1 Global Methane Budget 2000-2020

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- 114 Abstract. Understanding and quantifying the global methane (CH₄) budget is important for assessing realistic pathways to
- 115 mitigate climate change. CH4 is the second most important human-influenced greenhouse gas in terms of climate forcing
- after carbon dioxide (CO₂) and both emissions and atmospheric concentrations of CH₄ continue to increase since 2007 after
- 117 a temporary pause. The relative importance of CH4 emissions compared to those of CO2 for temperature change is related
- 118 to its shorter atmospheric lifetime, stronger radiative effect, and acceleration in atmospheric growth rate over the past decade,
- 119 the causes of which are still debated. Two major challenges in guantifying the factors responsible for the observed
- 120 atmospheric growth rate arise from diverse, geographically overlapping CH4 sources and from the uncertain magnitude and
- 121 temporal change in the destruction of CH4 by short-lived and highly variable hydroxyl radicals (OH). To address these
- challenges, we have established a consortium of multi-disciplinary scientists under the umbrella of the Global Carbon Project
- to improve, synthesise and update the global CH₄ budget regularly and to stimulate new research on the methane cycle.
- Following Saunois et al. (2016, 2020), we present here the third version of the living review paper dedicated to the decadal
- 125 CH₄ budget, integrating results of top-down CH₄ emission estimates (based on in-situ and greenhouse gas observing satellite
- 126 (GOSAT) atmospheric observations and an ensemble of atmospheric inverse-model results) and bottom-up estimates (based 127 on process-based models for estimating land-surface emissions and atmospheric chemistry, inventories of anthropogenic
- emissions, and data-driven extrapolations). We present a budget for the most recent 2010-2019 calendar decade (the latest
- 129 period for which full datasets are available), for the previous decade of 2000-2009 and for the year 2020.

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139 The revision of the bottom-up budget in this 2024 edition benefits from important progress in estimating inland freshwater 140 emissions, with better accounting of emissions from lakes and ponds, reservoirs, and streams and rivers. This budget also 141 reduces double accounting across freshwater and wetland emissions and, for the first time, includes an estimate of the 142 potential double accounting that may exist (average of 23 Tg CH4 yr⁻¹). Bottom-up approaches show that the combined 143 wetland and inland freshwater emissions average 248 [159-369] Tg CH₄ yr⁻¹ for the 2010-2019 decade. Natural fluxes are 144 perturbed by human activities through climate, eutrophication, and land use. In this budget, we also estimate, for the first 145 time, this anthropogenic component contributing to wetland and inland freshwater emissions. Newly available gridded products also allowed us to derive an almost complete latitudinal and regional budget based on bottom-up approaches. 146 147 For the 2010-2019 decade, global CH4 emissions are estimated by atmospheric inversions (top-down) to be 575 Tg CH4 yr 148 ¹ (range 553-586, corresponding to the minimum and maximum estimates of the model ensemble). Of this amount, 369 Tg 149 CH4 yr⁻¹ or ~65% are attributed to direct anthropogenic sources in the fossil, agriculture and waste and anthropogenic biomass burning (range 350-391 Tg CH yr⁻¹ or 63-68%). For the 2000-2009 period, the atmospheric inversions give a 150 slightly lower total emission than for 2010-2019, by 32 Tg CH₄ yr⁻¹ (range 9-40). The 2020 emission rate is the highest of 151 152 the period and reaches 608 Tg CH₄ yr⁻¹ (range 581-627), which is 12% higher than the average emissions in the 2000s. Since 153 2012, global direct anthropogenic CH₄ emission trends have been tracking scenarios that assume no or minimal climate 154 mitigation policies proposed by the Intergovernmental Panel on Climate Change (shared socio-economic pathways SSP5 155 and SSP3). Bottom-up methods suggest 16% (94 Tg CH4 yr⁻¹) larger global emissions (669 Tg CH4 yr⁻¹, range 512-849) than top-down inversion methods for the 2010-2019 period. The discrepancy between the bottom-up and the top-down 156 157 budgets has been greatly reduced compared to the previous differences (167 and 156 Tg CH₄ yr⁻¹ in Saunois et al. (2016, 158 2020), respectively), and for the first time uncertainty in bottom-up and top-down budgets overlap. Although differences 159 have been reduced between inversions and bottom-up, the most important source of uncertainty in the global CH4 budget is 160 still attributable to natural emissions, especially those from wetlands and inland freshwaters. 161 The tropospheric loss of methane, as the main contributor to methane lifetime, has been estimated at 563 [510-162 663] Tg CH₄ yr⁻¹ based on chemistry climate models. These values are slightly larger than for 2000-2009 due to the impact 163 of the rise in atmospheric methane, and remaining, large uncertainty (~25%). The total sink of CH4 is estimated at 633 164 [507-796] Tg CH4 yr-1 by the bottom-up approaches and at 554 [550-567] Tg CH4 yr-1 by top-down approaches. Though, 165 most of the top-down models use the same OH distribution, which introduces less uncertainty to the global budget than is 166 likely justified. 167 For 2010-2019, agriculture and waste contributed an estimated 228 [213-242] Tg CH4 yr⁻¹ in the top-down budget and 211 168 [195-231] Tg CH4 yr⁻¹ in the bottom-up budget. Fossil fuel emissions contributed 115 [100-124] Tg CH4 yr⁻¹ in the top-169 down budget and 120 [117-125] Tg CH4 yr⁻¹ in the bottom-up budget. Biomass and biofuel burning contributed 27 [26-

170 27] Tg CH₄ yr⁻¹ in the top-down budget and 28 [21-39] Tg CH₄ yr⁻¹ in the bottom-up budget

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191 We identify five major priorities for improving the CH4budget: i) producing a global, high-resolution map of water-saturated 192 soils and inundated areas emitting CH₄ based on a robust classification of different types of emitting ecosystems; ii) further 193 development of process-based models for inland-water emissions; iii) intensification of CH4 observations at local (e.g., 194 FLUXNET-CH4 measurements, urban-scale monitoring, satellite imagery with pointing capabilities) to regional scales 195 (surface networks and global remote sensing measurements from satellites) to constrain both bottom-up models and 196 atmospheric inversions; iv) improvements of transport models and the representation of photochemical sinks in top-down 197 inversions, and v) integration of 3D variational inversion systems using isotopic and/or co-emitted species such as ethane 198 as well as information in the bottom-up inventories on anthropogenic super-emitters detected by remote sensing (mainly oil 199 and gas sector but also coal, agriculture and landfills) to improve source partitioning.

200 The data presented here can be downloaded from https://doi.org/10.18160/GKQ9-2RHT (Martinez et al., 2024).

201 1 Introduction

202 The average surface dry air mole fraction of atmospheric methane (CH4) reached 1912 ppb in 2022 (Fig. 1-Lan et 203 al., 2024), 2.6 times greater than its estimated pre-industrial value in 1750. This increase is attributable in large part to 204 increased anthropogenic emissions arising primarily from agriculture (e.g., livestock production, rice cultivation, biomass 205 burning), fossil fuel production and use, waste disposal, and alterations to natural CH4 fluxes due to increased atmospheric 206 CO₂ concentrations, land use (Woodward et al., 2010, Fluet-Chouinard et al., 2023) and climate change (Ciais et al., 2013; 207 Canadell et al., 2021). An equal mass of CH4 emissions have a stronger impact on climate than carbon dioxide (CO2), which 208 is reflected by its global warming potential (GWP) relative to CO₂ on a given time horizon. For a 100-yr time horizon the 209 GWP of CH4 emitted by fossil sources is 29.8 (GWP of CH4 emitted by microbial sources is 27), whereas the values reach 210 82.5 over a 20-year horizon for CH4 emitted by fossil sources and 79.7 for CH4 emitted by microbial sources (Forster et al., 211 2021). Although global anthropogenic emissions of CH₄ are estimated at around 359 Tg CH₄ yr⁻¹ (Saunois et al., 2020), 212 representing around 2.5% of the global CO₂ anthropogenic emissions when converted to units of carbon mass flux for the 213 recent decade, the emissions-based effective radiative forcing of CH4 concentrations has contributed ~31% (1.19 W m⁻²) to 214 the additional radiative forcing from anthropogenic emissions of greenhouse gases and their precursors (3.84 W m⁻²) over 215 the industrial era (1750-2019) (Forster et al., 2021). Changes in other chemical compounds such as nitrogen oxides (NO_x) 216 or carbon monoxide (CO) also influence atmospheric CH4 through changes to its atmospheric lifetime. Emissions of CH4 217 contribute to the production of ozone, stratospheric water vapour, and CO2, and most importantly affect its own lifetime 218 (Myhre et al., 2013; Shindell et al., 2012). CH₄ has a short lifetime in the atmosphere (about 9 years for the year 2010, 219 Prather et al., 2012; Szopa et al., 2021). Hence a stabilisation or reduction of CH4 emissions leads to the stabilisation or 220 reduction of its atmospheric concentration (assuming no change in the chemical oxidants), and therefore its radiative forcing, 221 in only a few decades. While reducing CO₂ emissions is necessary to stabilise long-term warming, reducing CH₄ emissions

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is recognized as an effective option to limit climate warming in the near-term (Shindell et al., 2012; Jackson et al., 2020;
 Ocko et al., 2021; UNEP, 2021), because of its shorter lifetime compared to CO₂.

237 The momentum around the potential of CH4 to limit near-term warming has led to the launch of the Global Methane 238 Pledge at the November 2021 Conference of the Parties (COP 26). Signed by 158 countries (update on October 2024), this 239 collective effort aims at reducing global CH4 anthropogenic emissions at least 30 percent from 2020 levels by 2030 (Global 240 Methane Pledge, 2023). Given that global baseline CH₄ emissions are expected to grow through 2030 (by an additional 20-241 50 Million tons (Mt) of CH4, UNEP 2022), the CH4 emission reductions currently needed to reach the Global Methane 242 Pledge objective (UNEP, 2022) correspond to 36% of the projected baseline emissions in 2030 (ie. if no further emission 243 reductions were implemented). This implies that large reductions of CH4 emissions are needed to meet the Global Methane 244 Pledge that is consistent also with the 1.5-2°C target of the Paris Agreement (UNEP, 2022). Moreover, because CH₄ is a 245 precursor of important air pollutants such as ozone, CH4 emissions reductions are required by two international conventions: 246 the United Nations Framework Convention on Climate Change (UNFCCC) and the Convention on Long Range Transport 247 of Air Pollution (CLRTAP), making this global CH₄ budget assessment all the more critical.

248 Changes in the magnitude and temporal variation (annual to interannual) of CH4 sources and sinks over the past 249 decades are characterised by large uncertainties (e.g., Kirschke et al., 2013; Saunois et al., 2017; Turner et al., 2019). Also, 250 the decadal budget suggests relative uncertainties (hereafter reported as min-max ranges) of 20-35% for inventories of 251 anthropogenic emissions in specific sectors (e.g., agriculture, waste, fossil fuels (Tibrewal et al., 2024)), 50% for biomass 252 burning and natural wetland emissions, and up to 100% for other natural sources (e.g., inland waters, geological sources). 253 The uncertainty in the chemical loss of CH₄ by OH, the predominant sink of atmospheric CH₄, has been estimated using 254 Prather et al. (2012) and Rigby et al. (2017). The former study estimated this uncertainty at ~10% from the uncertainty in 255 the reaction rate between CH4 and OH, and the latter study was based on methyl-chloroform measurements. Bottom-up 256 approaches (chemistry transport models) estimate the uncertainty of the chemical loss by OH at around 15-20% (Saunois et 257 al., 2016, 2020). This uncertainty on the OH induced loss translates, in the top-down methods, into the minimum relative 258 uncertainty associated with global CH4 emissions, as other CH4 sinks (atomic oxygen and chlorine oxidations, soil uptake) 259 are much smaller and the atmospheric growth rate is well-defined (Dlugokencky et al., 2009). Globally, the contribution of 260 natural CH4 emissions to total emissions can be quantified by combining lifetime estimates with reconstructed pre-industrial 261 atmospheric CH₄ concentrations from ice cores (assuming natural emissions have not been perturbed during the 262 anthropocene) (e.g., Ehhalt et al., 2001). Regionally or nationally, uncertainties in emissions may reach 40-60% (e.g., for 263 South America, Africa, China, and India, see Saunois et al., 2016). Another difficulty of the CH4 budget lies in the necessity to also match the isotopic signal and in particular reflect the decreasing methane isotopic signal ¹³C (Nisbet et al., 2016; 264 265 2019). The previous budgets were tested against the isotopic observations (Saunois et al., 2017) and follow an exhaustive 266 assessment (Zhang et al., 2021b). To date only a couple of atmospheric inverse systems are able to assimilate both CH4 267 mixing ratios and stable isotopic signal to retrieve fluxes at the global scale (Thanwerdas et al., 2024; Basu et al., 2022), but

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276	these systems still need improvements in terms of configuration set-up and computing time resources, in addition to	
277	characterisation of source signatures and chemical kinetic effect (Chandra et al., 2024). We hope to be able to report isotopic	
278	constrained budgets in the coming years, or at least test the budget against the isotopic balance.	

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279 To monitor emission reductions, for example to help conduct the Paris Agreement's stocktake, sustained and long-280 term monitoring of anthropogenic emissions per sector is needed in particular for hotspots of emissions that may be missed 281 in inventories (Bergamaschi et al., 2018a; Pacala, 2010; Lauvaux et al., 2022). At the same time, reducing uncertainties in 282 all individual CH₄ sources, and thus in the overall CH₄ budget remains challenging for at least four reasons. First, CH₄ is 283 emitted by multiple processes, including natural and anthropogenic sources, point and diffuse sources, and sources 284 associated with at least three different production origins (i.e., microbial, thermogenic, and pyrogenic). These multiple 285 sources and processes require the integration of data from diverse scientific communities and across multiple temporal and 286 spatial scales. The production of accurate bottom-up estimates is complicated by the fact that anthropogenic emissions result 287 from leakage from fossil fuel production with large differences between countries depending on technologies and practices, 288 the fact that many large leak events are sporadic, and the location of many emissions hotspots is not well known, and from 289 uncertain emission factors used to summarise complex microbial processes in the agriculture and waste sectors. For the 290 latter, examples include difficulties in upscaling methane emissions from livestock without considering the variety of animal 291 weight, diet and environment, and difficulties in assessing emissions from landfills depending on waste type and waste 292 management technology. Second, atmospheric CH₄ is removed mainly by chemical reactions in the atmosphere involving 293 OH and other radicals that have very short lifetimes (typically~1s). Due to the short lifetime of OH, the spatial and temporal 294 distributions of OH are highly variable. While OH can be measured locally, calculating global CH₄ loss through OH 295 measurements requires high-resolution global OH measurements (typically half an hour to integrate cloud cover, and 1 km 296 spatially to consider OH high reactivity and heterogeneity) which is impossible from direct OH observations. As a result, 297 OH can only be calculated through large scale atmospheric chemistry modelling. Those simulated OH concentrations from 298 transport-chemistry models prescribed with emissions of precursor species affecting OH still show uncertain spatio-temporal 299 distribution from regional to global scales (Zhao et al., 2019). Third, only the net CH4 budget (sources minus sinks) is well 300 constrained by precise observations of atmospheric growth rates (Dlugokencky et al., 2009), leaving the sum of sources and 301 the sum of sinks uncertain. One distinctive feature of CH4 sources compared to CO2 fluxes is that the oceanic contribution 302 to the global CH₄ budget is small (~1-3%), making CH4 source estimation predominantly a terrestrial endeavour (USEPA, 303 2010b). Finally, we lack comprehensive observations to constrain 1) the areal extent of different types of wetlands and 304 inland freshwater (Kleinen et al., 2012, 2020, 2021, 2023; Stocker et al., 2014; Zhang et al., 2021), 2) models of wetland 305 and inland freshwater emission rates (Melton et al., 2013; Poulter et al., 2017; Wania et al., 2013; Bastviken et al., 2011; 306 Wik et al., 2016a; Rosentreter et al., 2021; Bansal et al., 2023; Lauerwald et al., 2023a; Stanley et al. 2023), 3) inventories 307 of anthropogenic emissions (Höglund-Isaksson et al., 2020; Crippa et al., 2023; USEPA, 2019), and 4) atmospheric 308 inversions, which aim to estimate CH4 emissions from global to regional scales (Houweling et al., 2017; Jacob et al., 2022).

