comparisons of *NonWetl* fluxes were made using five BU estimates (one for each termite estimate) and the mean.

2.3.3 Top-down estimates

A total of 22 different gridded TD total source estimates were provided from the nine inversion systems included in Sauniois et al. (2020). Of these, half were based solely on surface CH₄ data (TD SURF) and half included GOSAT satellite data (TD GOSAT). Separate TD SURF and TD GOSAT annual and decadal means were calculated globally and for each region. Unlike Sauniois et al. (2020), here the decadal mean, median and box plot calculations are based only on the SURF data. In some cases, multiple simulations were provided for a given inversion system. To avoid over-weighting these inversion systems the mean of each inversion system's simulations,

rather than each individual simulation, was used to calculate the mean timeseries. However, to demonstrate the spread in the TD estimates the individual simulations were included in the time series figures. See Table S2 for further information.

Total TD anthropogenic emissions were calculated separately for each inversion simulation as the sum of the *Fossil*, *BB&F* and *Ag&Waste* emissions. Similarly, total TD natural emissions were calculated separately for each inversion simulation as the sum of the *NonWetl* (including ocean emissions) and *Wetl* emissions.

3 Results

We first focus on commonalities between the BU and TD estimates, identifying robust features in the spatial distribution and temporal trends of CH₄ emissions with the aim of addressing the four key questions outlined earlier (Section 3.1). Secondly, we highlight key TD and BU differences (Section 3.2).

In the text we report median BU values along with their 25th to 75th percentile range as median [25th-75th percentile] Tg CH₄ yr⁻¹ with the equivalent measures for TD estimates following in parentheses. This reporting approach differs from Saunio et al. (2020), who reported the mean and min-max range. Similarly, when discussing BU and TD estimates as a percentage or changes in emissions rates we report the BU value and give the TD value in parentheses, i.e. BU (TD).

For consistency and completeness when discussing sectoral CH₄ emissions we report both the BU and TD values. However, it is important to note that TD sectoral estimates are dependent on BU estimates and due to the methodology used there are significant uncertainties associated with the TD sectoral estimates (Section 2.2).

3.1 BU and TD commonalities, trends and regional hotspots

3.1.1 Total CH₄ emissions

Mean global total CH₄ emissions for 2008-2017 are estimated to be BU = 689 [25th-75th percentile = 677-697] (TD = 572 [568-581]) Tg CH₄ yr⁻¹. Top-down and bottom-up regional total emissions for the most recent decade (2008-2017) are shown in Fig. 2. Five regions dominate total emissions: China, Southeast Asia, USA, South Asia and Brazil. Together these five regions

account for BU = 43% (TD = 49%) of total global CH₄ emissions, with 298 [279-312] (273 [256-305]) Tg CH₄ yr⁻¹ in the 2008-2017 decade. Comparisons of the 2000-2009, 2003-2012 and 2008-2017 decadal means show these regional rankings to be fairly consistent over time (Table S3). Information in relation to latitudinal flux distribution can be found in Table 5 of Saunio et al. (2020).

3.1.2 Anthropogenic and natural CH₄ emissions

Natural emissions are dominated by five regions - Brazil, Canada, Equatorial Africa, Russia and Southeast Asia (Fig. 3). Together these five regions produce BU = 48% (TD = 52%) of the global natural emissions, with BU = 158 [133-181] (TD = 104 [95-121]) Tg CH₄ yr⁻¹ (Fig. 3). In comparison, regional anthropogenic emissions can be divided into three groups, those > 40 Tg CH₄ yr⁻¹, those 20 to 40 Tg CH₄ yr⁻¹ and those < 20 Tg CH₄ yr⁻¹ (Fig. 3 and Fig. S2a). The two regions in the first group, China, 59 [50-68] (45 [41-55]) Tg CH₄ yr⁻¹, and South Asia, 45 [41-47] (46 [43-49]) Tg CH₄ yr⁻¹, together account for 29% (26%) of global anthropogenic emissions. The second group of regions, which together account for 55% (56%) of anthropogenic emissions, include the USA, Brazil, Europe, Northern and Equatorial Africa, Russia, Middle East and Southeast Asia, each emitting between 20 and 40 Tg CH₄ yr⁻¹. The rest of the world (nine regions) emit ≤ 10 Tg CH₄ yr⁻¹ per region. Median anthropogenic per capita emissions for 2008 to 2017 (Fig. S2d) range from a low of 0.02 Tg CH₄ yr⁻¹ million people⁻¹ (Korea and Japan) to a high of 0.2 Tg CH₄ yr⁻¹ million people⁻¹ (Oceania).

The fraction of regional anthropogenic emissions can differ between TD and BU estimates, with only 8 of the 19 regions agreeing within the 25-75th percentile range (Fig. 3c). However, the relative region-to-region pattern has some similarities with the methods having the five lowest regions in common – namely Canada, Oceans, Southwest South America, Brazil and Northwest South America. Similarly, Europe, Korea & Japan, Middle East, South Asia and China are common to both the TD and BU top six regions when ranked by anthropogenic proportion. Some regions, including China, Middle East and South Asia, are dominated by anthropogenic emissions (BU and TD > 75% of total emissions), while in other regions (e.g. Canada) most emissions come from natural sources (> 70%) (Fig. 3c).

3.1.3 Sectoral CH₄ emissions

The sectoral breakdowns for each of the five global hotspot regions – USA, China, South Asia, Southeast Asia and Brazil – show distinct patterns (Fig. 4). The majority of Chinese emissions BU

= 75% (TD = 81%), totalling BU = 52 [49-59] (TD = 42 [37-56]) Tg CH₄ yr⁻¹, are driven by a roughly 50:50 split of *Fossil* and *Ag&Waste* emissions. In comparison, Brazil and Southeast Asia (along with Southwest South America, Central America and Oceania), show strong *Ag&Waste* emissions, 31% (33%) and 32% (38%), respectively, accompanied by high *WetI* emissions, 45% (58%) and 39% (38%), respectively. While the majority of South Asian CH₄ emissions, 68% (64%), come from *Ag&Waste*. Finally, the USA emissions are split relatively evenly between natural emissions (*WetI* + *NonWetI*), *Fossil* and *Ag&Waste* with 45% (30%), 25% (27%) and 29% (42%), respectively.

Fossil fuel

Fossil emissions are defined here as the sum of coal, oil, gas, industrial and transport emissions, including fugitive emissions from exploration, production and distribution. This sector can in turn be broken down in to four sub-sectors: *Coal*, *Oil&Gas*, *Industry* and *Transport*. Emissions hotspots include China, Russia, the Middle East and the USA who together produced 75 [67-82] (53 [46-77]) Tg CH₄ yr⁻¹ over the 2008-2017 period. This equates to 60% (49%) of total global *Fossil* CH₄ emissions which were 124 [110-143] (107 [88-139]) Tg CH₄ yr⁻¹ (Fig. S3b). Although TD emissions estimates are unable to inform a deep sectoral attribution of these emissions, BU emissions inventories can. These inventories suggest different sub-sectoral mixes between these regions with Chinese *Fossil* emissions dominated (> 80%) by *Coal*, the USA showing a shift from *Coal* to *Oil&Gas* emissions (from 30% to 10% *Coal* and from 60 to 80% *Oil&Gas* over the 2000 to 2017 period), and Russian and Middle Eastern emissions predominantly (> 80%) *Oil&Gas* related (Höglund-Isaksson et al., 2020; Janssens-Maenhout et al., 2019; USEPA, 2012). A detailed sub-sectoral breakdown of the *Fossil* sector for all 19 regions and each of the four BU inventories is given in Fig. S4.