311 The global CH4 budget inferred from atmospheric observations by atmospheric inversions relies on regional 312 constraints from atmospheric sampling networks, which are relatively dense for northern mid-latitudes, with various high-313 precision and high-accuracy surface stations, but are sparser at tropical latitudes and in the Southern Hemisphere 314 (Dlugokencky et al., 2011). Recently, the density of atmospheric observations has increased in the tropics due to satellite-315 based platforms that provide column-average CH4 mixing ratios. Despite continuous improvements in the precision and 316 accuracy of space-based measurements (e.g., Buchwitz et al., 2016), systematic errors greater than several ppb on total 317 column observations can still limit the usage of such data to constrain surface emissions (e.g., Jacob et al., 2022). The 318 development of robust bias corrections on existing data can help overcome this issue (e.g., Inoue et al., 2016, Lorente et al., 319 2023; Balasu et al., 2023) and satellite data are now widely used in atmospheric inversions where they provide more global 320 information on the distribution of fluxes and highly complement the surface networks (e.g., Lu et al., 2021).

321 In this context, the Global Carbon Project (GCP) seeks to develop a complete picture of the carbon cycle by 322 establishing common, consistent scientific knowledge to support policy development and actions to mitigate greenhouse gas 323 emissions to the atmosphere (www.globalcarbonproject.org). The objective of this paper is to analyse and synthesise the 324 current knowledge of the global CH4 budget, by gathering results of observations and models to better understand and 325 quantify the main robust features of this budget, its remaining uncertainties, and to make recommendations for improvement. 326 We combine results from a large ensemble of bottom-up approaches (e.g., process-based models for natural wetlands, data-327 driven approaches for other natural sources, inventories of anthropogenic emissions and biomass burning, and atmospheric 328 chemistry models), and top-down approaches (including CH4 atmospheric observing networks, atmospheric inversions 329 inferring emissions and sinks from the assimilation of atmospheric observations into models of atmospheric transport and 330 chemistry). The focus of this work is to update the previous assessment made for the period 2000-2017 (Saunois et al., 2020) 331 to the more recent 2000-2020 period. More in-depth analyses of trends and year-to-year changes are left to future 332 publications. Our current paper is a living review, published at about four-year intervals, to provide an update and new 333 synthesis of available observational, statistical, and model data for the overall CH4 budget and its individual components. 334 Kirschke et al. (2013) was the first CH4 budget synthesis followed by Saunois et al. (2016) and Saunois et al. 335 (2020), with companion papers by Stavert et al. (2021) on regional CH4 budgets and Jackson et al. (2020) focusing on the

(2020), with companion papers by Statert et al. (2021) on regional CH4 budgets and Jackson et al. (2020) focusing on the last year of the budget (2017). Saunois et al. (2020) covered 2000-2017 and reported CH4 emissions and sinks for three time periods: 1) the latest calendar decade at that time (2000-2009), 2) data for the latest available decade (2008-2017), and 3) the latest available year (2017) at the time. Here, the Global Methane Budget (GMB) covers 2000-2020 split into the 2000-2009 decade, the 2010-2019 decade (where data are available), the year 2020 affected by COVID induced changes in human activity, and briefly for 2021-2023 as per data availability (Section 6). The CH4 budget is presented at global, latitudinal, and regional scales and data can be downloaded from https://doi.org/10.18160/GKQ9-2RHT (Martinez et al., 2024). A global, regional and sectoral assessment of methane emission changes over the last two decades is discussed in Jackson et

343 <u>al. (2024) based on the data of Martinez et al. (2024).</u>

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346 Six sections follow this introduction. Section 2 presents the methodology used in the budget: units, definitions of 347 source categories, regions, data analysis; and discusses the delay between the period of study of the budget and the release 348 date. Section 3 presents the current knowledge about CH4 sources and sinks based on the ensemble of bottom-up approaches 349 reported here (models, inventories, data-driven approaches). Section 4 reports atmospheric observations and top-down 350 atmospheric inversions gathered for this paper. Section 5, based on Sections 3 and 4, provides the updated analysis of the 351 global CH4 budget by comparing bottom-up and top-down estimates and highlighting differences. Section 6 discusses the 352 recent changes in atmospheric CH4 in relation with changes in CH4 sources and sinks. Finally, Section 7 discusses future 353 developments, missing components, and the most critical remaining uncertainties based on our update to the global CH4 354 budget. For easier reading, the list of Contents of this manuscript is presented in the first section of the Supplementary 355 Material.

356 2 Methodology

357 2.1 Units used

358 Unless specified, fluxes are expressed in teragrams of CH₄ per vear (1 Tg CH₄ vr⁻¹ = 10^{12} g CH₄ vr⁻¹), while atmospheric 359 mixing ratios are expressed as dry air mole fractions, in parts per billion (ppb), with atmospheric CH4 annual increases, GATM, expressed in ppb yr⁻¹. In the tables, we present mean values and ranges for the two decades 2000-2009 and 2010-360 2019, together with results for the most recent available year (2020). Results obtained from previous syntheses (i.e., Saunois 361 et al., 2020 and Saunois et al., 2016) are also given for the decade 2000-2009. Following Saunois et al. (2016) and 362 363 considering that the number of studies is often relatively small for many individual source and sink estimates, uncertainties 364 are reported as minimum and maximum values of the available studies, given in brackets. In doing so, we acknowledge that 365 we do not consider the uncertainty of the individual estimates, and we express uncertainty as the range of available mean estimates, i.e., differences across measurements/methodologies considered. These minimum and maximum values are those 366 367 presented in Section 2.5 and exclude identified outliers.

368 The CH₄ emission estimates are provided with up to three significant digits, for consistency across all budget flux

369 components and to ensure the accuracy of aggregated fluxes. Nonetheless, given the values of the uncertainties in the CH₄ 370 budget, we encourage the reader to consider not more than two digits as significant for the global total budget.

371 2.2 Period of the budget and availability of data

372 The bottom-up estimates rely on global anthropogenic emission inventories, an ensemble of process-based models for

wetlands emissions, and published estimates in the literature for other natural sources. The global gridded anthropogenic

inventories (see Section (3.1.1) are updated irregularly, generally every 3 to 5 years. The last reported years of available

inventories were 2018 or 2019 when we started the top-down modelling activity. In order to cover the period 2000-2020, it

376 was necessary to extrapolate the anthropogenic inventory EDGARv6 (Crippa et al., 2021) to 2020 to use it as prior 377 information for the anthropogenic emissions in the atmospheric inversion systems as explained in the supplementary 378 material. Though EDGARv7 (EDGAR, 2022; Crippa et al., 2023) spanning until 2021 was then released, and was used for 379 the bottom-up budget. EDGARv8 (EDGAR, 2023; Crippa et al., 2023) spanning until 2022 and released in 2024, was used 380 in Section 6 to discuss the post 2020 methane budget. The land surface (wetland) models were run over the full period 2000-381 2020 using dynamical wetland areas, derived by remote sensing data or other models of flooded area variability (Sect. 3.2.1). 382 The atmospheric inversions run until mid-2021, but the last year of reported inversion results is 2020, which represents a 383 three-year lag with the present. This is due to the long time period it takes to acquire atmospheric in-situ data and integrate 384 models. Even though satellite observations are processed operationally and are generally available with a latency of days to weeks, by contrast surface observations can lag from months to years because of the time for flask analyses and data quality 385 386 checks in (mostly) non-operational chains. In addition, the final six months of inversions must be generally ignored because 387 the estimated fluxes are not constrained by as many observations as the previous periods. Lastly, this budget presents an 388 extended synthesis of the most recent development regarding inland water emissions (Sect. 3.2.2) and corrections associated 389 with double counting with wetlands.

390 2.3 Definition of regions

391 Geographically, emissions are reported globally and for three latitudinal bands (90°S-30°N, 30-60°N, 60-90°N, only for 392 gridded products). When extrapolating emission estimates forward in time (see Sect. 3.1.1), and for the regional budget 393 presented by Stavert et al. (2021), a set of 19 regions (oceans and 18 continental regions, see supplementary Fig. S3) were 394 used. As anthropogenic emissions are often reported by country, we define these regions based on a country list (Table S1). 395 This approach was compatible with all top-down and bottom-up approaches considered. The number of regions was chosen 396 to be close to the widely used TransCom inter-comparison map (Gurney et al., 2004) but with subdivisions to separate the 397 contribution from important countries or regions for the CH₄ cycle (China, South Asia, Tropical America, Tropical Africa, 398 United States of America, and Russia). The resulting region definition is the same as that used for the Global Carbon Project 399 (GCP) N₂O budget (Tian et al., 2020). Compared to Saunois et al. (2020), the Oceania region has been replaced by 400 Australasia including only Australia and New Zealand. Other territories formerly in Oceania were included in Southeast 401 Asia.

402 2.4 Definition of source and sink categories

403 CH₄ is emitted by different processes (i.e., biogenic, thermogenic, or pyrogenic) and can be of anthropogenic or natural 404 origin. Biogenic CH₄ is the final product of the decomposition of organic matter by methanogenic *Archaea* in anaerobic 405 environments, such as water-saturated soils, swamps, rice paddies, marine and freshwater sediments, landfills, sewage and 406 wastewater treatment facilities, or inside animal digestive systems. Thermogenic methane is formed on geological time Supprimé:

408 scales by the breakdown of buried organic matter due to heat and pressure deep in the Earth's crust. Thermogenic CH4 409 reaches the atmosphere through marine and land geological gas seeps. These CH₄ emissions are increased by human 410 activities, for instance, the exploitation and distribution of fossil fuels. Pyrogenic CH4 is produced by the incomplete 411 combustion of biomass and other organic materials. Peat fires, biomass burning in deforested or degraded areas, wildfires, 412 and biofuel burning are the largest sources of pyrogenic CH4, CH4 hydrates, ice-like cages of frozen CH4 found in continental 413 shelves and slopes and below sub-sea and land permafrost, can be of either biogenic or thermogenic origin. Each of these 414 three process categories has both anthropogenic and natural components. 415 In the following, we present the different CH4 sources depending on their anthropogenic or natural origin, which is relevant 416 to climate policy. Compared to the previous budgets, marginal changes have been made regarding source categories (naming 417 and grouping), to reflect the improved estimates for inland water sources and their indirect anthropogenic component. In the 418 previous Global Methane Budgets (Saunois et al., 2016, 2020), natural and anthropogenic emissions were split in a way that 419 did not correspond exactly to the definition used by the UNFCCC following the IPCC guidelines (IPCC, 2006), where, for 420 pragmatic reasons, all emissions from managed land are typically reported as anthropogenic. For instance, we considered 421 all wetlands as natural emissions, despite some wetlands being on managed land and their emissions being partly reported 422 as anthropogenic in UNFCCC national communications. Separating natural from anthropogenic sources could be quite 423 challenging, especially over regions where sources overlap, as over heavily human-dominated floodplain deltas for example. 424 The human induced perturbation of climate, atmospheric CO2, and nitrogen and sulfur deposition may also cause changes 425 in wetland sources we classified as natural. Following our previous definition, emissions from wetlands, inland freshwaters, 426 thawing permafrost, or geological leaks are accountable for "natural" emissions, even though we acknowledge that climate 427 change and other human perturbations (e.g., eutrophication) may cause changes in those emissions. CH₄ emissions from 428 reservoirs were also considered as natural even though reservoirs are human-made. Indeed, since the 2019 refinement to the 429 IPCC guidelines (IPCC, 2019) emissions from reservoirs and other flooded lands are considered to be anthropogenic by the 430 UNFCCC and should be reported as such. However, these estimates are not provided by inventories and not systematically 431 reported by all countries (especially non Annex-I countries). In this budget we rename "natural sources" to "natural and 432 indirect anthropogenic sources" to acknowledge that CH4 emissions from reservoirs, as well as from water bodies that were 433 perturbed by agricultural activities (drainage, eutrophication, land use change) are indirect anthropogenic emissions. As a 434 result, here, "natural and indirect anthropogenic sources" refer to "emissions that do not directly originate from fossil, 435 agricultural, waste, and biomass burning sources" even if they are perturbed by anthropogenic activities and climate change. 436 Natural and indirect anthropogenic emissions are split between "Wetlands and Inland Freshwaters" and "Other natural" 437 emissions (e.g., wild animals, termites, land geological sources, oceanic geological and biogenic sources, and terrestrial 438 permafrost). "Anthropogenic direct sources" are caused by direct human activities since pre-industrial/pre-agricultural time 439 (3000-2000 BC, Nakazawa et al., 1993) including agriculture, waste management, fossil fuel-related activities and biofuel 440 and biomass burning (yet we acknowledge that a small fraction of wildfires are naturally ignited). Direct anthropogenic

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emissions are split between: "Agriculture and waste emissions", "Fossil fuel emissions", and "Biomass and biofuel burning emissions", assuming that all types of fires are caused by anthropogenic activities. To conclude, this budget reports "direct anthropogenic", and "natural and indirect anthropogenic" methane emissions for the five main source categories explained above for both bottom-up and top-down approaches.

446 The sinks of methane are split into the soil uptake that can be derived from land-surface models in the bottom-up budget, 447 and the chemical sinks. The chemical sinks are estimated by either chemistry climate or chemistry transport models in the

448 bottom-up budget, and are further detailed in terms of vertical distribution (troposphere and stratosphere) and oxidants.

449 Bottom-up estimates of CH4 emissions for some processes are derived from process-oriented models (e.g., biogeochemical

450 models for wetlands, models for termites), inventory models (agriculture and waste emissions, fossil fuel emissions, biomass

451 and biofuel burning emissions), satellite-based models (large scale biomass burning), or observation-based upscaling models

452 for other sources (e.g., inland water, geological sources). From these bottom-up approaches, it is possible to provide

453 estimates for more detailed source subcategories inside each main category described above (see budget in Table 3).
454 However, the total CH₄ emission derived from the sum of independent bottom-up estimates remains unconstrained.

455 For atmospheric inversions (top-down approach), atmospheric methane concentration observations provide a constraint on the global methane total source if we assume the global sink is known (OH and other oxidant prescribed), or inversions are 456 457 optimising also for the chemical sink. OH estimates are constrained by methyl chloroform-inversion (Montzka et al., 2011; 458 Rigby et al., 2017; Patra et al., 2021). The inversions reported in this work solve for the total net CH₄ flux at the surface 459 (sum of sources minus soil uptake) (e.g., Pison et al., 2013), or a limited number of source categories (e.g., Bergamaschi et 460 al., 2013). In most of the inverse systems the atmospheric oxidant concentrations were prescribed with pre-optimized or scaled OH fields, and thus the atmospheric sink is not optimised. The assimilation of CH4 observations alone, as reported in 461 462 this synthesis, can help to separate sources with different locations or temporal variations but cannot fully separate individual 463 sources where they overlap in space and time in some regions. Top-down global and regional CH4 emissions per source 464 category were nevertheless obtained from gridded optimised fluxes, for the inversions that separated emissions into the five 465 main GCP categories. Alternatively, for the inversion that only solved for total emissions (or for other categories other than 466 the five described above), the prior contribution of each source category at the spatial resolution of the inversion was scaled 467 by the ratio of the total (or embedding category) optimised flux divided by the total (or embedding category) prior flux 468 (Kirschke et al., 2013). In other words, the prior relative mix of sources at model resolution is kept in each grid cell while 469 total emissions are given by the atmospheric inversions. The soil uptake was provided separately to report total gross surface 470 emissions instead of net fluxes (sources minus soil uptake).

471 In summary, bottom-up models and inventories emissions are presented for all relevant source processes and grouped if

472 needed into the five main categories defined above. Top-down inversion emissions are reported globally and for the five 473 main emission categories.

474 2.5 Processing of emission maps and box-plot representation of emission budgets

475 Common data analysis procedures have been applied to the different bottom-up models, inventories and atmospheric 476 inversions whenever gridded products exist. Gridded emissions from atmospheric inversions, land-surface models for 477 wetland or biomass burning were provided at the monthly scale. Emissions from anthropogenic inventories are usually 478 available as yearly estimates. These monthly or yearly fluxes were provided on a 1°x1° grid or re-gridded to 1°x1°, then 479 converted into units of Tg CH₄ per grid cell. Inversions with a resolution coarser than 1° were downscaled to 1° by each 480 modelling group. Land fluxes in coastal pixels were reallocated to the neighbouring land pixel according to our 1° land-sea 481 mask, and vice-versa for ocean fluxes. Annual and decadal means used for this study were computed from the monthly or 482 yearly gridded 1°x1° maps.

Budgets are presented as boxplots with quartiles (25%, median, 75%), outliers, and minimum and maximum values without outliers. Outliers were determined as values below the first quartile minus three times the interquartile range, or values above the third quartile plus three times the interquartile range. Mean values reported in the tables are represented as "+" symbols in the corresponding figures.

487 **3 Methane sources and sinks: bottom-up estimates**

For each source category, a short description of the relevant processes, original data sets (measurements, models) and related methodology are given. More detailed information can be found in original publication references, in Annex A2 where the sources of data used to estimate the different sources and sinks are summarised and compared with those used in Saunois et al. (2020) and in the Supplementary Material of this study when specified in the text. The emission estimates for each source category are compared with Saunois et al. (2020) in Table 3 and with Saunois et al. (2016) in Table S12 for the decade 2000-2009.

494 **3.1 Anthropogenic direct sources**

495 3.1.1 Global inventories

The main bottom-up global inventory datasets covering direct anthropogenic emissions from all sectors (Table 1) are from the United States Environmental Protection Agency (USEPA, 2019), the Greenhouse gas and Air pollutant Interactions and Synergies (GAINS) model developed by the International Institute for Applied Systems Analysis (IIASA) (Höglund-Isaksson et al., 2020) and the Emissions Database for Global Atmospheric Research (EDGARv6 and v7, Crippa et al., 2021, 2023) compiled by the European Commission Joint Research Centre (EC-JRC) and Netherlands Environmental Assessment Agency (PBL). We also used the Community Emissions Data System for historical emissions (CEDS) (Hoesly et al., 2018) developed for climate modelling and the Food and Agriculture Organization (FAO) FAOSTAT emission database (Tubiello

504 inventories are not independent as they may use the same activity data or emission factors, as discussed below. 505 These inventory datasets report emissions from fossil fuel production, transmission, and distribution; livestock enteric 506 fermentation; manure management and application; rice cultivation; solid waste and wastewater. Since the level of detail provided by country and by sector varies among inventories, the data were reconciled into common categories according to 507 508 Table S2. For example, agricultural waste-burning emissions treated as a separate category in EDGAR, GAINS and FAO, 509 are included in the biofuel sector in the USEPA inventory and in the agricultural sector in CEDS. The GAINS, EDGAR and FAO estimates of agricultural waste burning were excluded from this analysis (these amounted to 1-3 Tg CH4 yr⁻¹ in recent 510 decades) to prevent any potential overlap with separate estimates of biomass burning emissions (e.g., GFEDv4.1s; Giglio et 511 512 al. (2013); van der Werf et al (2017)). In the inventories used here, emissions for a given region/country and a given sector 513 are usually calculated following IPCC methodology (IPCC, 2006), as the product of an activity factor and its associated 514 emission factor. An abatement coefficient may also be used, to account for any regulations implemented to control emissions 515 (see e.g., Höglund-Isaksson et al., 2015). These datasets differ in their assumptions and data used for the calculation; however, they are not completely independent because they often use the same activity data and some of them follow the 516 same IPCC guidelines (IPCC, 2006). While the USEPA inventory adopts emissions reported by the countries to the 517 518 UNFCCC, other inventories (FAOSTAT, EDGAR and the GAINS model) produce their own estimates using a consistent 519 approach for all countries, typically IPCC Tier 1 methods or deriving IPCC Tier 2 emission factors from country-specific 520 information using a consistent methodology. These other inventories compile country-specific activity data and emission factor information or, if not available, adopt IPCC default factors (Tibrewal et al., 2024; Oreggioni et al., 2021; Höglund-521 522 Isaksson et al., 2020; Tubiello, 2019). CEDS takes a different approach (Hoesly et al., 2018) and combines data from 523 GAINS, EDGAR and FAO depending on the sector. Then their first estimates are scaled to match other individual or region-524 specific inventory values when available. This process maintains the spatial information in the default emission inventories 525 while preserving consistency with country level data. The FAOSTAT dataset (hereafter FAO-CH4) provides estimates at 526 the country level and is limited to agriculture (CH4 emissions from enteric fermentation, manure management, rice 527 cultivation, energy usage, burning of crop residues, and prescribed burning of savannahs) and land-use (peatland fires and 528 biomass burning). FAO-CH4 uses activity data mainly from the FAOSTAT crop and livestock production database, as 529 reported by countries to FAO (Tubiello et al., 2013), and applies mostly the Tier 1 IPCC methodology for emissions factors 530 (IPCC, 2006), which depends on geographic location and development status of the country. For manure, the country-scale 531 temperature was obtained from the FAO global agro-ecological zone database (GAEZv3.0, 2012). Although country 532 emissions are reported annually to the UNFCCC by annex I countries, and episodically by non-annex I countries, data gaps 533 of those national inventories do not allow the inclusion of these estimates in this analysis. 534 In this budget, we use the following versions of these inventories that were available at the start and during the analysis (see

et al., 2022), which covers emissions from agriculture and land use (including peatland fires and biomass fires). These

535 Table 1):

503

- EDGARv6 which provides yearly gridded emissions by sectors from 1970 to 2018 (Crippa et al., 2021; Oreggioni et al., 2021; EDGARv6 website https://edgar.jrc.ec.europa.eu/dataset_ghg60; Monforti Ferrario et al., 2021),
- EDGARv7, which provides yearly gridded emissions by sectors from 1970 to 2020 (monthly for some sectors),
 but emissions from fossil fuel energy are not separated (oil and gas, and coal are lumped together see Table S2)
 (EDGARv7 website https://edgar.jrc.ec.europa.eu/dataset ghg70; Crippa et al., 2023).
- GAINS model scenario version 4.0 (Höglund-Isaksson et al., 2020) which provides an annual sectorial gridded
 product from 1990 to 2020 both by country and gridded. USEPA (USEPA, 2019), which provides 5-year sectorial
 totals by country from 1990 to 2020 (estimates from 2015 onward are a projection), with no gridded distribution
 available. The USEPA dataset was linearly interpolated to provide yearly values from 1990-2020.
- CEDS version v_2021_04_21 which provides gridded monthly and annual country-based emissions by sectors from 1970 to 2019 (Hoesly et al., 2018; O'Rourke et al., 2021). Fossil fuel emissions for 2020 have been updated using the methodology described for CO in Zheng et al. (2023).
- FAO-CH4 (database accessed in December 2022, FAO, 2022) containing annual country level data for the period
 1961-2020, for rice, manure, and enteric fermentation; and 1990-2020 for burning savannah, crop residue and non agricultural biomass burning.

551 3.1.2 Total anthropogenic direct emissions

552 We calculated separately the total anthropogenic emissions for each inventory by adding its values for "Agriculture and waste", "Fossil fuels" and "Biofuels" with additional large-scale biomass burning emissions data (Sect. 3.1.5). This method 553 554 avoids double counting and ensures consistency within each inventory. This approach was used for the EDGARv6 and v7, 555 CEDS and GAINS inventories, but we kept the USEPA inventory as originally reported because it includes its own estimates 556 of biomass burning emissions. FAO-CH4 was only included in the range reported for the "Agriculture and waste" category. 557 For the latter, we calculated the range and mean value as the sum of the mean and range of the three anthropogenic subcategory estimates "Enteric fermentation and Manure", "Rice", and "Landfills and Waste". The values reported for the 558 559 upper-level anthropogenic categories ("Agriculture and waste", "Fossil fuels" and "Biomass burning & biofuels") are 560 therefore consistent with the sum of their subcategories, although there might be small percentage differences between the reported total anthropogenic emissions and the sum of the three upper-level categories. This approach provides a more 561 562 accurate representation of the range of emission estimates, avoiding an artificial expansion of the uncertainty attributable to 563 subtle differences in the definition of sub-sector categorisations between inventories.

Based on the ensemble of databases detailed above, total direct anthropogenic emissions were 358 [329-387] Tg CH_4 yr⁻¹

for the decade 2010-2019 (Table 3, including biomass and biofuel burning) and 331 [305-365] Tg CH₄ yr⁻¹ for the decade 2000-2009. Our estimate for the 2000-2009 decade is within the range of Saunois et al. (2020) (334 [321-358]), Saunois et

567 al. (2016) (338 Tg CH4 yr-1 [329-342]) and Kirschke et al. (2013) (331 Tg CH4 yr-1 [304-368]) for the same period. The

569 waste and fossil emissions associated with the lowest estimate of biomass burning. 570 Figure 2 (left) summarises or projects global CH₄ emissions of anthropogenic sources (including biomass and biofuel 571 burning) by different datasets between 2000 and 2050. The datasets consistently estimate total anthropogenic emissions of 572 ~300 Tg CH₄ yr⁻¹ in 2000. For the Sixth Assessment Report of the IPCC, seven main Shared Socioeconomic Pathways 573 (SSPs) were defined for future climate projections in the Coupled Model Intercomparison Project 6 (CMIP6) (Gidden et al., 574 2019: O'Neill et al., 2016) ranging from 1.9 to 8.5 W m⁻² radiative forcing by the year 2100 (as shown by the number in the SSP names). For the 1970-2015 period, historical emissions used in CMIP6 (Feng et al., 2019) combine anthropogenic 575 emissions from CEDS (Hoesly et al., 2018) and a climatological value from the GFEDv4.1s biomass burning inventory (van 576 577 Marle et al., 2017). The harmonised scenarios used for CMIP6 activities start in 2015 at 388 Tg CH₄ yr⁻¹, which corresponds 578 to the higher range of our estimates. Since CH4 emissions continue to track scenarios that assume no or minimal climate 579 policies (SSP5 and SSP3), it may indicate that climate policies, when present, have not yet produced sufficient results to 580 change the emissions trajectory substantially (Nisbet et al., 2019). After 2015, the SSPs span a range of possible outcomes, 581 but current emissions appear likely to follow the higher-emission trajectories over the past decade in terms of trend, and the 582 peak year has not yet been reached. High or medium emission reduction rates as suggested by scenarios SSP1 and SSP2 583 have not yet happened. This illustrates the challenge of methane mitigation that lies ahead to help reach the goals of the 584 Paris Agreement (Nisbet et al., 2020; Shindell et al., 2024). In addition, estimates of methane atmospheric concentrations 585 (Meinshausen et al., 2017, 2020) from the harmonised scenarios (Riahi et al., 2017) indicate that observations of global CH4 586 concentrations fall well within the range of scenarios in absolute values but their trend over the past few years is closest to 587 those of scenario SSP5-8.5 (Fig. 2 right). The CH₄ concentrations are estimated using a simple exponential decay with 588 inferred natural emissions (Meinshausen et al., 2011), and the emergence of any trend between observations and scenarios 589 needs to be confirmed in the following years. However, the current observed concentrations and emissions estimates lie in 590 the upper range of the former RCPs scenarios starting in 2005 (Fig. S1). In the future, it will be important to monitor the 591 trends from 2015 (the Paris Agreement) and from 2020 (Global Methane Pledge) estimated in inventories and from 592 atmospheric observations, and compare them to various scenarios.

slightly larger range reported herein with respect to previous estimates is due to the USEPA lower estimate for agriculture,

593 3.1.3 Fossil fuel production and use

568

Most anthropogenic CH₄ emissions related to fossil fuels come from the exploitation, transportation, and usage of coal, oil, and natural gas. Additional emissions reported in this category include small industrial contributions such as the production of chemicals and metals, fossil fuel fires (e.g., underground coal mine fires and the Kuwait oil and gas fires), and transport (road and non-road transport). CH₄ emissions from the oil processing industry (e.g., refining) and production of charcoal are estimated to be a few Tg CH₄ yr⁻¹ only, and are included in the transformation industry sector in the inventory. Fossil fuel fires are included in the subcategory "Oil & Gas". Emissions from industries, road and, non-road transport are reported

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apart from the two main subcategories "Oil & Gas" and "Coal", as in Saunois et al. (2020) and contrary to Saunois et al. (2016); each of these amounts to about 2 to 5 Tg CH₄ yr⁻¹ (Table 3). The large range (1-9 Tg CH₄ yr⁻¹) is attributable to

607 difficulties in allocating some sectors to these sub-sectors consistently among the different inventories (See Table S2). The

so, antennes in anotating some sectors to more sub-sectors consistently anong the antennes (see Factors 2). The

spatial distribution of CH4 emissions from fossil fuels is presented in Fig. 3 based on the mean gridded maps provided by

609 CEDS, EDGARv6, and GAINS for the 2010-2019 decade; USEPA lacks a gridded product.

610 Global mean emissions from fossil fuel-related activities, other industries and transport are estimated from the four global

inventories (Table 1) to be of 120 [117-125] Tg CH_4 yr⁻¹ for the 2010-2019 decade (Table 3), but with large differences in

612 the rate of change during this period across inventories. The sector accounts on average for 34% (range 31-42%) of total

613 global anthropogenic emissions in 2010-2019, This contribution has slightly increased from 32% on average in 2000-2009.

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615 Coal mining.

614

616 During mining, CH₄ is emitted primarily from ventilation shafts, where large volumes of air are pumped in and out of the 617 mine to keep the CH4 mixing ratio below 0.5% to avoid accidental ignition, and from dewatering operations. In countries of 618 the Organization for Economic Co-operation and Development (OECD), coalbed CH4 is often extracted as fuel up to ten 619 years before the coal mine starts operation, thereby reducing the CH4 channelled through ventilation shafts during mining. 620 In many countries, large quantities of ventilation air CH4 are still released to the atmosphere or flared, despite efforts to 621 extend coal mine gas recovery under the UNFCCC Clean Development Mechanisms (http://cdm.unfccc.int). CH4 leaks also 622 occur during post-mining handling, processing, and transportation. Some CH4 is released from coal waste piles and abandoned mines; while emissions from these sources were believed to be low (IPCC, 2000), recent work has estimated 623 624 these at 22 billion m3 (compared to 103 billion m3 from functioning coal mines) in 2010 with emissions projected to increase

625 into the future (Kholod et al., 2020).

In 2020, more than 35% (IEA, 2023a) of the world's electricity is still produced from coal. This contribution grew in the

2000s at the rate of several percent per year, driven by Asian economic growth where large reserves exist, but global coal

consumption declined between 2014 and 2020. In 2020, the top ten largest coal producing nations accounted for ~90% of

total world CH₄ emissions from coal mining; among them, the top three producers (China, United States of America, and

630 India) produced almost two-thirds (66%) of the world's coal (IEA, 2021).

631 Global estimates of CH₄ emissions from coal mining show a reduced range of 37-44 Tg CH₄ yr⁻¹ for 2010-2019 (Table 3),

632 compared to the previous estimate for 2008-2017 in Saunois et al. (2020) reporting a range of 29-61 Tg CH₄ yr⁻¹ for 2008-

2017. This reduced range probably results from using similar activity data (mostly from IEA statistics) in the different

inventories. The highest value of the range in Saunois et al. (2020) came from the CEDS inventory while the lowest came

from USEPA. CEDS seems to have revised downward their estimate compared to the previous version used in Saunois et

al. (2020). There were previously large discrepancies in Chinese coal emissions, with a large overestimation from

637 EDGARv4.2 on which CEDS was based. As highlighted by Liu et al. (2021a), a county-based inventory of Chinese methane

inventories globally (Fig. 2).
For the 2010-2019 decade, methane emissions from coal mining represent 33% of total fossil fuel-related emissions of CH4
(40 [37-44] Tg CH4 yr⁻¹, <u>Table 3</u>). An additional <u>assumed</u> very small source corresponds to fossil fuel fires, <u>which are mostly</u>
underground coal fires. <u>This source is estimated at around 0.15 Tg yr⁻¹ in EDGARv7, though this value remains the same</u>
across EDGAR versions and for all years despite the changes in coal production, which could influence this estimate.
However, to date, insufficient data is available to better estimate this largely unknown source.

emissions also confirms the overestimation of previous EDGAR inventories and estimated total anthropogenic Chinese

emissions at 38.2±5.5 Tg CH₄ yr¹ for 2000-2008 (Liu et al., 2021a). Coal mining emission factors depend strongly on the

type of coal extraction (underground mining emits up to 10 times more than surface mining), the geological underground

structure (region-specific), history (basin uplift), and the quality of the coal (brown coal (lignite) emits more than hard coal

(anthracite)). Finally, the different emission factors derived for coal mining is the main reason for the differences between

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651 Oil and natural gas systems.