Agriculture & waste

Together South Asia and China account for 32% (33%) of all global *Ag&Waste* emissions producing 65 [61-69] (71 [64-77]) Tg CH₄ yr⁻¹ between 2008 and 2017 (Fig. S3a). A second group of regions, South East Asia, Europe, USA and Brazil, produce > 15 Tg CH₄ yr⁻¹ each over the same period and in combination account for a further 34% (37%) of global emissions. For the following discussion we decompose the *Ag&Waste* sector into three sub-sectors: *Livestock*, *Waste* and *Rice*.

While the exact distribution of Chinese sub-sectoral *Ag&Waste* emissions differs between BU inventories (Fig. S5) most suggest a relatively even split during 2008-2017 between the *Livestock*

(31 to 44%), *Rice* (22 to 31%) and *Waste* (31 to 38%) sectors (Höglund-Isaksson et al., 2020; Janssens-Maenhout et al., 2019; USEPA, 2012). This distribution is significantly different to the South Asian sectoral distribution where *Livestock* alone accounts for 48 to 63% of these types of emissions with the remaining emissions split between *Waste* (18 to 23%) and *Rice* (19 to 32%). The predominance of *Livestock* emissions is again apparent in the Brazil, Europe and USA with an average of 81%, 60% and 56%, respectively, of all 2008-2017 *Ag&Waste* emissions coming from *Livestock*.

Biomass burning and biofuels

The *BB&F* emissions (composed of *Biomass Burning* and *Biofuel* emissions) are generally higher in the African regions with 33% (31%) of total global 2008-2017 *BB&F* emissions, 8 [8-9] (9 [8-10]) Tg CH₄ yr⁻¹, coming from the African continent due to extensive savanna burning. Other regional hotspots of *Biomass Burning* include Southeast Asia 14% (18%), China 13% (13%), South Asia 11% (9%), Brazil 6% (7%) and Russia 5% (6%).

Globally BU emissions inventories together with satellite biomass burning products, indicate a roughly 50:50 split between *Biofuels* and *Biomass Burning*, however, this is not common across individual regions (Höglund-Isaksson et al., 2020; Janssens-Maenhout et al., 2019; Randerson, Chen, van der Werf, Rogers, & Morton, 2012; USEPA, 2012). The regions with the largest populations in this study, China and South Asia, show predominantly (87 to 95%) *Biofuel* emissions driven by the use of biofuels for cooking and household heating in rural areas. In comparison, Brazil and Southern Africa, regions with significant deforestation and drought- and human-induced fire activity (Andela & van der Werf, 2014; Aragão et al., 2018), show mostly (78 to 91%) *Biomass Burning* emissions. While the proportion of *Biomass Burning* to *Biofuel* emissions are generally relatively constant over time within a given region the Southeast Asian region shows a unique pattern. This region has fairly constant *Biofuel* emissions across the 2000-2017 time period of about 1 Tg CH₄ yr⁻¹, however, mean *Biomass Burning* emissions show significant IAV, fluctuating between 0.8 and 5 Tg CH₄ yr⁻¹ (Fig. S1).

Ratios of *Biofuel* to *Fossil* emissions vary between regions and between BU inventories (due to differing biofuel sectorial definitions, see Supplementary S5). However, the relative regional rankings are fairly consistent between BU inventories with South Asia, Equatorial Africa and Southern Africa showing the highest *Biofuel:Fossil* fuel ratios, averaging 0.5, 0.3 and 0.2 respectively (Fig. S6).

Wetlands

Wetl emissions show a distinctive spatial distribution with the three largest contributing regions, Brazil, 15% (19%), Southeast Asia, 15% (12%), and Equatorial Africa, 14% (11%), of total *Wetl* emissions (147 [101-186] (168 [145-204]) Tg CH₄ yr⁻¹) all located on the equator. *Wetl* emissions are also a significant proportion of total emissions in these regions with 45% (58%), 39% (38%) and 41% (46%) of the total regional emissions coming from *Wetl* for Brazil, Southeast Asia and Equatorial Africa, respectively. Other key contributing regions emitting > 10 Tg CH₄ yr⁻¹ each, include Southwest South America, Russia and Canada. On a continental basis South America emits 33% (34%) of all global *Wetl* emissions averaging 48 [36-60] (57 [52-65]) Tg CH₄ yr⁻¹ during the 2008-2017 period.

3.1.4 Total emission rates and temporal trends

The overall temporal trends in TD and BU CH₄ emission means are very similar within each region, with all but one region (Canada) showing agreement in the direction (positive or negative) of trends (Fig. 5 and Fig. S7). Like global CH₄ emissions, which have been steadily increasing over the 2000-2017 period, most regions show an increase in total CH₄ emissions over time. Only Europe and the combined Korea & Japan region exhibit a decrease in total CH₄ emissions between 2000 and 2017. While absolute emission rates often differ between TD and BU estimates (Fig. 5) no region, including Canada, shows a statistically significant difference between the TD and BU emission trend (slope) over the 2000 to 2017 period (student t-test, p=0.05, See Supplementary S4).

Global emissions have increased on average, BU = 4 (TD = 3) Tg CH₄ yr⁻¹ yr⁻¹ or BU = 0.7 (TD = 0.6) % yr⁻¹ relative to 2000. However, rates of change differ between regions (Fig. 5 and Fig. S7). For instance, China increased at more than double the global rate, 2.4 (1.5) % yr⁻¹ relative to 2000. In contrast, Europe and Korea & Japan declined by 0.7 (0.5) and 0.2 (1) % yr⁻¹ relative to 2000, respectively (Figure S6 b).

Both TD and BU estimates link the reduction in European emissions to a 28% (15%) decline in *Fossil* emissions and a 17% (11%) decline in *Ag&Waste* emissions since 2000. BU inventories which separate *Coal* emissions from other *Fossil* emissions show that declines are driven by decreases in *Coal* emissions (> 20% relative to 2000) (Höglund-Isaksson et al., 2020; Janssens-Maenhout et al., 2019; USEPA, 2012). While the GAINS and USEPA inventories show a concurrent decrease in *Oil&Gas* emissions (Höglund-Isaksson et al., 2020; USEPA, 2012).

BU inventories indicated that changes in European *Ag&Waste* emissions were evenly split between reductions in *livestock* emissions (1-2 Tg CH₄ yr⁻¹, 9-15% relative to 2000) and reductions in *waste* emissions (2-3 Tg CH₄ yr⁻¹, 27-30% relative to 2000) (Hoesly et al., 2018; Höglund-Isaksson et al., 2020; Janssens-Maenhout et al., 2019; USEPA, 2012). Inventories that provide further sub-sectoral information show that the decrease in *waste* emissions are solely driven by solid waste (landfill and incineration) processing (Janssens-Maenhout et al., 2019) and that the majority of the *livestock* emissions reductions are due to decreases in enteric fermentation.

The sectoral drivers of decline in Korean & Japanese emissions are not consistent between the four BU inventories. All four inventories show small (0.1 to 0.3 Tg CH₄ yr⁻¹) decreases in *Rice&Livestock* and *Waste* emissions, relative to 2000 with the subsector breakdown of the EDGAR, GAINS and USEPA inventories attributing the decline in *Rice&Livestock* to the *Rice* sector. However, *Fossil* emission trends differ with the CEDS and USEPA inventories showing an increase of 0.2-0.4 Tg CH₄ yr⁻¹ and the more recent inventories, EDGAR and GAINS, a small decline (-0.04 to -0.06 Tg CH₄ yr⁻¹) (Fig. S10). In contrast, all TD emissions estimates show a decline in both total *Fossil* and *Ag&Waste* emissions (Fig. 6).