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652 This sub-category includes emissions from both conventional and shale oil and gas exploitation. Natural gas is composed 653 primarily of CH4, so both fugitive and planned emissions during the drilling of wells in gas fields, extraction, transportation, 654 storage, gas distribution, end use, and incomplete combustion in gas flares emit CH4 (Lamb et al., 2015; Shorter et al., 1996). 655 Persistent fugitive emissions (e.g., due to leaky valves and compressors) should be distinguished from intermittent emissions 656 due to maintenance (e.g., purging and draining of pipes) or incidents. During transportation, fugitive emissions can occur in 657 oil tankers, fuel trucks and gas transmission pipelines, attributable to corrosion, manufacturing, and welding faults. According to Lelieveld et al. (2005), CH₄ fugitive emissions from gas pipelines should be relatively low, however, old 658 659 distribution networks in some cities may have higher rates, especially those with cast-iron and unprotected steel pipelines 660 (Phillips et al., 2013). Measurement campaigns in cities within the USA (e.g., McKain et al., 2015) and Europe (e.g., 661 Defratyka et al., 2021) revealed that significant emissions occur in specific locations (e.g., storage facilities, city natural gas fueling stations, well and pipeline pressurisation/depressurisation points, sewage systems, and furnaces of buildings) along 662 the distribution networks (e.g., Jackson et al., 2014a; McKain et al., 2015; Wunch et al., 2016). However, CH4 emissions 663 664 vary significantly from one city to another depending, in part, on the age of city infrastructure and the quality of its 665 maintenance, making urban emissions difficult to scale-up from measurement campaigns, although attempts have been made (e.g., Defratyka et al., 2021). In many facilities, such as gas and oil fields, refineries, and offshore platforms, most of the 666 667 associated and other waste gas generated will be flared for security reasons with almost complete conversion to CO2, 668 however, due to the large quantities of waste gas generated, small fractions of gas still being vented make up relatively large 669 quantities of methane. These two processes are usually considered together in inventories of oil and gas industries. In 670 addition, single-point failure of natural gas infrastructure can leak CH4 at high rate for months, such as at the Aliso Canyon 671 blowout in the Los Angeles, CA (Conley et al., 2016) or the shale gas well blowout in Ohio (Pandey et al., 2019), thus

677 hampering emission control strategies. Production of natural gas from the exploitation of hitherto unproductive rock

formations, especially shale, began in the 1970s in the US on an experimental or small-scale basis, and then, from the early

679 2000s, exploitation started at a large commercial scale. The shale gas contribution to total dry natural gas production in the

680 United States reached 82% in 2023, growing rapidly from 48% in 2013 (IEA, 2023b). The possibly larger emission factors

form shale gas compared to conventional gas, have been widely debated (e.g., Cathles et al., 2012; Howarth, 2019; Lewan,

682 2020). The latest studies tend to inferemission factors from the oil gas production chain of about 1% to 6% (e.g., Schneising 683 et al., 2020; Varon et al., 2023; Zhang et al., 2020), but loss rate could be has high as more than 10% in low producing well

684 <u>sites (e.g., Omara et al., 2022, Williams et al., 2024)</u>

CH₄ emissions from oil and natural gas systems vary greatly in different global inventories (67 to 80 Tg yr⁻¹ in 2020, Table 685 686 3). The inventories generally rely on the same sources and magnitudes for activity data, with the derived differences 687 therefore resulting primarily from different methodologies and parameters used, including emission factors. Those factors 688 are country- or even site-specific and the few field measurements available often combine oil and gas activities (Brandt et 689 al., 2014), resulting in high uncertainty in emission estimates for many major oil and gas producing countries. Depending 690 on the region, the IPCC 2006 default emission factors may vary by two orders of magnitude for oil production and one order 691 for gas production. For instance, the GAINSv4.0 estimate of CH₄ emissions from US oil and gas systems in 2015 is 16 692 Tg, which is almost twice as high as EDGARv8.0 (EDGAR, 2024) at 8.4 Tg and USEPA (UNFCCC, 2023) at 9.5 Tg. The 693 difference can partly be explained by GAINS using a bottom-up methodology to derive country- and year-specific flows of 694 associated petroleum gas and attributing these to recovery/reinjection, flaring or venting (Höglund-Isaksson, 2017), and 695 partly to GAINS using a higher emission factor for unconventional gas production (Höglund-Isaksson et al., 2020). Recent 696 quantifications using satellite observations and inversion estimate a relatively stable trend for US oil and gas systems emissions since 2010, with Lu et al. (2023) estimating 14.6 Tg for 2010, 15.9 Tg for 2014 and 15.6 Tg for 2019, Shen et al. 697 698 (2022) estimating a mean of 12.6 Tg for 2018-2020, and Maasakkers et al (2021) a mean of 11.1 Tg for 2010 to 2015. The 699 stable top-down trend for the US appears not well captured in the bottom-up inventories from GAINS and EDGAR, which 700 tend to show an increasing trend driven by increase in production volumes. 701 Most recent studies (e.g., Zhang et al., 2020; Shen et al., 2023; Li et al., 2024, Tibrewal et al., 2024; Sherwin et al., 2024)

702 still, suggest that the methane emissions from oil and gas industry are underestimated by inventories, industries, and agencies,

including the USEPA and UNFCCC reporting. Lauvaux et al. (2022) showed that emissions from a few high-emitting
 facilities, i.e., super-emitters (> 20 t hr⁻¹), which are usually sporadic in nature, and not accounted for in the inventories,

could represent 8-12% of global oil & gas emissions, or around 8 Tg CH_4 yr⁻¹. These high emitting points, located on the conventional part of the facilities, could be avoided through better operating conditions and repair of malfunctions. Over the last decade, absolute CH_4 emissions almost certainly increased, since USA crude oil production doubled and natural gas

production rose by about 50% (IEA, 2023a). However, global implications of the rapidly growing shale gas activity in the

709 US remain to be determined precisely.

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Supprimé: Alvarez et al., 2018; Brandt et al., 2014; Iackson et al., 2014b; Karion et al., 2013; Moore et al., 2014; Olivier and Janssens-Maenhout, 2014; Pétron et al., 2014; Zavala-Araiza et al., 2015), albeit not all (Allen et al., 2013; Cathles et al., 2012; Peischl et al., 2015

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For the 2010-2019 decade, CH₄ emissions from upstream and downstream oil and natural gas sectors are estimated to represent about 56% of total fossil CH₄ emissions (67 [57-74] Tg CH₄ yr⁻¹, Table 3) based on global inventories, with a lower uncertainty range than for coal emissions for most countries. However, it is worth noting that 8 Tg CH₄ yr⁻¹ should

726 be added on top of this estimate to acknowledge the ultra-emitters contribution, as done in Tibrewal et al (2024).

727 3.1.4 Agriculture and waste sectors

This main category includes CH₄ emissions related to livestock production (i.e., enteric fermentation in ruminant animals and manure management), rice cultivation, landfills, and wastewater handling. Of these activities, globally and in most countries, livestock is by far the largest source of CH₄, followed by waste handling and rice cultivation. Conversely, field burning of agricultural residues is a minor source of CH₄ reported in emission inventories (a few Tg at the global scale). The spatial distribution of CH₄ emissions from agriculture and waste handling is presented in Fig. 3 based on the mean gridded maps provided by CEDS, EDGARv6 and GAINS over the 2010-2019 decade.

Global emissions from agriculture and waste for the period 2010-2019 are estimated to be 211 [195-231] Tg CH₄ yr⁻¹ (Table Characteristic Characteristi

3), representing 60% of total direct anthropogenic emissions. Agriculture emissions amount to 144 Tg CH₄ yr¹, 40% of the direct anthropogenic emissions, with the rest coming from the fossil fuel sector (34%), waste (19%) and biomass (5%) and

737 biofuel (3%) burning.

Livestock: Enteric fermentation and manure management. Domestic ruminants such as cattle, buffalo, sheep, goats, and camels emit CH₄ as a by-product of the anaerobic microbial activity in their digestive systems (Johnson et al., 2002). The very stable temperatures (about 39°C) and pH (6.5-6.8) within the rumen of domestic ruminants, along with a constant plant matter flow from grazing (cattle graze many hours per day), allow methanogenic *Archaea* residing within the rumen to produce CH₄. CH₄ is released from the rumen mainly through the mouth of multi-stomached ruminants (eructation, ~90% of emissions) or absorbed in the blood system. The CH₄ produced in the intestines and partially transmitted through the rectum is only ~10% (Hill et al. 2016).

- 745 The total number of livestock continues to grow steadily. There are currently (2020) about 1.5 billion cattle globally, almost 746 1.3 billion sheep, and nearly as many goats (http://www.fao.org/faostat/en/#data/GE). Livestock numbers are linearly related 747 to CH4 emissions in inventories using the Tier 1 IPCC approach such as FAOSTAT. In practice, some non-linearity may 748 arise due to dependencies of emissions on the total weight of the animals and their diet, which are better captured by Tier 2 749 and higher approaches. Cattle, due to their large population, large individual size, and particular digestive characteristics, 750 account for the majority of enteric fermentation CH4 emissions from livestock worldwide (Tubiello, 2019; FAO, 2022), 751 particularly in intensive agricultural systems in wealthier and emerging economies, including the United States (USEPA, 752 2016). CH4 emissions from enteric fermentation also vary from one country to another as cattle may experience diverse
- rs3 living conditions that vary spatially and temporally, especially in the tropics (Chang et al., 2019).
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754 Anaerobic conditions often characterise manure decomposition in a variety of manure management systems globally (e.g., 755 liquid/slurry treated in lagoons, ponds, tanks, or pits), with the volatile solids in manure producing CH4. In contrast, when 756 manure is handled as a solid (e.g., in stacks or dry-lots) or deposited on pasture, range, or paddock lands, it tends to 757 decompose aerobically and to produce little or no CH4. However aerobic decomposition of manure tends to produce nitrous oxide (N₂O), which has a larger global warming impact than CH₄. Ambient temperature, moisture, energy contents of the 758 759 feed, manure composition, and manure storage or residency time affect the amount of CH4 produced. Despite these 760 complexities, most global datasets used herein apply a simplified IPCC Tier 1 approach, where amounts of manure treated depend on animal numbers and simplified climatic conditions by country. 761

762 Global CH₄ emissions from enteric fermentation and manure management are estimated in the range of 114-124 Tg CH₄ yr 763 ¹, for the year 2020, in the GAINS model and CEDS, USEPA, FAO-CH₄ and EDGARv7 inventories (Table 3). Using the 764 Tier 2 method adopted from the 2019 Refinement to 2006 IPCC guidelines, a recent study (Zhang et al., 2022) estimated 765 that global CH₄ emissions from livestock increased from 31.8 [26.5-37.1] (mean [minimum-maximum of 95% confidence interval) Tg CH₄ yr⁻¹ in 1890 to 131.7 [109.6-153.7] Tg CH₄ 766 yr⁻¹ in 2019, a fourfold increase in the past 130 years. Chang et al. (2021) estimates enteric fermentation and manure 767 management emissions based on mixed Tier 1&2 and Tier1 approaches and calculate livestock emissions being 120±13 and 768 769 136±15 Tg CH₄ yr⁻¹ respectively for 2018. Chang et al. (2021) and Zhang et al. (2022) estimates for 2018 or 2019 are on average a bit higher than the inventories estimates but in agreement considering the uncertainties. It is worth recalling here 770 771 that the ranges provided in this study correspond to the minimum-maximum of the existing estimates and do not include the

772 <u>uncertainty of the individual estimate; these uncertainties could be larger than the range proposed here.</u>

For the period 2010-2019, we estimated total emissions of 112 [107-118] Tg CH₄ yr⁻¹ for enteric fermentation and manure management, about one third of total global anthropogenic emissions (Table 3).

775 Rice cultivation. Most of the world's rice is grown in flooded paddy fields (Baicich, 2013). The water management systems, 776 particularly flooding, used to cultivate rice are one of the most important factors influencing CH4 emissions and one of the 777 most promising approaches for CH4 emission mitigation: periodic drainage and aeration not only cause existing soil CH4 to 778 oxidise, but also inhibit further CH4 production in soils (Simpson et al., 1995; USEPA, 2016; Zhang, 2016). Upland rice 779 fields are not typically flooded, and therefore are not a significant source of CH4. Other factors that influence CH4 emissions 780 from flooded rice fields include fertilisation practices (i.e., the use of urea and organic fertilisers), soil temperature, soil type 781 (texture and aggregated size), rice variety and cultivation practices (e.g., tillage, seeding, and weeding practices) (Conrad et al., 2000; Kai et al., 2011; USEPA, 2011; Yan et al., 2009). For instance, CH4 emissions from rice paddies increase with 782 783 organic amendments (Cai et al., 1997) but can be mitigated by applying other types of fertilisers (mineral, composts, biogas 784 residues) or using wet seeding (Wassmann et al., 2000).

785 The geographical distribution of rice emissions has been assessed by global (e.g., Janssens-Maenhout et al., 2019; Tubiello, 2019; USEPA, 2012) and regional (e.g., Castelán-Ortega et al., 2014; Chen et al., 2013; Chen and Prinn, 2006; Peng et al.,

2016; Yan et al., 2009; Zhang and Chen, 2014) inventories and land surface models (Li et al., 2005; Pathak et al., 2005; Ren et al., 2011; Spahni et al., 2011; Tian et al., 2010, 2011; Zhang, 2016). The emissions show a seasonal cycle, peaking in the summer months in the extra-tropics associated with monsoons and land management. Emissions from rice paddies are influenced not only by the extent of rice field area, but also by changes in the productivity of plants (Jiang et al., 2017) as these alter the CH₄ emission factor used in inventories. However, the inventories considered herein are largely based on IPCC Tier 1 methods, which mainly scale with cultivated areas and include regional specific emission factors but do not account for changes in plant productivity and detailed cultivation practices.

The largest emissions from rice cultivation are found in Asia accounting for 30 to 50% of global emissions (Fig. 3). The decrease of CH_4 emissions from rice cultivation over recent decades is confirmed in most inventories, because of the decrease in rice cultivation area, changes in agricultural practices, and a northward shift of rice cultivation since the 1970s, as in China (e.g., Chen et al., 2013).

Based on the global inventories considered in this study, global CH_4 emissions from rice paddies are estimated to be 32 [25-37] Tg CH_4 yr⁻¹ for the 2010-2019 decade (Table 3), or about 9% of total global anthropogenic emissions of CH_4 . These estimates are consistent with the 29 Tg CH_4 yr⁻¹ estimated for the vear 2000 by Carlson et al. (2017).

Waste management. This sector includes emissions from managed and non-managed landfills (solid waster management).

Waste management. This sector includes emissions from managed and non-managed landfills (solid waste disposal on land), and wastewater handling, where all kinds of waste are deposited. CH₄ production from waste depends on the pH,

803 moisture, and temperature of the material. The optimum pH for CH4 emission is between 6.8 and 7.4 (Thorneloe et al.,

804 2000). The development of carboxylic acids leads to low pH, which limits methane emissions. Food or organic waste, such

as leaves and grass clippings, ferment quite easily, while wood and wood products generally ferment slowly, and cellulose and lignin even more slowly (USEPA, 2010a).

Waste management was responsible for about 11% of total global direct anthropogenic CH₄ emissions in 2000 (Kirschke et al., 2013). A recent assessment of CH₄ emissions in the USA found landfills to account for almost 26% of total USA anthropogenic CH₄ emissions in 2014, the largest contribution of any single CH₄ source in the United States of America (USEPA, 2016). In Europe, gas control has been mandatory on all landfills since 2009, and more importantly for CH₄ emissions, the EU Landfill Directive (1999) with subsequent amendments, has diverted most biodegradable waste away

812 from landfills towards source separation, recycling, composting and energy recovery, and with a legally binding target not

to landfill more than 10% of municipal solid waste by 2035.

814 Wastewater from domestic and industrial sources is treated in municipal sewage treatment facilities and private effluent

treatment plants. The principal factor in determining the CH4 generation potential of wastewater is the amount of degradable

816 organic material in the wastewater. Wastewater with high organic content is treated anaerobically, which leads to increased

emissions (André et al., 2014). Excessive and rapid urban development worldwide, especially in Asia and Africa, could

enhance methane emissions from waste unless adequate mitigation policies are designed and implemented rapidly.

819 The GAINS model and CEDS and EDGAR inventories give robust emission estimates from solid waste in the range of 37-

- $42 \text{ Tg CH}_4 \text{ yr}^{-1}$ for the year 2019, and more uncertain wastewater emissions in the range 20-45 Tg CH₄ yr⁻¹1.
- 821 In our study, the global emission of CH₄ from waste management is estimated in the range of 56-80 Tg CH₄ yr⁻¹ for the
- 822 2010-2019 period with a mean value of 69 Tg CH4 yr⁻¹, about 19% of total global anthropogenic emissions (Table 3).

823 3.1.5 Biomass and biofuel burning

This category includes CH₄ emissions from biomass burning in forests, savannahs, grasslands, peats, agricultural residues, as well as, from the burning of biofuels in the residential sector (stoves, boilers, fireplaces). Biomass and biofuel burning emit CH₄ under incomplete combustion conditions (i.e., when oxygen availability is insufficient for complete combustion), for example in charcoal manufacturing and smouldering fires. The amount of CH₄ emitted during the burning of biomass depends primarily on the amount of biomass, burning conditions, fuel moisture and the specific material burned.