Both China and the Middle East, regions with emissions rates significantly higher than the global mean, show significant increases in *Fossil* and *Ag&Waste* emissions over the 2000-2017 period. *Fossil* emissions from China increased by 141% (78%) (See also Section 3.2.3) and *Ag&Waste* by 18% (14%), between 2000 and 2017. While Middle Eastern *Fossil* and *Ag&Waste* emissions have climbed by 32% (42%) and 26% (26%), respectively, over the same period. The EDGAR BU inventory alone suggests that the Middle East increase in *Fossil* emissions is dominated by increases in natural gas related emissions (Janssens-Maenhout et al., 2019), while all inventories agree on increases in *livestock* and *waste* emissions (Hoesly et al., 2018; Höglund-Isaksson et al., 2020; Janssens-Maenhout et al., 2019; USEPA, 2012).

Only Southeast Asia and Russia show similar patterns in TD and BU mean IAV. A sectoral decomposition of these regions (Fig. 6) shows that this IAV is driven by a combination of *WetI* and *BB&F*.

While the absolute magnitude of the TD and BU total emissions estimates differ significantly for most regions, there are some regions, USA, Southeast Asia and the three African regions, which appear to agree well (average absolute difference 2000 to 2017 < 15%, SURF data only). On average, absolute differences between regional TD and BU estimates decrease by 3% with the addition of the GOSAT data. Agreement tends to improve in equatorial regions with a paucity of

long-term ground-based observations (Fig. 1). Specifically, Northern South America, Southwest South America, Equatorial Africa and South Asia show a reduction in TD and BU total emission difference of > 7% when using both GOSAT and SURF data (2010 to 2017) (Fig. S8).

Some regions, including USA, Central Asia and Northern Africa, show an acceleration in emissions post 2010 (Fig 5). Both BU and TD methods link the USA post-2010 increase to the *Fossil* sector (Fig 6) with the GAINS and EDGAR BU estimates indicating a > 100% increase in *Oil&Gas* emissions. Other regions, for example Brazil, show a small decline in emissions between 2009 and 2017 with sectoral estimates linking this to a 14% (9%) decline in *Wetl* emissions.

3.2 TD and BU differences

3.2.1 Total CH₄ emissions

As identified in Saunois et al. (2020) and demonstrated again here, there is a significant difference in the global TD and BU total emissions estimates, most likely driven by double counting of fluxes from wetlands and inland waters, and extrapolation issues (e.g. Thornton, Wik, & Crill, 2016). Here we find the 2008-2017 median BU estimate to be 136 Tg CH₄ yr⁻¹ higher than the median TD estimate. This offset (along with the total and sectoral TD and BU emissions estimates) differs from that of Saunois et al. (2020) as here we exclude the GOSAT estimates from the decadal means, use a new inland waters estimate and use one less BU agricultural estimate (See Section 2). However, while the 2008-2017 global mean BU total of this paper is 48 Tg CH₄ yr⁻¹ lower than the Saunois et al. (2020) mean it is within the (wide) range of their global BU emissions estimates, 594 to 881 Tg CH₄ yr⁻¹. Our lower estimate is predominantly due to a lower inland waters estimate, 129 cf. 159 Tg CH₄ yr⁻¹, and a lower oceanic emissions estimate 3 cf. 13 Tg CH₄ yr⁻¹.

For 18 of the 19 regions TD total CH₄ posterior emissions estimates are closer to their prior than the BU estimate (Fig. 2). However, this trend is not consistent across the subsectors (e.g. Fossil, Fig. S3b). For 16 of the 19 regions examined median regional BU estimates of total CH₄ flux are higher than posterior TD estimates (Fig. 2). However, only four regions (Canada, Central Asia, Russia and Oceania, Fig. 2) do not agree within the 25-75th percentile range, with all these regions showing total BU >> TD. As with global estimates regional BU – TD discrepancies are driven by BU *NonWetl* emissions estimates (Fig. S3a-d cf. S3 e), primarily elevated *Inland Waters* emissions (Fig. S9).

Although TD sectoral partitioning can be problematic (Section 2.2), it is interesting that for the two land regions with higher TD than BU estimates, Brazil and South Asia, this difference is caused primarily by *Wetl* emissions. Here the Brazil and South Asia TD *Wetl* estimates, 33 [31-35] and 13 [11-15] Tg CH₄ yr⁻¹, respectively, are > 45% higher than the BU estimates, 23 [15-28] and 4 [3-5] Tg CH₄ yr⁻¹ (Figs. 4 and 6). Even though this is a large discrepancy between median estimates, there are some BU wetland models that do find higher *Wetl* emissions. For example, the LPX-Bern model has a Brazilian mean 2008-2017 *Wetl* emission estimate of 31 Tg CH₄ yr⁻¹ and the DLEM model a South Asian mean 2008-2017 *Wetl* emission estimate of 16 Tg CH₄ yr⁻¹ (see Table 2 Saunio et al. (2020) for further information on individual wetland models). In addition, the mean of BU wetland models run using a prognostic extent mode (where wetland extent is determined by the model) is significantly closer to those of the TD estimates for these two regions, 40 and 11 Tg CH₄ yr⁻¹ for Brazil and South Asia, respectively. While the higher TD *Wetl* estimates may not be correct there are known issues with wetland mapping, particularly in flooded forest regions where closed canopies make remote sensing of water-saturated soils challenging (Prigent, Jimenez, & Bousquet, 2020). As such, the TD-BU *Wetl* discrepancy along with the difference in prognostic and diagnostic BU wetland models suggests that there may be room for improvement in the diagnostic wetland map in the Brazilian and South Asian regions. It is also interesting to note that the addition of the GOSAT data to the TD inversion estimate decreases the TD *Wetl* estimate in the South Asia region by approximately 5 Tg CH₄ yr⁻¹. This result suggests that where surface site numbers are limited (Fig. 1) the TD-BU discrepancy may be partially driven by data coverage, emphasizing the importance of satellite data.

3.2.2 Temporal trends in total CH₄ emissions

Canada is the only region to show a statistically different trend in TD and BU emissions, with TD estimates showing a gradual decrease in CH₄ emissions and BU estimates showing an increase (Figs 5 and 6). The sectoral breakdown of the BU and TD emissions estimates shows this difference to be driven by *Wetl* emissions with a downward trend in *Wetl* emissions, ~0.3 Tg CH₄ yr⁻², present in all but one of the TD SURF emissions estimates. This contrasts with BU process model estimates which, while varying in absolute magnitude, all show a small upward trend in *Wetl* emissions.

3.2.3 Sectoral CH₄ emissions

As discussed earlier (Section 3.2.1), high BU *Inland Waters* estimates drive discrepancies in TD and BU total emissions estimates. These also lead to higher BU estimates of natural emissions

(Fig. 3a) and in turn (for all regions other than South Asia), higher TD estimates of anthropogenic proportion (Fig. 3c). The discrepancy in the Central Asia region is particularly large with a median TD anthropogenic proportion of 89% and a BU estimate of 37%. This difference is driven by the far higher (~15 times) BU estimate of natural emissions, 86% of which is *Inland Waters* (Fig. S9). Unlike land region estimates, in the ocean region a higher TD estimate of anthropogenic proportion, is driven by a higher (~3.5 times) TD estimate of anthropogenic *Fossil* emissions.

Fig. 6 shows annual mean values for the TD and BU sectoral emissions estimates for the 2000 to 2017 period. This figure shows clear differences between the temporal trends in TD and BU estimates of *Wetl*, *NonWetl* (both discussed previously in Section 3.2.1) and *Fossil* emissions for many regions. While there can be significant methodologically driven uncertainties with TD sectoral estimates (Section 2), when optimising for total CH₄ fluxes seven of the nine TD estimates (Table S1) used here are independently optimised for at least anthropogenic (if not *Fossil* specific) emissions. As such, the TD *Fossil* estimates while treated with caution are reported here and used to inform the discussion.