829 In this study, we use large-scale biomass burning (forest, savannah, grassland, and peat fires) from five biomass burning 830 inventories (described below) and the biofuel burning contribution from anthropogenic emission inventories (EDGARv6

- and v7, CEDS, GAINS and USEPA). The spatial distribution of emissions from the burning of biomass and biofuel over
 the 2010-2019 decade is presented in Fig. 3 based on data listed in Table 1.
- At the global scale, during the period of 2010-2019, biomass and biofuel burning generated CH₄ emissions of 28 [21-39] Tg
- $CH_4 \text{ yr}^{-1}$ (Table 3), of which 30-50% is from biofuel burning.
- 835

836 Biomass burning. Fire is an important disturbance event in terrestrial ecosystems globally (van der Werf et al., 2010), and 837 can be of either natural (typically ~10% of fires, ignited by lightning strikes or started accidentally) or anthropogenic origin 838 (~90%, human initiated fires) (USEPA, 2010b, chapter 9.1). As previously noted all fires are accounted as anthropogenic in 839 Table 3. Anthropogenic fires are concentrated in the tropics and subtropics, where forests, savannahs and grasslands may 840 be burned to clear land for agricultural purposes or to maintain pastures and rangelands. Small fires associated with 841 agricultural activity, such as field burning and agricultural waste burning, are often not detected by moderate resolution 842 remote sensing methods and are instead estimated based on cultivated area or through in-situ measurements such as 843 dedicated airborne campaigns (e.g., Barker et al., 2023).

Emission rates of biomass burning vary with biomass loading (depending on the biomes) at the location of the fire, the efficiency of the fire (depending on the vegetation type), the fire type (smouldering or flaming) and emission factor (mass of the considered species / mass of biomass burned). Depending on the approach, these parameters can be derived using satellite data and/or biogeochemical model, or through simpler IPCC default approaches.

848 In this study, we use five products to estimate biomass burning emissions. The Global Fire Emission Database (GFED) is

- 849 the most widely used global biomass burning emission dataset and provides estimates from 1997 onwards. Here, we use
- 850 GFEDv4.1s (van der Werf et al., 2017), based on the Carnegie-Ames-Stanford-Approach (CASA) biogeochemical model

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852 (van der Werf et al., 2010) driven by satellite derived vegetation characteristics and burned area mostly from the MODerate 853 resolution Imaging Sensor, MODIS (Giglio et al., 2013). GFEDv4.1s (with small fires) is available at a 0.25° resolution and 854 on a daily basis from 1997 to 2020. One characteristic of the GFEDv4.1s burned area is that small fires are better accounted 855 for compared to GFEDv4.1 (Randerson et al., 2012), increasing carbon emissions by approximately 35% at the global scale. The latest version GFEDv5 (Chen et al., 2023) suggest 61% higher burned area than GFEDv4.1s, in closer agreement with 856 857 burned area products from higher resolution satellite sensors. The next budget would benefit from GFEDv5 to revisit the 858 estimates of biomass burning emissions (which would likely go up) based on more specific comparison studies. The Quick Fire Emissions Dataset (QFED) is calculated using the fire radiative power (FRP) approach, in which the thermal 859 energy emitted by active fires (detected by MODIS) is converted to an estimate of CH4 flux using biome specific emissions 860

factors and a unique method of accounting for cloud cover. Further information related to this method and the derivation of
 the biome specific emission factors can be found in Darmenov and da Silva (2015). Here we use the historical QFEDv2.5

product available daily on a 0.1x0.1 grid for 2000 to 2020.

The Fire INventory from the National Center for Atmospheric Research (FINNv2.5, Wiedinmyer et al., 2023) provides daily, 1 km resolution estimates of gas and particle emissions from open burning of biomass (including wildfire, agricultural

fires and prescribed burning) over the globe for the period 2002-2020. FINNv2.5 uses MODIS and VIIRS satellite observations for active fires, land cover and vegetation density.

We use v1.3 of the Global Fire Assimilation System (GFAS, Kaiser et al., 2012), which calculates emissions of biomass

burning by assimilating Fire Radiative Power (FRP) observations from MODIS at a daily frequency and 0.5° resolution and
 is available for 2000-2020.

The FAO-CH₄ yearly biomass burning emissions are based on the most recent MODIS 6 burned area products (Prosperi et al., 2020), coupled with a pixel level (500 m) implementation of the IPCC Tier 1 approach, and are available from 1990 to 2020 (Table 1).

The differences in emission estimates for biomass burning arise from specific geographical and meteorological conditions

and fuel composition, which strongly impact combustion completeness and emission factors. The latter vary greatly

according to fire type, ranging from 2.2 g CH₄ kg⁻¹ dry matter burned for savannah and grassland fires up to 21 g CH₄ kg⁻¹

877 dry matter burned for peat fires (van der Werf et al., 2010). Biomass burning emissions encountered large interannual 878 variability related to meteorological conditions, with generally higher emissions during El-Nino periods as in 2019 (20 [14-

28] Tg CH₄ yr⁻¹), 2015 (22 [15-28] Tg CH₄ yr⁻¹) and 2010 to a lesser extent (18 [15-29] Tg CH₄ yr⁻¹).

In this study, based on the five aforementioned products, biomass burning emissions are estimated at 17 Tg CH₄ yr⁻¹ [12-

881 24] for 2010-2019, representing about 5% of total global anthropogenic CH₄ emissions (Table 3).

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883 Biofuel burning. Burning of biomass to produce energy for domestic, industrial, commercial, or transportation purposes is 884 hereafter called biofuel burning. A largely dominant fraction of CH₄ emissions from biofuel burning comes from domestic

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cooking or heating in stoves, boilers, and fireplaces, mostly in open cooking fires where wood, charcoal, agricultural 886 887 residues, or animal dung are burned. It is estimated that more than two billion people, mostly in developing countries, use 888 solid biofuels to cook and heat their homes daily (André et al., 2014), and yet CH4 emissions from biofuel combustion have 889 received relatively little attention. Biofuel burning estimates are gathered from the CEDS, USEPA, GAINS and EDGAR inventories. Due to the sectoral breakdown of the EDGAR and CEDS inventories the biofuel component of the budget has 890 891 been estimated as equivalent to the "RCO - Energy for buildings" sector as defined in Worden et al. (2017) and Hoesly et al. (2018) (Table S2). This is equivalent to the sum of the IPCC 1A4a Commercial-institutional, 1A4b_Residential, 892 893 1A4c Agriculture-forestry-fishing and 1A5 Other-unspecified reporting categories. This definition is consistent with that used in Saunois et al. (2016) and Kirschke et al. (2013). While this sector incorporates biofuel use, it also includes the use 894 of other combustible materials (e.g., coal or gas) for small-scale heat and electricity generation within residential and 895 896 commercial premises. Data provided by the GAINS inventory suggests that this approach may overestimate biofuels 897 emissions by between 5 and 50%. Further study into this category would be needed to better disentangle biofuels from fossil 898 combustibles.

In our study, biofuel burning is estimated to contribute 11 [8-14] Tg CH₄ yr⁻¹ to the global CH₄ budget, about 3% of total global anthropogenic CH₄ emissions for 2010-2019 (Table 3).

901 3.1.6 Other anthropogenic sources (not explicitly included in this study)

902 Other anthropogenic sources not included in this study are related to agriculture and land-use management. In particular, 903 increases in agricultural areas (such as global palm oil production) have led to the clearing of natural peat forests, reducing 904 natural peatland area and associated natural CH4 emissions. Peatlands planted to forests (like in Northern Europe) also lead 905 to reduced CH4 emissions. While studies have long suggested that CH4 emissions from peatland drainage ditches are likely 906 to be significant (e.g., Minkkinen and Laine, 2006, Peacock et al., 2021), CH4 emissions related to palm oil plantations have 907 vet to be properly quantified (e.g., Manning et al, 2019). Taylor et al. (2014) have quantified global palm oil wastewater treatment fluxes to be 4 ± 32 Tg CH₄ yr⁻¹ for 2010-2013. This currently represents a small and highly uncertain source of 908 909 methane but one potentially growing in the future.

910 3.2 Natural and indirect anthropogenic sources

As introduced in section 2.4, natural and indirect anthropogenic sources refer to pre-agricultural CH4 emissions even if they are perturbed by anthropogenic climate change or other global change factors (e.g., eutrophication), and indirect emissions resulting from anthropogenic perturbation of the landscape (reservoirs) and the biogeochemical characteristics of soil. They include vegetated wetland emissions and inland freshwater systems (lakes, small ponds, reservoirs, and rivers), land geological sources (gas-oil seeps, mud volcanoes, microseepage, geothermal manifestations, and volcanoes), wild animals, wildfires, termites, thawing terrestrial and marine permafrost, and coastal and oceanic sources (biogenic, geological and

917 hydrate). In water-saturated or flooded ecosystems, the decomposition of organic matter gradually depletes most of the 918 oxygen in the soil or the sediment zone, resulting in anaerobic conditions and CH₄ production. Once produced, CH₄ can 919 reach the atmosphere through a combination of three processes: (1) diffusive loss of dissolved CH₄ across the air-water 920 boundary; (2) ebullition flux from sediments; and (3) flux mediated by emergent aquatic macrophytes and terrestrial plants 921 (plant transport). On its way to the atmosphere, in the soil or water columns, CH₄ can be partly or completely oxidised by 922 microorganisms, which use CH₄ as a source of energy and carbon (USEPA, 2010b). Concurrently, methane from the 923 atmosphere can diffuse into the soil column and be oxidised (See Sect. 3.3.4 on soil uptake).

924 3.2.1 Wetlands

925 Wetlands are generally defined as ecosystems in which mineral or peat soils are water-saturated at some depth or where 926 surface inundation (permanent or not) has a dominating influence on the soil biogeochemistry and determines the ecosystem 927 species composition (USEPA, 2010b). To refine such an overly broad definition for CH4 emissions, we define wetlands as 928 ecosystems with inundated or saturated soils or peats where anaerobic conditions below the water table lead to CH4 929 production (Matthews and Fung, 1987; USEPA, 2010b). Brackish water emissions are discussed separately in Sect. 3.2.6. 930 Our definition of wetlands includes ombrotrophic and minerotrophic peatlands (i.e., bogs and fens), mineral soil wetlands 931 (swamps and marshes), and seasonal or permanent floodplains. It excludes exposed water surfaces without emergent 932 macrophytes, such as lakes, rivers, estuaries, ponds, and reservoirs (addressed in the next section), as well as rice agriculture 933 (see Sect. 3.1.4, rice cultivation paragraph), and wastewater ponds. It also excludes coastal vegetated ecosystems 934 (mangroves, seagrasses, salt marshes) with salinities usually >0.5 (See Sect. 3.2.6). Even with this definition, some wetlands 935 could be considered as anthropogenic systems, being affected by human land-use changes such as impoundments, drainage, 936 or restoration (Woodward et al., 2012). In the following, we retain the generic denomination "wetlands" for natural and 937 human-influenced wetlands, as discussed in Sect. 2.2.

The three most important factors influencing CH₄ production in wetlands are the spatial and temporal extent of anoxia
(linked to water saturation), temperature, and substrate availability (Valentine et al., 1994; Wania et al., 2010; Whalen, 2005;
Delwiche et al., 2021; Knox et al., 2021).

941 Land surface models estimate CH4 emissions through a series of processes, including CH4 production, oxidation, and 942 transport. The models are then forced with inputs accounting for changing environmental factors (Melton et al., 2013; Poulter et al., 2017; Tian et al., 2010; Wania et al., 2013; Xu et al., 2010). CH4 emissions from wetlands are computed as 943 944 the product of an emission flux density and a CH4 producing area or surface extent (see Supplementary Material; Bohn et 945 al., 2015; Melton et al., 2013). The areal extent of different wetland types (having large differences in areal CH4 emission 946 rates) appears to be a primary contributor to uncertainties in the absolute flux of CH₄ emissions from wetlands, with 947 meteorological response being the main source of uncertainty for seasonal and interannual variability (Poulter et al., 2017; 948 Kuhn et al., 2021; Parker et al., 2022; McNicol et al., 2023; Karlson and Bastviken 2023). However, large uncertainty

remains in both spatial and temporal emission distributions, especially over tropical wetlands where data are lacking to evaluate the models but are nevertheless a key region for climate feedbacks (Nisbet, 2023; Zhang et al., 2023). Direct

951 measurement campaigns and remote sensing are providing key insights where to improve the land surface models (e.g.,

952 France et al., 2022; Shaw et al., 2022).

953 In this work, sixteen land surface models computing net CH₄ emissions (Table 2) were run under a common protocol with 954 a spin-up using repeated climate data from 1901-1920 to pre-industrial conditions followed by a transient simulation through 955 the end of 2020. Of the 16 models, 13 previously contributed to Saunois et al. (2020), and three models were new to this 956 release (CH4MODwetland (Li et al., 2010), ISAM (Shu et al., 2020; Xu et al., 2021), and SDGVM (Beerling and Woodward, 957 2001; Hopcroft et al., 2011; Hopcroft et al., 2020) (Table 2, see also in the Supplementary Material Table S3 for a history 958 of the contributing models). Climatic forcing uncertainties are considered in the ensemble estimate by using two climate 959 datasets, CRU/CRU-JRA55 (Harris, 2014) and GSWP3-W5E5 (Dirmeyer et al., 2006; Kim 2017; Lange, 2019; Cucchi et 960 al., 2020). Atmospheric CO₂ was also prescribed in the models. For all models, two wetland area dynamic schemes were 961 applied: a diagnostic scheme using a remote sensing-based wetland area and dynamics dataset called WAD2M (Wetland 962 Area Dynamics for Methane Modeling; Zhang et al., 2021a; 2021b) available at 0.25 degree of horizontal resolution, as in 963 Saunois et al. (2020), and a prognostic scheme using internal model-specific hydrologic models. 964

The diagnostic wetland extent product WAD2Mv1.0 (Zhang et al., 2021a) has been updated since Saunois et al. (2020) to 965 WAD2Mv2.0 (Zhang et al., 2021b) and extended to 2020. It uses the same Surface Water Microwave Product Series 966 (SWAMPSv3.2) for capturing inundation dynamics (Jenson and McDonald, 2019), which was extended to 2020. To reduce 967 potential double-counting with the freshwater budget, the surface areas of rivers/streams and lakes/ponds are excluded by 968 using the products Global River Widths from Landsat (GRWL) database v01.01 (Allen and Pavelsky, 2018) and HydroLakes v1.0 (Messenger et al., 2016), instead of the Global Surface Water (GSW) product (Pekel et al., 2016) used in WAD2Mv1.0. 969 970 The GRWL and Hydrolakes are also the datasets used separately in the upscaling of the freshwater budget allowing for a 971 more consistent approach between the wetland and freshwater CH4 budgets (Sect. 3.2.2). This update in WAD2M leads to 972 a downward revised annual average wetland extent by 0.5 Mkm² for the mid-high latitudes (mainly due to larger lake extent 973 in HydroLakes than in the GSW dataset) with small impacts in other regions. However, since HydroLakes includes only 974 vectorized lakes larger than 0.1 km², smaller lakes/ponds under 0.1 km² are implicitly still included as wetlands in 975 WAD2Mv2.0. For the high-latitude region, the recent peatland extent product from Hugelius et al. (2020) is applied, which 976 indicates a slightly higher peatland area by 0.2 Mkm² primarily in regions above 60°N, compared to the Northern 977 Circumpolar Soil Carbon Database (NCSCD) product (Hugelius et al., 2013) used in WAD2Mv1.0. Rice agriculture was 978 removed using the Monthly Irrigated and Rainfed Crop Areas (MIRCA2000, Portmann et al. (2010)) dataset from circa 979 2000, as a fixed distribution.

The combined remote-sensing and inventory WAD2Mv2.0 product leads to a maximum wetland area of 13.6 Mkm² during the peak season (7.9 Mkm² on annual average, with a range of 7.5 to 8.4 Mkm² from 2000-2020, about 5.2% of the global Supprimé:

983 land surface). The largest wetland areas in WAD2Mv2.0 are in Amazonia, the Congo Basin, and the Western Siberian 984 Lowlands, which in previous studies were underestimated by inventories (Bohn et al., 2015). However, the SWAMPS v3.2 985 dataset which serves as a proxy of temporal variations of wetland extent, has discontinuity issues over a few tropical hotspots 986 since 2015 and hence affects the temporal variations of WAD2M. Consequently, this affects CH4 emissions estimates for a 987 subset of land surface models that are particularly sensitive to inundation in these hotspots. Meanwhile, prognostic estimates 988 show moderate consistency in capturing the spatial distribution of wetland area with WAD2M, with an annual average 989 wetland area of 8.0±2.0 Mkm² during the peak season for 2000-2020. The ensemble mean of annual wetland area anomaly 990 by the prognostic models show reasonable agreement with satellite-based estimates in capturing the response of wetland 991 area to climate variations (Zhang et al., in review), with higher agreement over temperate and boreal regions than in the 992 tropics.