For most regions, the TD and BU *Fossil* estimates are similar over the 2000-2017 period; however, there are notable differences, particularly for China, the Middle East, Russia and Northern South America where TD and BU estimates differ by up to 10 Tg CH₄ yr⁻¹. During the year 2000 the Chinese CEDS *Fossil* estimate (upper blue line, Fig. S10) is similar to the other three BU inventories but then rapidly accelerates and by 2017 is almost double that of the other three BU inventories. As noted earlier, the *Fossil* component of the CEDS inventory is based on an earlier EDGAR v4.2 inventory which is known to overestimate *Coal* emissions in China (See Section 2.1.1). However, limiting the analysis to the remaining three BU inventories only reduces the annual mean TD and BU difference by 20%. In fact, the mean BU estimate (CEDS excluded) is consistently higher than all but two of the individual TD estimates with only one of the BU estimates, USEPA, within the range of the majority of the TD estimates. A closer examination of the sub-sectoral breakdown of the BU inventories suggests that the EDGAR and GAINS *Fossil* estimates are higher than the other inventories due to a combination of higher *Oil&Gas* emissions (10 times the USEPA inventory, Fig. S4) and *Coal* emissions (up to 50% higher than the USEPA inventory, Fig. S4).

There are large discrepancies between the TD and individual BU inventory estimates of Northern South American *Fossil* emissions (Fig. S10). The CEDS estimate is consistently (> 4 times) higher than the other three BU inventories and the TD estimates (Fig. S4 and S10). Interestingly, a closer examination of the gridded CEDS data shows the majority of these emissions occurring from a single point source in Venezuela. This assessment agrees with other inversion studies

(Maasakkers et al., 2019; Y. Zhang et al., 2020) which suggest an under-estimate in Venezuelan *Oil&Gas* emissions in the EDGAR and UNFCCC estimates. However Maasakkers et al. (2019) only revised these estimates to $\sim 5 \text{ Tg CH}_4 \text{ yr}^{-1}$ i.e. far less than the $\sim 12 \text{ Tg CH}_4 \text{ yr}^{-1}$ allocated by the CEDS emissions inventory.

4 Discussion

4.1 Emissions and policy relevance

Four key regions (China, South Asia, Southeast Asia and USA) together account for $> 40\%$ of global anthropogenic CH_4 emissions (Fig. 3 and Fig. S2a). Most of these emissions come from a combination of the *Fossil* (in particular *Coal* and *Oil&Gas*) and *Ag&Waste* (in particular *Livestock*) sectors (Section 3.1.3). The predominance of *Fossil* and *Ag&Waste* emissions in these regions of high total CH_4 emissions highlights the importance of these sectors as opportunities for significant emissions mitigation.

In general, anthropogenic emissions can be divided into three groups roughly correlating with the size of their regional populations (Fig. 3 and Fig. S2a). The group 1 regions (China and South Asia), those with the largest anthropogenic emissions (combined $> 25\%$ of global anthropogenic emissions) are also the regions with the largest populations ($> 40\%$ of the global population (United Nations, 2019)). While the second group of regions account for $> 55\%$ of anthropogenic emissions and the third group $< 20\%$. This breakdown suggests that relatively large mitigation opportunities exist in group 1 and 2 nations, while smaller but substantial opportunities exist in other regions.

China has the largest anthropogenic CH_4 emissions of any region ($> 13\%$ of the total) and one of the fastest growing trends with a BU = 51% (TD = 27%) increase between 2000 to 2017. This growth is driven predominantly by increases in *Coal* and *Ag&Waste* emissions, with BU inventories suggesting that the increase in Chinese *Coal* emissions alone accounts for $> 15\%$ of the global total emissions increase. Increases in Chinese coal mining activities have been linked to rapid economic growth, which relies predominantly on coal-based energy sources (Jackson et al., 2018; Miller et al., 2019; Peters et al., 2017). However, recent Chinese inventory studies suggest a decline in coal mining emissions since 2012 (Gao, Guan, & Zhang, 2020; Sheng, Song, Zhang, Prinn, & Janssens-Maenhout, 2019). Similarly, increases in Chinese *Waste* emissions have been driven by economic growth and urbanisation. This sector, however, has

great mitigation potential as previous work reports that changes in landfill procedures could lead to a 30-50% reduction in landfill emissions (Cai et al., 2018). The increases in agricultural CH₄ emissions are being driven by diet changes associated with the rising standard of living and the promotion of the agricultural economy (Tian, Zhang, & He, 2014; B. Zhang & Chen, 2010). Tian et al. (2014) also emphasize that given China's economic dependence on agriculture, decreases in agricultural emissions will only come from changes in production methods not a reduction in agricultural capacity.

In contrast, anthropogenic emissions from Europe and Korea & Japan have decreased by 20% (13%) and 7% (14%), respectively. Specifically, decreases in European *Livestock* emissions (mainly enteric fermentation) have been linked to decreases in livestock numbers driven by reforms to the EU common agricultural policy (CAP) (EUROSTAT, 2017; Vergé, De Kimpe, & Desjardins, 2007). However, livestock numbers have remained unchanged since 2008 (FAO, 2019a) and future reductions may instead need to come from changes in farm management, feed composition and additives and selective breeding (EIP-AGRI, 2017; Pellerin, Bamière, & Pardon, 2014; Roque et al., 2021). Reductions in European solid waste emissions have been driven by the EU Landfill Directive 1999/31/EC which aimed to reduce the amount of biodegradable waste entering landfills sites (EUROSTAT, 2014). Although the economic and policy drivers of European CO₂ emissions have been examined in detail elsewhere (e.g. Le Quéré et al. (2019)), similar studies of CH₄ appear lacking in the literature. Reports describing total Japanese greenhouse gas emissions (as opposed to solely CH₄ emissions) link the decline in these emissions to the adoption of renewable energy, a resumption of nuclear energy and a reduction in energy consumption due to improved energy conservation and reduced manufacturing production (Ministry of the Environment, 2021). This agrees well with the reductions in TD *Fossil* emissions and those of the *Coal* sector evident in the GAINS and EDGAR BU estimates. Similarly, reductions in Japanese *Waste* and *Rice* emissions (evident in EDGAR, GAINS and USEPA inventory estimates) align with Japanese governmental policies which propose decreasing methane emissions through the reduction of landfill organic waste disposal and altering rice farming practices (Ministry of the Environment, 2014). However, decreases in these emission estimates were evident prior to the 2014 policies making the contribution of environmental policy unclear.

Several TD studies have found that USA anthropogenic (in particular *Oil&Gas* and *Livestock*) emissions are underestimated in BU inventories (e.g. Maasackers et al., 2021; Williams, Regehr, & Kang, 2021; Y. Zhang et al., 2020). Considering that the USA is the third largest contributor to global anthropogenic emissions and that BU, rather than TD, estimates are used to track and

assess policy outcomes, resolving this discrepancy would be valuable. However, while the median BU USA anthropogenic emissions estimate (28 Tg CH₄ yr⁻¹ for 2008-2017) is lower than the TD estimate (31 Tg CH₄ yr⁻¹) they do agree within the 25-75th percentile range (Fig. 3) and as such our data suggest that the methods may well be consistent.

4.2 Methodological and science implications

In this paper we explored a range of TD and BU emissions estimates with the aim of providing policy-relevant information with sector specific emissions and trend data that is necessary for developing effective regional CH₄ mitigation strategies and protocols. Due to methodological limitations (Sections 2.2 and 3.2.3), comparisons between TD and BU emissions estimates should be treated with care. However, our comparisons of Brazilian and South Asian TD and BU *Wetl* estimates (Section 3.2.1) and TD and BU *Fossil* estimates (Section 3.2.3) demonstrate their power to identify methodological issues in current reporting approaches and areas requiring further study. While, clearly useful in their current form, TD and BU estimates could be improved.

Regional BU inventory estimates could be enhanced with the implementation of time varying regional and technologically specific emissions factors and checks on the reported activity data. There are also significant differences in the spatial distribution of sectoral emissions between inventories suggesting that further work is required to accurately map emissions regionally. Encouragingly, efforts to implement these suggestions and to use TD estimates to inform and improve European BU inventory estimates are already underway as part of the VERIFY project (<https://verify.lsce.ipsl.fr/index.php>). However, it is important that such efforts are implemented world-wide, particularly in other regions with large anthropogenic emissions.

The low accuracy of natural flux estimates limits our ability to separate anthropogenic and natural flux estimates. Required improvements are the development of gridded inland water estimates, supported by a long-term global inland water monitoring network, the development of process-based models, use of observed site-level CH₄ emissions for model benchmarking (e.g., FLUXNET-CH₄; Delwiche et al. (2021)), and the avoidance of double counting with the use of comprehensive inland water models encompassing all water body types (e.g. lakes, ponds and rivers) and using spatial masks consistent with those used by wetland models.

Many TD methods rely on the use of fixed OH climatology (i.e. no IAV) which may artificially bias TD emissions estimates. In our study, emissions calculated with independent OH field optimisation (allowing for IAV) show similar or only slightly larger CH₄ emissions than those without OH IAV. Similarly, Zhao et al. (2019) examined a range of OH scenarios and suggested

that IAV OH offsets 7-20% of the increase in global CH₄ emissions between 2000-2010. This increase roughly equates to a global emissions reduction of 3-11 Tg CH₄ yr⁻¹. While small compared to the global 136 Tg CH₄ yr⁻¹ TD-BU discrepancy any improvement in accuracy is important. Similarly, the spatial distribution of the TD chemical sink prior can influence the regional distribution of TD derived CH₄ sources. However, this effect is likely to be small with Zhao et al. (2019) finding that large (~50%) variability in the OH spatial distribution only equated to a 1.5% variability in regional CH₄ mixing ratios. While only a proxy for variability in the true OH CH₄ sink this is in an order of magnitude smaller than the mean range of TD emissions estimates and easily contained within the current uncertainty range.

Other key developments in TD approaches include improvements in atmospheric transport models (especially vertical transport, stratospheric transport and the simulation of boundary layer dynamics), methodological improvements in TD sector specific flux estimation using isotopic tracers and co-emitted compounds (e.g. ethane) as additional constraints and the use of the latest satellite data (e.g. TROPOMI <http://www.tropomi.eu>).

5 Conclusions

Five regions – China, Southeast Asia, USA, South Asia and Brazil – account for > 40% of all CH₄ emissions during the decade of 2008-2017. Emissions from most of these regions, like global emissions, are equally contributed by anthropogenic and natural sources, except China and South Asia where emissions are predominantly (> 75%) anthropogenic. *Livestock* (46%) and *Rice* (20%) account for the bulk of anthropogenic emissions from South Asia, while *Fossil* accounts for 40% of China's anthropogenic emissions with most of the remaining emissions equally distributed between *Livestock*, *Rice* and *Waste*.

Annual global CH₄ emissions increased by ~10% (50-70 Tg CH₄) between 2000 and 2017 (Jackson et al., 2020). Regionally China, the Middle East, South Asia and Southeast Asia are the largest contributors to this rise, together accounting for 62-67% (33-46 Tg CH₄) of the global increase. Increases in China and the Middle East are driven by *Fossil* emissions, with the increase in *Coal* emissions from China alone accounting for ~15% of the global emissions increase. Increases in South Asia are related predominantly to *Livestock* and *Waste*, whereas those in Southeast Asia are more equally divided between *Coal*, *Rice*, *Livestock* and *Waste* emissions.

In contrast, emissions from Europe and Korea & Japan decreased by ~10%. European emissions reductions were linked to decreases in *Waste* (mainly solid waste disposal), *Livestock* and *Coal*,

which on average accounted for 43%, 26% and 22% of the decline respectively. Japanese emissions decline at a similar rate to European emissions, driven by reductions in *Waste* and *Rice* emissions, however absolute emissions are an order of magnitude smaller.

The *Coal*, *Waste* and *Livestock* sub-sectors are common drivers of rapid increases and decreases in regional CH₄ emissions. As such, these sectors present the best opportunities for CH₄ emissions mitigation and the largest risks (drivers) of on-going emission increases.

Regions with significant natural emissions (e.g. Brazil with 22-34 Tg CH₄ yr⁻¹ from *Wetl*) are also key, with the influence of climate change on these sources an area of concern. Some studies (e.g. Z. Zhang et al., 2018) suggest that changes in temperature and precipitation patterns may have already led to increases in CH₄ *Wetl* emissions growth rates. However, as these flux types have no or limited mitigation potential the focus when implementing emission reduction schemes should be on regions with a large proportion of anthropogenic emissions.

Considering the large contribution of Global CH₄ emissions to the Earth's radiative budget, the predominance of increasing (rather than decreasing) CH₄ emissions is a large concern. However, the emissions decline observed in Europe and Korea & Japan provides evidence that a decrease in emissions in response to government policies and/or economic forces is possible.

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7 Conflict of interest

The authors declare that there are no competing financial interests.

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9 Data availability statement

A number of datasets were used to support the findings of this study. These datasets are either openly available in repositories as listed in the text and references, available on request from the original developers (see text and references there in) or are available from the corresponding author upon reasonable request.

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Figure captions

Fig. 1 – The locations of the surface (triangle) and ground-based profile (circle) observation sites and GOSAT XCH₄ data density and extent (number of data points per grid cell per month for NIES full physics retrievals) for 2014. Grid cells are 2.5 x 2.5°. Note that many of the in situ sites are not operational or are not reporting data to the Global Atmospheric Watch repository. Also note that many of the GOSAT grids do not have uniform data coverage in all months as the instrument cannot see through the cloud covered areas and during polar nights.

Fig. 2 – Boxplots of the total source estimates for the most recent decade (2008-2017) for each region for the bottom-up (BU – red boxes) and top-down surface sites only (TD SURF – blue boxes). The priors used in the TD SURF estimate are also shown (empty boxes). The box represents the 25th to 75th percentile range, “x” represents the outliers and the whiskers show the minimum and maximum values (outliers excluded).

Fig. 3 – (a) natural (green) emissions by region, (b) mean anthropogenic (brown) and (c) mean anthropogenic proportion as a percentage of total regional emissions (brown) in Tg CH₄ yr⁻¹ for 2008-2017. Bottom-up (BU) estimates are shown as filled boxes and top-down surface sites only (TD SURF) as open boxes. The box represents the 25th to 75th percentile range, “x” represents the outliers and the whiskers show the minimum and maximum values (outliers excluded).

Fig. 4 – The sectorial budget breakdown for each region in Tg CH₄ yr⁻¹ for the most recent decade (2008-2017) and a map of the 18 land regions used in this study. Each panel shows methane emissions for the five key sectors, from left to right: wetlands, biomass burning & biofuels, fossil fuels, agriculture & waste and other natural (including inland waters). Top-down estimates are shown on the left as light-coloured boxes and bottom-up estimates on the right as dark-coloured boxes. Sectoral emission estimates based on less than 5 data points are shown as open boxes and those with 5 or greater data points are shown as filled boxes. The box represents the 25th to 75th percentile range, “x” represents the outliers and the whiskers show the minimum and maximum values (outliers excluded).