993 For the wetland methane emissions estimate, we use the decadal mean from the prognostic runs and adjust these flux 994 estimates for double counting from inland waters (described in next section) given the reliance of the prognostic models on 995 satellite flooded area data like WAD2Mv2 to parameterize maximum wetland extent (Zhang et al., in review). The average 996 emission from wetlands for 2010-2019 for the 16 models is plotted in Fig. 3. The zones with the largest emissions are the 997 Amazon basin, equatorial Africa and Asia, Canada, western Siberia, eastern India, and Bangladesh. Regions where CH4 998 emissions have high inter-model agreement (defined as regions where mean flux is larger than the standard deviation of the 999 models, on a decadal mean) represent 72% of the total CH4 flux due to natural wetlands. The different sensitivities of the 1000 models to temperature, vapour pressure, precipitation, and radiation can generate substantially different patterns, such as in 1001 India. Emission estimates over regions with lower emissions (in total) are also consistently inferred between models (e.g., Scandinavia, Continental Europe, Eastern Siberia, Central United States of America, and Southern Africa). 1002

1003 The resulting global flux range for vegetated wetland emissions from the prognostic runs is 117-195 Tg CH_4 yr⁻¹ for the 1004 2000-2020 period, with an average of 157 Tg CH₄ yr¹ and a one-sigma standard deviation of 24 Tg CH₄ yr¹. Using the 1005 prognostic set of simulations, the average ensemble emissions were 159 [119-203] Tg CH₄ yr⁻¹ for the 2010-2019 period 1006 (Table 3). The estimated average ensemble annual total from the two sets of simulations by CRU/CRU-JRA55 and GSWP3-W5E5 are 158 [126-193] and 158 [118-203] for 2010-2019, respectively. Generally, the magnitude and interannual 1007 1008 variability agree between these two sets of simulations (Zhang et al., 2024). Wetland emissions represent about 25% of the 1009 total (natural plus anthropogenic) CH₄ sources estimated by bottom-up approaches. The large range in the estimates of 1010 wetland CH4 emissions results from difficulties in defining wetland CH4 producing areas as well as in parameterizing 1011 terrestrial anaerobic conditions that drive sources and the oxidative conditions leading to sinks (Melton et al., 2013; Poulter 1012 et al., 2017; Wania et al., 2013). The ensemble mean emission using the same simulation setup (i.e., diagnostic wetland 1013 extent and CRU/CRU-JRA55) in the models is 163 [117-195] Tg CH₄ yr⁻¹, higher by ~22 Tg CH₄ yr⁻¹ than the one previously 1014 reported (see Table 3, for 2000-2009 with comparison to Saunois et al., 2020). This difference is mainly due to the updated

model structure and parameterizations in the wetland CH₄ models compared to the versions in the previous budget and the
 inclusion of three new land surface models.

1017 For the last decade 2010-2019, we report in this budget an average ensemble estimate of 159 Tg CH₄ yr¹ with a range of

1018 119-203 (based on prognostic wetland extent runs, Table 3),

1019 **3.2.2 Inland freshwater <u>eco</u>systems (lakes, ponds, reservoirs, streams, rivers)**

1020 This category includes CH₄ emissions from freshwater systems (lakes, ponds, reservoirs, streams, and rivers). Numerous 1021 advances have been made in the freshwater greenhouse gases knowledge base in the last few years (Lauerwald et al., 2023a). 1022 These advances include improvements in the underlying databases used to estimate inland water surface areas and model 1023 their dynamics, a rapidly growing number of direct measurements of methane fluxes, and improvements in our process-1024 based understanding of methane biogeochemistry. Despite this, aspects of global freshwater methane estimates remain rather 1025 crude and continue to have large uncertainties. This includes the overall temperature dependency of methane emissions, the 1026 relative role of ebullition (i.e., bubble flux, which may represent the most important, but most difficult-to-capture emission 1027 path in many standing water bodies), fluxes from the smallest standing water bodies (sometimes referred to as ponds) having 1028 large emissions per m² but uncertain area extent, and the magnitude of anthropogenic influence on emissions, all which are 1029 discussed below.

1030

1031 Streams and rivers. The last global CH₄ budget used an estimate of 27 Tg CH₄ yr⁻¹ for global streams and rivers based 1032 largely on a data compilation by Stanley et al. (2016). This estimate was scaled from a simple data compilation without a 1033 spatial component or an estimate of ebullition. More recently, Rosentreter et al. (2021) performed a new data compilation 1034 of 652 flux estimates, including diffusive and ebullitive fluxes, coupled to an ice corrected surface area estimate of ~625,000 1035 km² that was aggregated to 5 latitudinal bands to come up with a global estimate of 6 and 31 ± 17 Tg CH₄ yr⁻¹ (respectively 1036 for the median and mean \pm c.i. 95%). We believe, due to better data representation in underlying datasets, that the mean 1037 estimate of Rosentreter et al. (2021) is more representative statistically because the median does not capture hotspots and 1038 hot moments of intense ebullitive fluxes. Finally, Rocher-Ros et al. (2023) used a new Global River Methane (GRiMeDB) 1039 database (Stanley et al., 2023) with > 24,000 observations of CH₄ concentrations to predict ~ 28 ± 17 Tg CH₄ yr⁻¹ (±c.i. 95%) 1040 river emissions globally. This approach used machine learning methods coupled to the latest spatially and temporally explicit 1041 mapping of monthly stream surface area (the smallest streams are still extrapolated) which incorporates drying and freezing effects (yearly average 672,000 km², Liu et al., 2022) and includes an ebullitive flux estimated from a correlation between 1042 1043 measured diffusive and ebullitive emissions in the GRiMeDB database (Stanley et al., 2023). Thus, for this study we use an 1044 estimate of 29±17 (±c.i. 95%) Tg CH₄ yr⁻¹ for streams and rivers (Figure 4), which averages the mean estimate of Rosentreter 1045 et al. (2021) and Rocher-Ros et al. (2023). Currently, ebullitive fluxes remain a major unknown quantity in streams and 1046 rivers but appear to be coarsely linearly correlated in a log-space to diffusive fluxes and of similar magnitude (Rocher-Ros Supprimé:)

1048 et al., 2023). Methodologically, the high-water velocity of many streams and rivers make measurement of ebullitive fluxes 1049 challenging (Robison et al., 2021). Effluxes are also linked to hydrology (Aho et al., 2021) although very few studies have 1050 sampled over a representative hydrograph. Plant-mediated effluxes of CH4 in running waters also remain difficult to 1051 constrain, with a recent compilation highlighting very few measurements (Bodmer et al. 2024). Connected adjacent wetlands 1052 is a common source of CH4 to streams and rivers (Borges et al., 2019) which may be important for the regulation of running 1053 water emissions but is currently difficult to assess at the global scale. Overall, the poor representation of sites and deficient mechanistic understanding make it difficult to model and predict methane evasion from streams and rivers using process-1054 1055 based models.

1056

Lakes and ponds. The previous global CH₄ budget used an estimate of 71 Tg CH₄ yr⁻¹ for lakes and 18 Tg CH₄ yr⁻¹ for 1057 1058 reservoirs. These estimates were based on an early study by Bastviken et al. (2011) coupled with a newer estimate for lakes 1059 north of 50°N (Wik et al., 2016b). There have been three new lake studies that have published their data with global estimates 1060 of 56 and 151 ± 73 (Rosentreter et al. (2021; respectively for the median and mean \pm c.i. 95%), 22 \pm 8 (Zhuang et al., 2023; 1061 ±lake-area-weighted normalised RMSE for all parameterized lake types), process-based model), and 41±36 Tg CH₄ yr⁻¹ 1062 (Johnson et al., 2022, mean ±c.i. 95%). This large range in estimated emissions can be attributed to the differences in the 1063 datasets and methods used to calculate the surface area of small waterbodies, as well as the differences between how the 1064 flux data were analyzed and extrapolated between studies. For instance, total surface areas of all lakes and ponds of 3712-1065 5688×10^3 km² (Rosentreter et al., 2021) and 2806×10^3 km² (Johnson et al., 2022) were used along with measurement data 1066 from 198 and 575 individual lake systems, respectively. In contrast, Zhuang et al. (2023) generated estimates using higher temporal resolution data from just 54 lakes to build a process-based model, which generated much lower flux estimates from 1067 1068 tropical lakes than previously implemented statistical approaches, but in line with the most recent assessments by Borges et 1069 al. (2022). For this study, we explicitly excluded lakes <0.1 km² which are treated separately (see below). If we re-assess 1070 these three studies for only lakes greater than 0.1 km², we obtain global effluxes of 17 and 42.9 \pm 20.8 Tg CH₄ yr⁻¹ 1071 (Rosentreter et al. (2021); median and mean ± c.i. 95% of global flux), 21.9±8.0. (Zhuang et al., 2023, ±lake-area-weighted 1072 normalised RMSE for all parameterized lake types), and 35.3±31.0 Tg CH4 yr⁻¹ (Johnson et al. 2022, ±95% C.I.) (with areas 1073 of 2556-3468 x10³, 2640x10³, and 2676x10³ km² respectively). Thus, for lakes >0.1 km², we propose an efflux of 33 ± 26 1074 Tg CH₄ yr⁻¹ (an average of the mean from Rosentreter et al., 2021 Zhuang et al., 2023, and Johnson et al., 2022, with the 1075 average c.i. 95% from Rosentreter et al., 2021 and Johnson et al. 2022) as represented in Figure 4. Small waterbody emissions, hereafter small lakes and ponds<0.1 km², remain difficult to assess. Evidence is emerging that 1076 1077 there is a lower limit to the power scaling laws that early studies used to extrapolate the surface area of these small systems 1078 (Bastviken et al., 2023; Kyzivat et al., 2022). Thus, for small lakes and ponds $< 0.1 \text{ km}^2$ (and $>0.001 \text{ km}^2$), we disregard the

higher end surface area used in Rosentreter et al., 2021 which relied on these earlier estimates and scale their numbers to

1080 the evasion estimates to the lower end surface area of $1,002x10^3$ to obtain a mean flux of 33 Tg CH₄ yr⁻¹ (Rosentreter et al.,

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2021). Johnson et al. (2022) estimated a surface area of only 166x103, km² for this size class to obtain an efflux of 6.3 Tg CH₄ yr⁻¹, which we acknowledge as a lower limit. Averaging these two values provide a conservative estimate of 20 [6-33] Tg CH₄ yr⁻¹, which is close to the number proposed by Holgerson and Raymond (2016) for diffusion effluxes only for this size class. The experts involved in this assessment have low confidence in this estimate. This <u>estimate</u> also does not include artificial ponds, which we discuss below. As a result, <u>combined</u> CH₄ emissions from Jarge lakes (>0.1 km²) and small lakes and ponds (<0.1km²) are estimated at 53 [19-86] Tg CH₄ yr⁻¹ (Figure 4), which is lower than the 71 Tg estimated in the previous budget.

- 1094 Reservoirs. New mean estimates of diffusive + ebullitive CH₄ emissions from reservoirs include 15 and 24±8 (the median 1095 and mean± c.i. 95% from Rosentreter et al., 2021), 10±4 (Johnson et al., 2021, mean±95% C.I.), 10 (Harrison et al., 2021, 1096 low and high c.i. 95% 7 and 22, respectively), and 2.1 Tg CH4 yr⁻¹ (Zhuang et al., 2023). We compile the first three estimates 1097 to a direct efflux of ~14 Tg CH₄ yr⁻¹ (with \pm c.i. 95% of 9 and 23). We note the fourth estimate as a lower bound, but exclude 1098 it from this budget given that it was generated via a model that only included data from six reservoir systems (Zhuang et al., 099 2023). We also add in an additional 12 Tg CH₄ yr⁻¹ (c.i. 95% 7 and 37) that is estimated to degas in dam turbines (Harrison 100 et al., 2021), which was not addressed in the studies by Rosentreter et al. (2021), Zhuang et al. (2023), or Johnson et al. 1101 (2021). Rocher-Ros et al. (2023) also excluded river observations below dams when executing their statistical model, and 1102 so did not capture downstream dam emissions. Thus, we use a direct reservoir emission here of ~13 [6-28] Tg CH4 yr⁻¹ and 1103 estimate an additional ~12 [7-37] Tg CH4 yr⁻¹ from dam turbine degassing fluxes, giving a total of 25 [13-65] Tg CH4 yr⁻¹ 1104 from reservoirs (Figure 4).
- 1105

1093

1106 Uncertainties and confidence levels. The emission estimates of lakes, reservoirs and ponds described above are limited by 1107 several uncertainties. First, a major unknown for lakes remains the size cut off and the representation of small lakes and 1108 ponds (Deemer and Holgerson, 2021), which are also more variable than larger water bodies in their CH₄ concentrations 1109 and fluxes (Rosentreter et al. 2021, Ray et al., 2023). Interestingly, there is also a lack of methane data representation from 1110 large lakes that are a large component of global lake surface area (Deemer and Holgerson, 2021; Messager et al., 2016). 1111 There is also a growing knowledge base on the importance of high CH4 fluxes from lake littoral zones that is not yet well 1112 incorporated into global scaling efforts (e.g., Grinham et al., 2011; Natchimuthu et al., 2016), and emergent vegetation 1113 (Bastviken et al., 2023; Kyzivat et al., 2022). Ebullition is more constrained in lakes/reservoirs compared to streams/rivers 1114 but is still difficult to measure and model accurately. Finally, for all inland water systems a greater scrutiny for the limiting 115 factors (including the impact of ice-cover and seasonality, stratification of the water column) of different CH₄ production, 1116 consumption and transport pathways is needed. In addition, a better understanding of the climatic, environmental and 117 geomorphological controls on key CH4_processes (e.g., sedimentary diffusive and ebullitive production, bubble dissolution, 118 CH4 oxidation) on the large-scale remains critically needed. For instance, the consistently lower global emissions determined Supprimé: ,000

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by the process-based model of Zhuang et al. (2023) compared to observations, suggest that current datasets are too limited
 to fully capture the spatio-temporal variability in CH₄ dynamics and their key control factors, possibly leading to biased high estimates.

The majority of the inland water CH₄ estimates are from a limited number of studies, some without spatial representation or reported statistical uncertainties. Furthermore, as mentioned above the knowledge base of the surface area of these ecosystems is new and rapidly expanding, but not standardised between studies leading to uncertainty (but see Lauerwald et al. 2023b), particularly for ponds. For this study, we are able to provide confidence intervals from the original studies for all fluxes except the smallest lake/pond size class.

1139

1140 The Surface Area of Inland Freshwaters. For all of these ecosystems, determining their surface area remains a central 1141 challenge. Since the last GMB, several methodological advances have reduced the uncertainty associated with the surface 1142 area estimates of rivers, streams, lakes, and reservoirs. Using a single geospatial dataset that includes both lakes and 1143 reservoirs (Messager et al., 2016) has decreased double counting of lakes and reservoirs (Johnson et al., 2022; Rosentreter 1144 et al., 2021). For rivers and streams, high-resolution global streamflow simulations, informed by satellite observations, 1145 enabled a much finer scale estimate of surface areas for rivers with a new temporal component (Allen and Pavelsky, 2018; 1146 Lin et al., 2019; Liu et al., 2022), although the surface for the smaller streams are still estimated indirectly, and mapping of 1147 human-created drainage ditches and canals is lacking. Seasonal ice cover and melt turnover corrections also have been newly 1148 incorporated into rivers, streams, lakes, and reservoirs (Harrison et al., 2021; Johnson et al., 2022; Lauerwald et al., 2023b; 1149 Rocher-Ros et al., 2023; Rosentreter et al., 2021; Zhuang et al., 2023). Finally, removing open water body surface areas 1150 from wetland surface areas based on geographic location has reduced double counting between these two land cover types, 1151 as described in the wetlands section of the GMB. Yet, the surface area of small lakes and ponds (<0.1 km2) is still highly 1152 uncertain, and new techniques for counting these systems and determining the overlap with wetland data bases is paramount. 1153 1154 Anthropogenic Contributions to Inland Freshwater Emissions. We argue that all reservoirs should be categorised as a 1155 direct anthropogenic source of emissions. Most of the surface area of reservoirs are human-made and reservoir construction 1156 leads to anoxic sediments and/or bottom waters with labile organic matter sourced from the watershed and to in-situ nutrient

augmented phytoplankton production (Deemer et al., 2016; Maavara et al., 2017; Prairie et al., 2018). It is also clear that the cultural eutrophication of natural lakes <u>driven by run-off of agricultural nitrogen fertilizer and manure</u> is augmenting CH4 emissions (DelSontro et al., 2018; Li et al., 2021), with shallow lakes particularly likely to experience eutrophication (Qin et al., 2020). For instance, Beaulieu et al. (2019) modelled a 15% reduction in lake CH4 with a 25% reduction in lake phosphorus concentrations. Several recent studies have estimated that anywhere between 30 and 50% of lakes are eutrophic (Cael et al., 2022; Qin et al., 2020; Sayers et al., 2015; Wu et al., 2022). These studies estimate numerical percentages (one by depth class: Qin et al., 2020), but none have estimated the percent of lake surface area that is eutrophic nor have any

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1165 determined the extent of anthropogenic vs. natural eutrophication. Still, numerous studies have noted widespread increases 1166 in eutrophication indicators across lakes due to nutrient loading and warming (Griffiths et al., 2022; Ho et al., 2019; Taranu 1167 et al., 2015), thus we estimate that ¹/₃, or 11 Tg CH₄ yr¹ of CH₄ emissions from lakes >0.1 km² could be anthropogenic (Figure 4). Similarly, CH4 emissions from small lakes and ponds are influenced by human factors, with emissions increasing 1168 1169 with eutrophication (Deemer and Holgerson, 2021), erosion and runoff in agricultural landscapes (Heathcote et al., 2013), 1170 and warming, the latter likely to have a disproportionately greater effect in small, shallow systems (Woolway et al., 2016). 1171 Thus, we adopt the same 1/3 number as for lakes for the proportion of anthropogenic emissions in small lakes and ponds 1172 (<0.1 km2), which amounts to 6 Tg CH₄ yr⁻¹ (Figure 4).