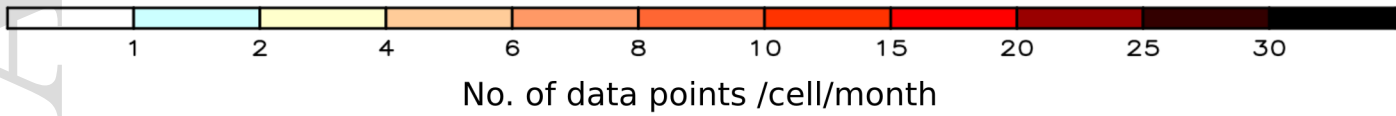
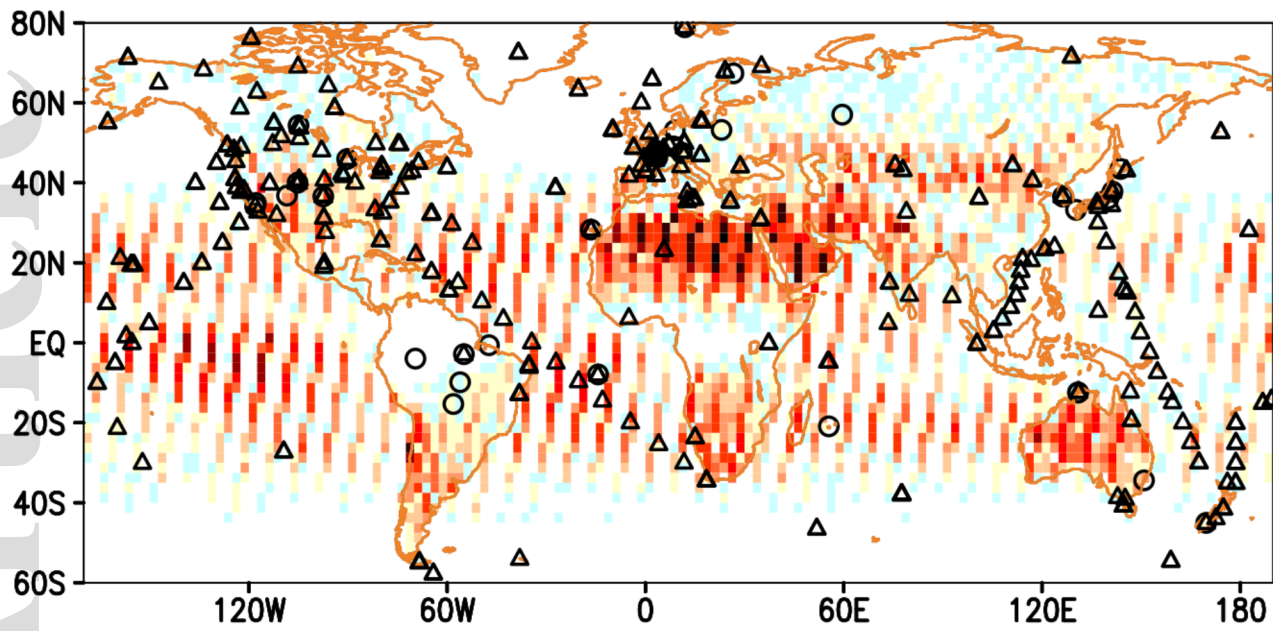
Fig. 5 – Annual estimates of total CH₄ emissions in Tg CH₄ yr⁻¹ for each of the 18 land regions, the ocean region and the global total over 2000-2017. Bottom-Up (blue) and Top-Down (red –

surface sites only, magenta – surface sites and satellite data). Thick lines indicate the mean estimate while thin lines represent individual estimates.

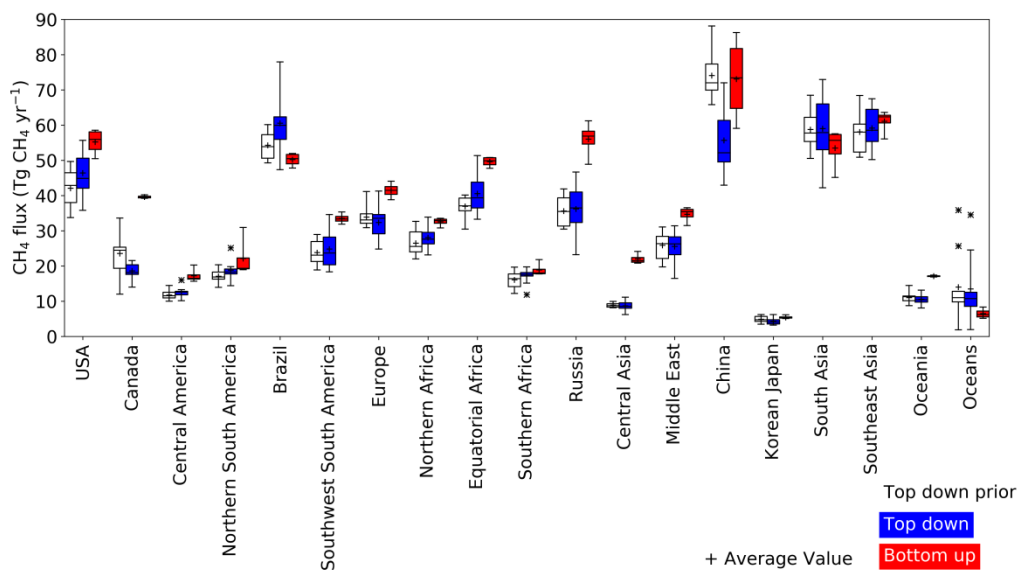
Fig. 6 – Yearly mean regional and global agriculture & waste (orange), biomass burning and biofuel (red), fossil fuels (black), other natural (blue) and wetland (green) fluxes in Tg CH₄ yr⁻¹ over 2000 to 2017. The mean of the bottom-up estimates are shown as a solid line, the mean of the top-down (surface sites only) as a dashed line and the mean of the top-down estimates including GOSAT data as a dotted line.

Table 1 – Components of the total bottom-up emission estimates and references. The inland water estimate was developed for this study and was not used in Saunois et al. (2020).