1173 There are also human-made small lakes and ponds, notably for agriculture, aquaculture, and recreation, that generally have conditions favourable for high CH4 emissions (Downing, 2010; Holgerson and Raymond, 2016; Malerba et al., 2022; 1174 1175 Ollivier et al., 2019; Zhao et al., 2021; Dong et al., 2023). Downing (2010) estimated that farm ponds comprise a global 1176 surface area of \sim 77,000 km²; using a conservative emission rate of 265 mg CH₄ m⁻² d⁻¹ and an ice correction factor of 0.6 1177 leads to an emission of 4.5 Tg yr⁻¹ that is anthropogenically sourced from farm ponds. Here the value is rounded to 5 Tg yr⁻¹ 1178 ¹ (Figure 4). Clearly, more work is required to assess the anthropogenic component of CH₄ emissions from lakes and ponds. 1179 It remains difficult to parse out an anthropogenic component to stream and river CH4 fluxes. Although some studies have 1180 noticed a temperature dependence with stream sediments (Comer-Warner et al., 2018; Zhu et al., 2020), Rocher-Ros et al. 1181 (2023) noted a small temperature dependence of CH₄ emissions in streams and rivers compared to other freshwater 1182 ecosystems, potentially due to the many other external processes affecting fluxes in these dynamic flowing ecosystems. 1183 Urbanisation can lead to elevated river CH₄ emissions, particularly in regions with elevated organic matter and nutrient loading due to limited wastewater treatment (Begum et al., 2021; Nirmal Raikumar et al., 2008; Wang et al., 2021a). Some 1184 1185 studies have found agricultural streams and ditches can have higher effluxes due to inputs of fine sediments (Comer-Warner 1186 et al., 2018; Crawford and Stanley, 2016), organic carbon, and nutrients (Borges et al., 2018) that lead to in-situ methane 1187 production. Furthermore, the creation of drainage ditches in organic soils tap CH4 rich waters from water-logged horizons 1188 and heighten emissions from ex-situ sources (Peacock et al., 2021), although limitations in both the geographic scope of 1189 existing ditch emission estimates our ability to estimate global surface area of ditches precludes their inclusion in this budget. 1190 Finally, extremely high rates of CH4 emission have been linked to ongoing permafrost thaw in Asia's Qinghai-Tibet Plateau 1191 (Zhang et al., 2020). However, the loss and disconnection of wetlands to rivers may have resulted in a decrease in the input 1192 of dissolved CH4 from this source. A recent expert elicitation (Rosentreter, et al. 2024) reported that 35% of all inland 1193 freshwater sources were anthropogenic and given that some of the river flux is from upstream reservoirs, we assign a 30% 1194 anthropogenic contribution to the stream and river flux (9 Tg CH_4 yr⁻¹, Figure 4), which approximates the expert elicitation 1195 via the impact of eutrophication and urban influences. 1196

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1198	Combination (lakes, ponds, reservoirs, streams and rivers, farm ponds). Combining the aforementioned emissions from	
1199	lakes >0.1 km ² (33 [13-53] Tg CH ₄ yr ⁻¹), small lakes and ponds < 0.1 km ² (20 [6-33] Tg CH ₄ yr ⁻¹), reservoirs (25 [13-65] model of the second se	Supprimé: ~
1200	Tg CH ₄ yr ⁻¹), streams and rivers (29 [12-46] Tg CH ₄ yr ⁻¹) and farm ponds (5 Tg CH ₄ yr ⁻¹), leads to a total of \sim 112 Tg CH ₄	
1201	yr ⁻¹ from freshwater systems, with a range of [49-202] Tg CH ₄ yr ⁻¹ . This estimate is about 50 Tg lower than in Saunois et	
1202	al. (2020) and is broadly consistent with the recent regionalized estimate by Lauerwald et al. (2023b) compiled for the	
1203	Regional Carbon Cycle Assessment and Processes (RECCAP2, https://www.globalcarbonproject.org/reccap/; 103 Tg CH4	
1204	yr ¹ , IQR= 82.1–134.8). The updated budget from these ecosystems and their anthropogenic components are represented in	Supprimé: 0
1205	Fig. 4. The gridded products for emissions from lakes and ponds by Johnson et al. (2022), from reservoirs by Johnson et al.	
1206	(2021) and from streams and rivers by Rocher-Ros et al. (2023) have been combined into a single map presented in Fig. 5.	
1207		
1208	Double-counting inland freshwater, ecosystems in the bottom-up estimates. To address the differences found between	Supprimé: aquatic
1209	bottom-up and top-down CH_4 budgets, and to acknowledge advances in addressing the central issue of double counting CH_4 $$	
1210	emissions for inland freshwater ecosystems, we introduce here a new correction term. Historically, the bottom-up estimate	
1211	of global CH_4 emissions has been higher than the top-down estimate, first recognized in Kirschke et al. (2013) and confirmed	
1212	in Saunois et al. (2016, 2020). The larger bottom-up emissions estimate has been partly attributed to double-counting	
1213	vegetated wetland emissions with inland freshwater emissions (including lakes, ponds, rivers, streams, and reservoirs) and	
1214	also the emissions of CH4 produced in vegetated wetlands and then transported via aquatic processes and emitted from	
1215	inland freshwaters (Pangala et al., 2017; Kirk and Cohen, 2023). The Saunois et al. (2020) CH4 budget addressed the issue	
1216	of double counting through the use of a revised vegetated wetland area dataset, WAD2M v1.0 (Zhang et al., 2021), that	
1217	removed inland waters from the SWAMPS (Jenson and McDonald, 2019) surface-inundation dataset, allowing for	
1218	independent vegetated wetlands and inland freshwater CH4 emissions to be compiled. Yet, the Saunois et al. (2020) CH4	
1219	budget still had a ~ 150 Tg CH ₄ yr ⁻¹ difference between bottom-up and top-down estimates. In this budget, we refined the	
1220	vegetated wetland area dataset with WAD2M v2.0 (see section 3.2.1, where HydroLakes is used to remove lakes and ponds	
1221	>0.1 km ²). Additionally, we applied numbers from peer-reviewed publications and expert elicitation to account for lateral	
1222	CH4 flux emissions. This most recent bottom-up budget estimates 159 [119-203] Tg CH4 yr ⁻¹ from vegetated wetlands for	Supprimé: BU
1223	$2010\text{-}2019 \text{ and } 112 \text{ Tg } \text{CH}_4 \text{ yr}^1 \text{ from inland freshwaters that includes 83 Tg } \text{CH}_4 \text{ yr}^1 \text{ from lakes, ponds, and reservoirs and}$	
1224	29 Tg CH ₄ yr ⁻¹ from rivers and streams, leading to a combined wetland and inland freshwater flux of 271 Tg CH ₄ yr ⁻¹ . Here,	
1225	we propose a correction of 20 Tg CH ₄ yr ⁻¹ to account for double counting of small lakes and ponds (< 0.1 km^2) that are	
1226	likely included in our vegetated wetlands estimate, and removing 1-3 Tg CH_4 yr ⁻¹ from river emissions due to lateral	
1227	transport of CH4 originating in adjacent vegetated wetlands. The river flux correction arises from assuming that for	
1228	catchments with >10% wetlands, rivers provide 5-10% of vegetated CH4 emissions. The total double-counting correction	
1229	term of 23 Tg CH4 reduces the bottom-up budget for combined wetlands and inland waters from 271 Tg CH4 yr ⁻¹ to 248 Tg	Supprimé: BU
1230	$CH_4\ yr^{\text{-}1}$ (see Fig. 4 and Table 3). Comparing the 2000-2009 decadal emissions from wetlands and inland freshwater	

ecosystems across the last three previous assessments of the budget shows a significant downward revision with 305 (183+122) Tg CH₄ yr¹, 356 (147+209) Tg CH₄ yr¹ and 248 (159+112-23) Tg CH₄ yr¹ (respectively from Saunois et al. (2016; 2020) and this work).

1239 Finally, it is worth noting that inland freshwater ecosystems can overlap with geological seepage systems in some areas,

1240 i.e., they may occur in correspondence with geological structures that emit fossil (microbial, thermogenic, or abiotic)

1241 CH4 generated in the Earth's crust. Examples have been documented in the Fisherman Lake in Canada (Smith et al., 2005), 1242 in the Baikal lake (Schmid et al, 2007), and in rice paddies in Japan (Etiope et al., 2011). Thus, some gas emissions in 1243 freshwater environments, particularly as bubble plumes, can be incorrectly attributed to modern biological (ecosystem)

1244 activities if appropriate isotopic and molecular analyses are not performed.

1245 3.2.3 Onshore and offshore geological sources

Significant amounts of CH4, produced within the Earth's crust, naturally migrate to the atmosphere through tectonic faults 1246 1247 and fractured rocks. Major emissions are related to hydrocarbon formation in sedimentary basins (microbial and thermogenic 1248 methane), through continuous or episodic exhalations from onshore and shallow marine hydrocarbon seeps and through 1249 diffuse soil microseepage (Etiope, 2015). Specifically, five source categories have been considered. Four are onshore 1250 sources: gas-oil seeps, mud volcanoes, diffuse microseepage, and geothermal manifestations including volcanoes. One 1251 source is offshore: submarine seepage, which may include the same types of gas manifestations occurring on land. Etiope et al. (2019) have produced the first gridded maps of geological CH4 emissions and their isotopic signature for these five 1252 categories, with a global total of 37.4 Tg CH4 yr⁻¹ (reproduced in Fig. 5). However, these maps are based on incomplete 1253 1254 data on geological sites due to missing information and difficulties in defining all current geological emitting sites. 1255 Combining the best estimates for the five categories of geological sources (from grid maps or from previous statistical and process-based models), the breakdown by category reveals that onshore microseepage dominate (24 Tg CH4 yr⁻¹), the other 1256 1257 categories having similar smaller contributions: as mean values, 4.7 Tg CH₄ yr⁻¹ for geothermal manifestations, about 7 Tg 1258 CH₄ yr⁻¹ for submarine seepage and 9.6 Tg CH₄ yr⁻¹ for onshore seeps and mud volcanoes. These values lead to a global 1259 bottom-up geological emission mean of 45 [27-63] Tg CH4 yr⁻¹ (Etiope and Schwietzke, 2019).

1260 While all bottom-up and some top-down estimates, following different and independent techniques from different authors, consistently suggest a global geo-CH4 emission in the order of 40-50 Tg yr-1, the radiocarbon (14C-CH4) data in ice cores 1261 1262 reported by Hmiel et al. (2020) appear to give a much lower estimate, with a minimum of about 1.6 Tg CH4 yr⁻¹ and a 1263 maximum value of 5.4 Tg CH₄ yr⁻¹ (95 percent confidence) for the pre-industrial period. Dyonisius et al. (2020) also suggest 1264 a low range of geological emissions over the last deglaciation period and for the late Holocene (0-10 Tg CH₄ yr⁻¹). The 1265 discrepancy between Hmiel et al. (2020) and all other estimates has been discussed in Thornton et al. (2021), which 1266 demonstrated that the global near-zero geologic CH4 emission estimate in Hmiel et al. (2020) is incompatible with the sum 1267 of multiple independent bottom-up estimates, based on a wide variety of methodologies, from individual natural geological

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1269 seepage areas: for example, from the Black Sea (up to 1 Tg CH4 yr⁻¹), the Eastern Siberian Arctic Shelf (ESAS, up to 4.6 1270 Tg CH₄ yr¹, referring mostly to thermogenic gas), onshore Alaska (up to 1.4 Tg CH₄ yr¹) and a single seepage site in 1271 Indonesia (releasing 0.1 Tg CH₄ yr⁻¹ as estimated by satellite measurement) (see Thornton et al. (2021) and references 1272 therein). Jackson et al. (2020) expressed doubt about the low Hmiel et al. (2020) estimates, noting that they are difficult to 1273 reconcile with the results of many other researchers and with bottom-up approaches in general. This discrepancy highlights 274 another main unresolved uncertainty in the methane budget and calls for further investigations to reconcile the different 275 estimates, and reduce the uncertainty on geological emissions. Waiting for further investigation to better understand 1276 discrepancies between radiocarbon approaches and other studies, we decided to keep the estimates from Etiope and 1277 Schwietzke (2019) for the mean values, and associate it to the lowest estimates reported in Etiope et al. (2019), as in Saunois 1278 et al. (2020). Thus, we report a total global geological emission of 45 [18-63] Tg CH₄ yr⁻¹, with a breakdown between 1279 offshore emissions of 7 [5-10] Tg CH4 yr⁻¹ and onshore emissions of 38 [13-53] Tg CH4 yr⁻¹ (Table 3), similar to Saunois et 1280 al. (2020). This bottom-up estimate is slightly lower than in the Saunois et al. (2016) budget mostly due to a reduction of

1281 estimated emissions of onshore and offshore seeps (see Sect. 3.2.6 for more offshore contribution explanations).