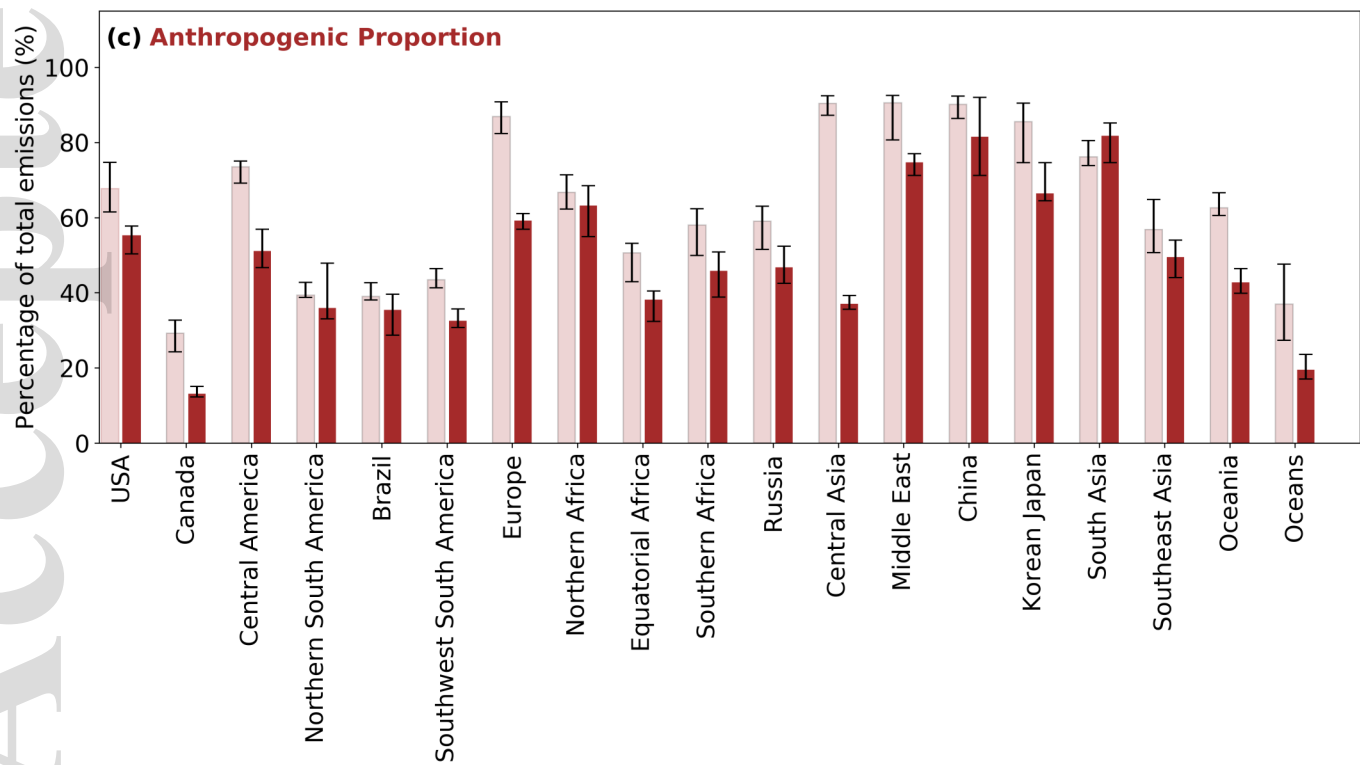
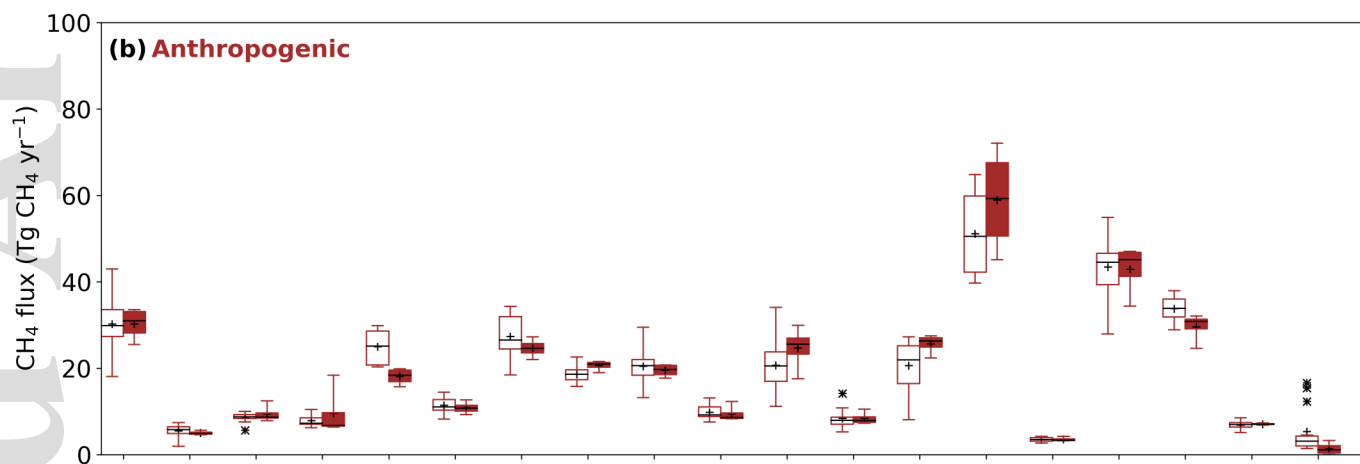
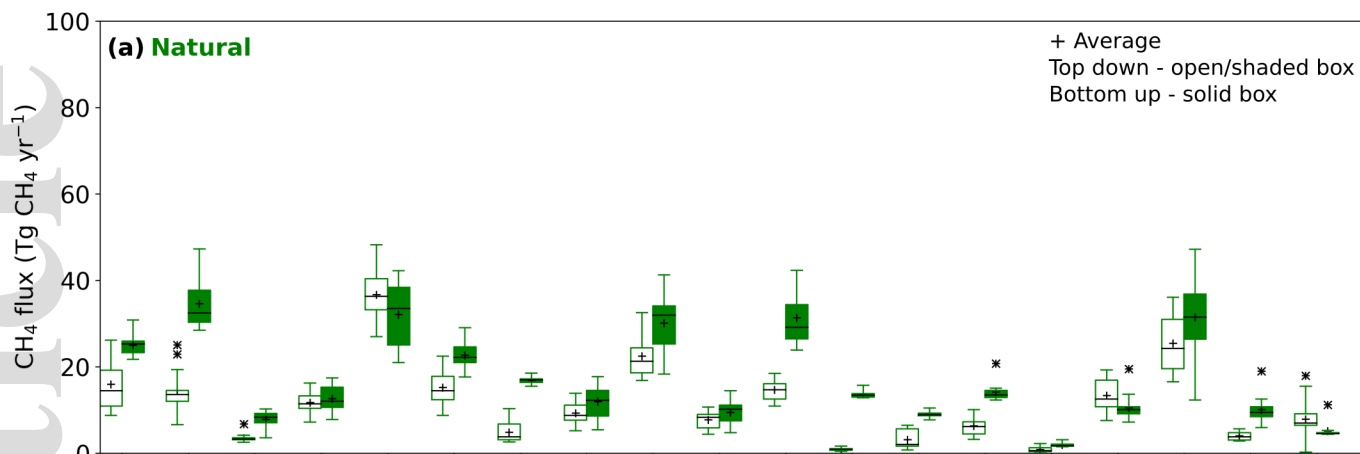
Sector abbreviation	Subsector	Information type	Bottom-up estimate			
			CEDS	EDGAR	GAINS	USEPA
Fossil	Total Fossil					
Ag&Waste	Agriculture & Waste		CEDS (Hoesly et al., 2018)	EDGAR v4.3.2 (Janssens-Maenhout et al., 2017)	GAINS ECPLISE v6 (Gómez-Sanabria et al., 2018; L. Höglund-Isaksson, 2012; Lena Höglund-Isaksson, 2017)	USEPA (USEPA, 2012)
BB&F	Biofuels	Temporally and spatially varying	BB4CMIP			
	Biomass Burning		(van Marle et al., 2017)		GFEDv4.1s (van der Werf et al., 2017)	
Wetl	Wetlands		Ensemble mean of wetlands models (Poulter, 2020)			
Non-Wetl	Geological	Spatially but not temporally varying	Etiopé et al. (2019)			
	Termites		Kirschke et al. (2013), Saunois et al. (2016) and Saunois et al. (2020)			
	Oceanic (biogenic) including estuaries and blue carbon		Ocean total of 6 Tg CH ₄ yr ⁻¹ as described in Saunois et al. (2020)			
	Inland waters – lakes and reservoirs	This paper section 2.1.4 and Supplementary S.4.				
	Wild animals	Global totals only	Excluded from total			



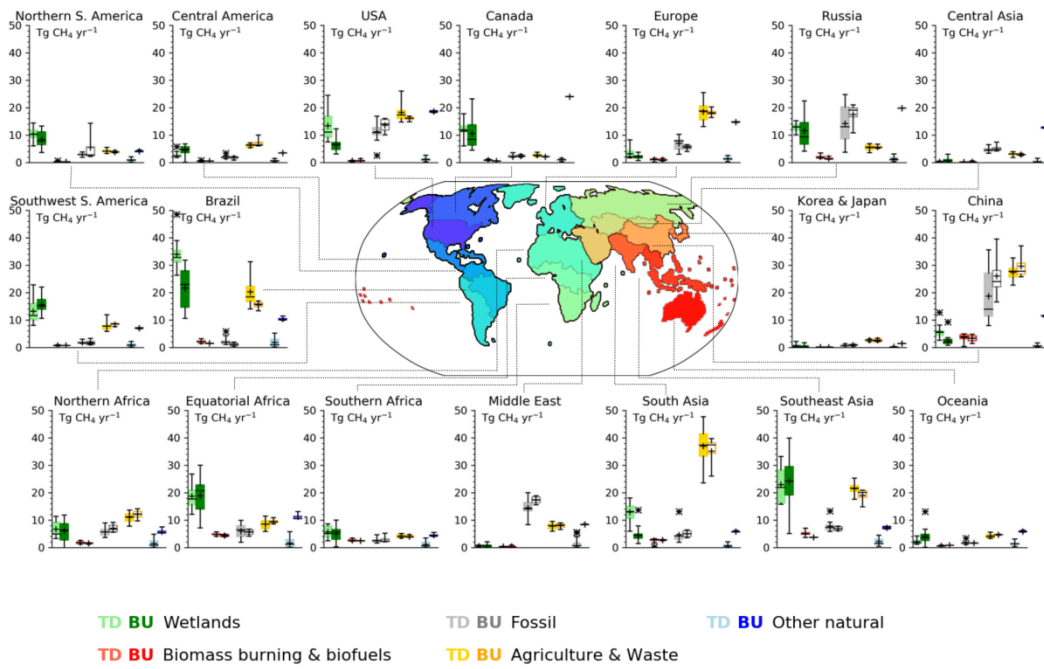
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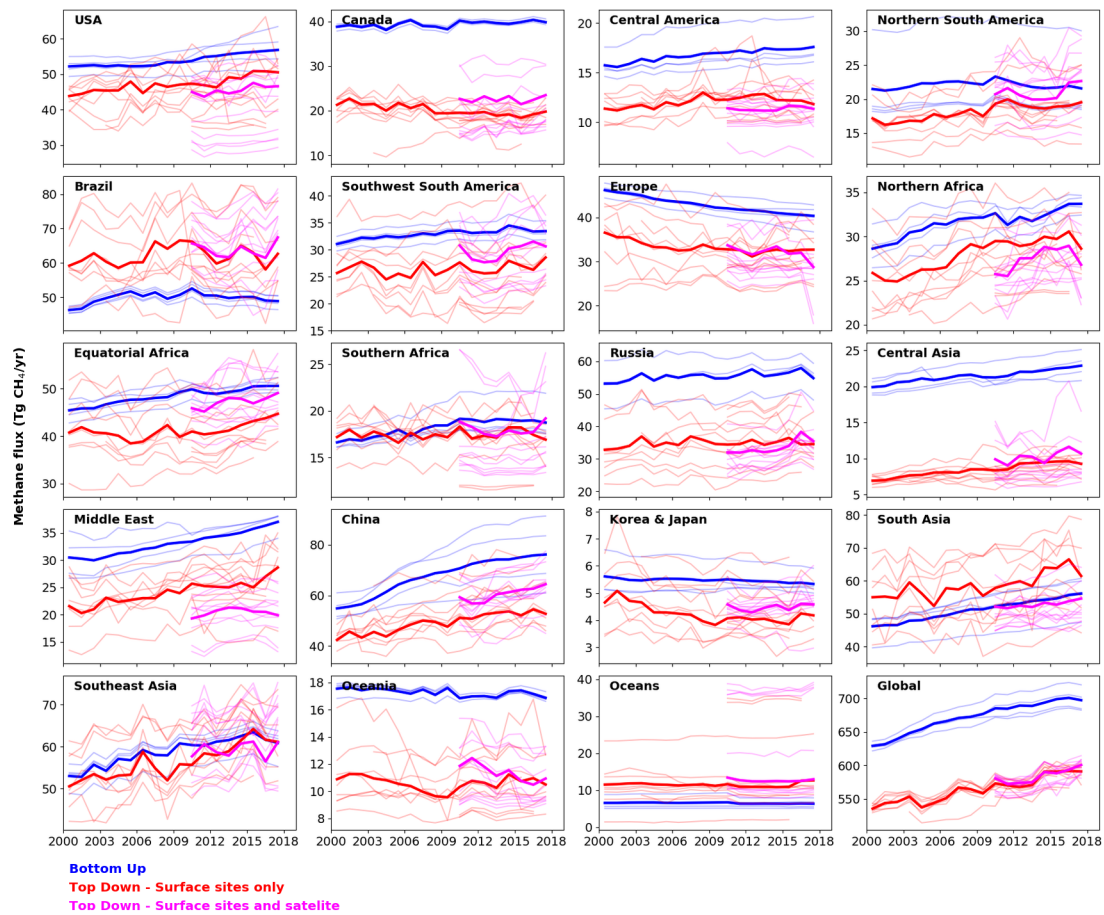
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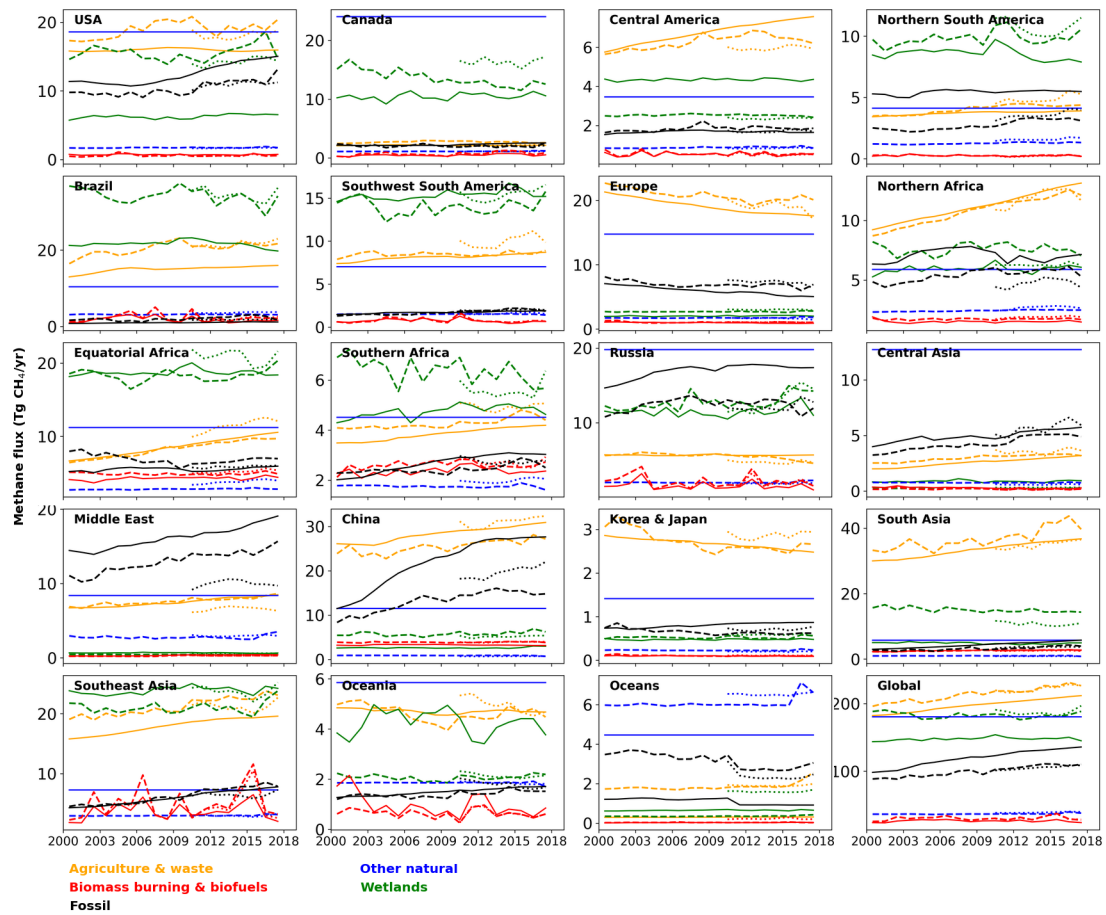
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