1282 **3.2.4 Termites**

1283 Termites are decomposers playing a central role in ecosystem nutrient fluxes at tropical and subtropical latitudes, in 1284 particular (Abe et al., 2000). Termites represent a natural CH4 source due to methanogenesis occurring in their hindgut 1285 during the symbiotic metabolic breakdown of lignocellulose (Sanderson, 1996; Brune, 2014). The upscaling of CH₄ emissions from termites from site to global level is characterised by high uncertainty (Sanderson, 1996; Kirschke et al., 1286 1287 2013; Saunois et al., 2016) due to the combination of factors that need to be considered and the scarcity of information for 1288 each of these factors for global upscaling. Needed data include termite biomass density (Sanderson, 1996), species distribution within and among ecosystems (Sugimoto et al., 1998), variation of termite CH4 emission rates per species and 1289 1290 dietary group (Sanderson, 1996), the role played by the termite mound structure in affecting the fraction of produced CH₄ 1291 that is effectively released into the atmosphere (Sugimoto et al., 1998; Nauer et al., 2018). In Kirschke et al. (2013) and 1292 Saunois et al. (2016) a global upscaling of termite CH4 emissions was proposed, where CH4 emissions, ECH4 (kg CH4 ha⁻¹yr 1293 ¹), were estimated as the product of three terms: termite biomass (Bioterm g fresh weight m²), a scalar correction factor 1294 (LU) expressing the effect of land use/cover change on termite biomass density, a termite CH4 emission factor (EFTERM, µg 1295 CH4 g⁻¹ Bio_{TERM} h⁻¹). The approach between the two re-analyses of CH4 emissions varied only for the data sources of gross primary productivity (GPP) and land use which were used to attribute biomass values of termite per ecosystem surface unit, 1296 1297 in order to cover different time spans, 1980s, 1990s and 2000s in Kirschke et al. (2013) and 2000-2007 and 2010-2016 in Saunois et al. (2016). For the present update, additional changes have been introduced compared with the previous versions. 1298 1299 Here we summarise the key data used for the new upscaling. CH4 fluxes were modelled between 45°S and 45°N and within 1300 35°S and 35°N. The termite biomass density, BioTERM, for tropical ecosystems was estimated as function (Kirschke et al., Supprimé:

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2013; BIOTERM=1.21 e 0.0008 GPP) of the gross primary production (GPP, g C m⁻² yr⁻¹) using the 0.25° native resolution 1303 1304 VODCA2GPP dataset covering the period 2001-2020 (Wild et al., 2022). Wetlands, barren areas, water bodies and artificial 1305 surfaces were excluded from this estimation and set as no data (no emissions). The scalar correction factor LU of 0.4 (60%) for agricultural areas (i.e., croplands) (Kirschke et al., 2013) was applied to the GPP value of the nearest natural areas to 1306 1307 account for anthropic disturbance. The annual (2001-2020) land cover information was obtained from the MODIS Terra+Aqua Combined Land Cover product (MCD12C1v006; https://lpdaac.usgs.gov/products/mcd12c1v006/), using the 1308 1309 International Geosphere-Biosphere Programme (IGP) classification with a 0.05° spatial resolution. For desert and arid lands, 1310 within 35°S and 35°N, a fixed BiotERM value of 1.56 g m⁻² was instead used (Sanderson, 1996; Heděnec et al., 2022). Similarly, fix values from the few available studies reported in literature were used to estimate BioTERM between 35°- 45° N 1311 1312 and 35°- 45° S as follows: 1.83 g m⁻² for temperate forests and grasslands (Wood and Sands, 1978; Petersen and Luxton, 1982; Sanderson, 1996; Bignell and Eggleton, 2000; King et al., 2013; conversion factor from dry to fresh biomass is 0.27 1313 from Petersen and Luxton, 1982), 5.3 g m⁻² for scrublands and Mediterranean areas of Australia (Sanderson, 1996), 1.09 g 1314 m⁻² for the other Mediterranean shrubland ecosystems (Heděnec et al., 2022). Other climates and land covers were set as no 1315 1316 data. Climate zoning was defined using the Climate Zones Köppen-Geiger dataset (Beck et al., 2018), this product is 1317 representative for the 1980-2016 time period and has a 0.0083° native resolution. The EFTERM was revised compared with 1318 previous estimates (Kirschke et al., 2013; Saunois et al., 2016), in order to consider the different distribution of termite 1319 families and subfamilies in the different continents and ecosystems, characterised by different feeding habits and nest 1320 typologies, as reported by Sugimoto et al. (1998), which might influence the EF. The species of each family and subfamily 1321 of the two major groups of lower and higher termites, listed by Sugimoto et al. (1998) were associated with EF values based 1322 on emissions from in-vitro experiments as reported by Sanderson (1996) and Eggleton et al. (1999), to which a correction 1323 factor (cf_{MOUND}) of 0.5 (Nauer et al., 2018; Chiri et al., 2020; 2021) was applied in order to take into account the mound 1324 effect on the CH₄ produced by termites, once inside the nest. The average EF_{TERM} for tropical and temperate areas was hence 1325 estimated as the weighted EFTERM derived from the product of the percentage weight of each family or subfamily of termites 1326 in the "community composition" in each geographical area and ecosystem (Sugimoto et al. (1998, Table 6), the respective 1327 calculated EF of each family or subfamily, a scalar or correction factor which considers the nest type (as in Table 5 from 1328 Sugimoto et al. 1998). For desert and arid lands and temperate areas, which were not reported in Sugimoto et al. (1998), EF 1329 rates were calculated directly from data reported in literature for the most representative species which were the genus 1330 Amitermes for the former (EF from data by Sanderson 1996, Eggleton et al. 1999, Jamali et al. 2011) and the genus 1331 Reticulitermes (family Rhinotermitidea) for the latter (EF from data by Odelson and Breznak, 1983; Sanderson, 1996; Eggleton et al., 1999; Myer et al., 2021). The following EF_{TERMS} were hence obtained to scale up emissions: $3.26 \pm 1.79 \ \mu g$ 1332 CH₄ g⁻¹ termite h⁻¹ (28.56 mg CH₄ g⁻¹ termite year⁻¹) for tropical ecosystems, $1.82 \pm 1.54 \mu g$ CH₄ g⁻¹ termite h⁻¹ for temperate 1333 1334 forests, grasslands, and Mediterranean areas, $1.24 \pm 1.22 \ \mu g \ CH_4 \ g^{-1}$ termite h⁻¹ for deserts and arid lands (warm climate). 1335 Annual CH4 fluxes were computed for all the years from 2001 to 2020 producing 20 global maps at 0.05° resolution of

1336 yearly total emissions. A further map of the estimated error representative of the entire time period was elaborated at the1337 same resolution as the emissions dataset.

1338Termite CH4 emissions over the period 2001-2020 varied between 9.7-10.8 Tg CH4 yr⁻¹, with an average of 10.2 ± 6.2 Tg1339CH4 yr⁻¹. Considering a 20-year average, tropical and subtropical moist broadleaf forests contributed to 46% of the total1340average flux, while tropical and subtropical grasslands, savannas, and shrublands to another 36%. In terms of regional1341contribution, 37.2% of fluxes were attributed to South America, 31.5% to Africa, 18.1% to Asia, 5.5% to Australia, 7.4%1342to North America and less than 1% to Europe. The present estimate value is within the range of previous up-scaling studies,1343spanning from 2 to 22 Tg CH4 yr⁻¹ (Ciais et al., 2013). In this study, we report a decadal value of 10 Tg CH4 yr⁻¹ with a1344range of [4-16] (Table 3).

1345 **3.2.5 Wild animals**

1346 Wild ruminants emit CH4 through microbial fermentation that occurs in their rumen, similarly to domesticated livestock species (USEPA, 2010b). Using a total animal population of 100-500 million, Crutzen et al. (1986) estimated the global 1347 emissions of CH4 from wild ruminants to be in the range of 2-6 Tg CH4 yr⁻¹. More recently, Pérez-Barbería (2017) lowered 1348 1349 this estimate to 1.1-2.7 Tg CH₄ yr⁻¹ using a total animal population estimate of 214 million (range of 210-219), arguing that 1350 the maximum number of animals (500 million) used in Crutzen et al. (1986) was poorly justified. Moreover Pérez-Barbería 1351 (2017) also stated that the value of 15 Tg CH₄ yr¹ found in the last IPCC reports is much higher than their estimate because 1352 this value comes from an extrapolation of Crutzen's work for the last glacial maximum when the population of wild animals 1353 was much larger, as originally proposed by Chappellaz et al. (1993). Recently, based on the modelling of grassland extent, Kleinen et al. (2023) also suggest that the population of wild animal during the last glacial maximum proposed by Crutzen 1354 1355 et al. (1986) and further used by Chappellaz et al. (1993) were overestimated. However, the estimate of 1-3 Tg CH₄ yr⁻¹ 356 seems underestimated when considering that Hempson et al. (2017) found actual CH4 emissions from African wildlife alone 357 to be around 9 Tg CH₄ yr⁻¹ but without discussing the uncertainty of this value. As a result, high uncertainty remains and 358 recalls the need for further investigation of this natural source of CH4,

Based on these findings and waiting for further global estimates, the range adopted in this updated CH₄ budget is 2 [1-3] Tg CH₄ yr⁻¹ (Table 3).

361 3.2.6 Coastal and oceanic sources

Coastal and oceanic sources comprise CH₄ release from estuaries, coastal vegetated habitats, as well as marine waters including seas and oceans. Possible sources of coastal and oceanic CH₄ include (1) in-situ biogenic production through various pathways in oxygenated sea-surface waters (Oremland, 1979; Karl et al., 2008; Lenhart et al., 2016; Repeta et al., 2016), a flux that can be enhanced in the coastal ocean because of submarine groundwater discharge (USEPA, 2010b); (2) production from shallow and marine (bare and vegetated) sediments including free gas or destabilised hydrates and thawing

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subsea permafrost containing modern (¹⁴C-bearing) microbial gas; (3) geological marine seepage (see also Sect. 3.2.3),
including hydrates, containing fossil (¹⁴C-free) microbial or thermogenic CH₄. CH₄ produced in marine sediments and
seabed CH₄ seepage can be transported across the water column to the sea-surface by upwelling waters (once at the surface
methane can cross the sea-air interface via diffusion) and gas bubble plumes (for instance from geological marine seeps;
e.g., Judd, 2004; Etiope et al., 2019). Gas bubble plumes generally reach the atmosphere in relatively shallow waters (<400
m) of continental shelves depending on the intensity of the events (e.g., Westbrook et al., 2009); however massive deepwater seepage events could contribute a significant amount of CH4 to the atmosphere, even from depths > 1000 m (e.g.,

Schmale et al., 2005.; Greinert et al., 2006; Solomon et al., 2009), In coastal vegetated habitats, CH4 can also be transported

to the atmosphere through the aerenchyma of emergent aquatic plants (Purvaja et al., 2004).

We distinguish between coastal and oceanic "geological" and "modern biogenic" CH₄ sources. Coastal and oceanic 1376 1377 "geological" emissions refer to CH4 seepage from the Earth's crust (mostly in hydrocarbon-rich sedimentary basins), which 1378 is typically evaluated by combining geochemical analyses (isotopic and molecular, including radiocarbon, ¹⁴C, analyses) 1379 and geological observations (degassing along faults, seeps, mud volcanoes). Geological emissions do not contain modern 1380 biogenic gas that is fossil (14C-free). Coastal and oceanic "biogenic" CH4 refers to CH4 formed in situ in coastal and marine 1381 sediments and in the water column by recent or modern microbial activity (therefore with measurable amounts of 1382 radiocarbon (14C)). To avoid double-counting, we assume that all diffusive CH4 emissions outside of geological seepage 1383 regions (identified in global grid maps; Etiope et al., 2019) are fuelled by biogenic CH4. Finally, we briefly discuss the case 1384 of CH4 hydrates, which can be considered either a "geological" source when they host fossil CH4 or a "biogenic" source 1385 when they host modern CH4.

Coastal and oceanic modern biogenic methane emissions. Area-integrated diffusive modern biogenic CH4 emissions 1386 1387 from coastal ecosystems are 1-2 magnitudes lower than from inland freshwaters but significantly higher than biogenic 1388 emissions from the open ocean (Rosentreter et al., 2021; Rosentreter et al., 2023; Weber et al., 2019). Particularly the shallow 1389 vegetated coastline fringed by mangroves, salt marshes, and seagrasses is a CH4 hotspot in the coastal ocean, characterised 1390 by significantly higher flux densities than other coastal settings such as estuaries or the continental shelves (Rosentreter et 1391 al., 2021; Rosentreter et al., 2023). Coastal ecosystems are thus being increasingly recognized as weak global sources to the 1392 atmosphere (Weber et al., 2019; Saunois et al., 2020; Rosentreter et al., 2021). Hydrogenotrophic and acetoclastic 1393 methanogenesis are largely outcompeted by sulphate reduction in coastal/marine sediments, which is often shown by a 1394 decreasing trend of CH4 concentrations with increasing salinity from upper tidal (low salinity) to marine (high salinity) 1395 regions. Much of the CH4 produced below the sulfate-reduction zone is indeed re-oxidized by sedimentary anaerobic 1396 methane oxidation or re-oxidized in the water column, leading to small emissions despite much larger production (Knittel 1397 and Boetius 2009; Regnier et al., 2011). Methylated compounds such as methylamines and methyl sulphides are non-1398 competitive substrates that are exclusively used by methanogens, therefore methylated methanogenesis can occur in coastal 1399 regions with high sulphate concentrations, for example, in organic-rich (Maltby et al., 2018), vegetated (Schorn et al., 2022),

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and hypersaline coastal sediments (Xiao et al., 2018). Coastal CH₄ can be driven by the exchange of pore water or
 groundwater (high in CH₄) with coastal surface waters in tidal systems, referred to as tidal pumping (Ovalle et al., 1990;
 Call et al., 2015). Anthropogenic impacts such as wastewater pollution and land-use change can increase CH₄ fluxes in

estuaries (Wells et al., 2020). A large increase of CH₄ emissions follows the conversion of natural coastal habitats to
aquaculture farms (Yuan et al., 2019; Yang et al., 2022).

1408 Currently available global modern biogenic CH4 flux data show high spatiotemporal variability within and between coastal 1409 systems, but also because of the overall global paucity of data. Therefore, global estimates have high uncertainties and show large ranges in both empirical (Rosentreter et al., 2021) and machine-learning based approaches (Weber et al., 2019). 1410 1411 According to a recent data-driven meta-data analysis, global estuaries, including tidal systems and deltas, lagoons, and fords, are estimated to emit median (Q1-Q3) 0.25 (0.07-0.46) Tg CH₄ yr⁻¹ (Rosentreter et al., 2023). Coastal vegetation, 1412 1413 including mangrove forests, salt marshes, and seagrasses are estimated to emit 0.77 (0.47-1.41) Tg CH₄ yr⁻¹, which is 3 1414 times more than global estuaries (Rosentreter et al., 2023). The combined median (Q1-Q3) emission of 1.01 (0.54-1.87) Tg 1415 CH₄ yr⁻¹ for coastal vegetation and estuaries by Rosentreter et al. (2023) is lower than the recent observation-based global 1416 synthesis including tidal flats and aquaculture ponds (median 1.49 (0.22-6.48) Tg CH₄ yr⁻¹) by Rosentreter et al. (2021). 1417 Total shallow coastal modern biogenic CH4 emissions based on existing data including emissions from estuaries, coastal 1418 vegetation (Rosentreter et al., 2023), tidal flats, and man-made coastal aquaculture ponds (Rosentreter et al., 2021) amount to median (Q1-Q3) 1.8 (0.59-5.57) Tg CH4 yr⁻¹. This range is about 3-4 times lower than the earlier global assessment by 1419 1420 Borges and Abril (2011) and also lower than the value of 4-5 Tg CH₄ yr⁻¹reported in the previous CH₄ budget for inner and 1421 outer estuaries including marshes and mangroves (Saunois et al., 2020), which was based on a significantly smaller dataset 1422 (n=80) and larger estuarine surface areas (Laruelle et al., 2013) than used here (Laruelle et al., 2025). 1423 The near-shore (0-50 m), inner shelf diffusive modern biogenic CH₄ flux of median (Q1-Q3) 1.33 (0.93-2.10) Tg CH₄ yr⁻¹

1424 by Weber et al. (2019) based on machine-learning is similar to the combined shallow coastal (estuaries and coastal 1425 vegetation) median by Rosentreter et al. (2021, 2023). Adding the diffusive modern biogenic CH₄ flux for the outer shelf (50-200 m) (median (Q1-Q3) of 0.54 (0.40-0.73) Tg CH₄ yr⁻¹) and for the slope (200-2000m) (median (Q1-Q3) of 0.28 1426 (0.22-0.37) Tg CH₄ yr⁻¹) (Weber et al., 2019), and excluding geological seepage regions (Etiope et al., 2019; see below), 1427 1428 gives a total median (Q1-Q3) of 3.95 (2.14-8.77) Tg CH4 yr⁻¹ for combined coastal shallow, near-shore, outer shelf and slope diffusive modern biogenic CH₄ emissions. The previous budget by Saunois (2020) also included poorly constrained 1429 emissions (upper bound value: 1-2 Tg CH₄ vr⁻¹) from large river plumes protruding onto the shelves. However, here we 1430 1431 assume that emissions from large river plumes are accounted for in the near-shore and outer shelf estimates by Weber et al. 1432 (2019). Area-integrated diffusive CH₄ emissions from the open ocean and deep seas (>2000 m) are much lower than from 1433 other coastal systems but amount to median (Q1-Q3) 0.91 (0.75-1.12) Tg CH₄ yr⁻¹ because of the large surface area of the 1434 open ocean (>300 x 106 km²) (Weber et al., 2019). Overall, these marine biogenic emissions are sustained by a mixture of

sedimentary production and in-situ production in the sea-surface layers (including the methylphosphonate pathway), (e.g.,

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Karl et al., 2008; Repeta et al., 2016; Resplandy et al., 2024). The total coastal and ocean diffusive modern biogenic
emissions retained here amount to 5 (3-10) Tg CH₄ yr⁻¹ (Table 3).

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1441 Coastal and oceanic geological methane emissions Submarine geological CH4 emission is the offshore component of the 1442 general geological emissions of natural gas from the Earth's crust (Judd, 2004; Etiope, 2009; Etiope et al., 2019). The 1443 onshore components include terrestrial seeps, mud volcanoes, microseepage, and geothermal manifestations, addressed in 1444 Sect. 3.2.3. Natural gas seeping at the seabed as bubble plumes can reach the surface in relatively shallow waters (<400 m). 1445 but CH4-rich bubble plumes reaching the atmosphere from depths >500 m have been observed in some cases (e.g., Solomon 1446 et al., 2009), and upwelling of bottom marine waters can, in theory, transport geological CH4 (dissolved) to the surface from 1447 any depth. This represents, however, a small and poorly known fraction of geological CH₄ emission. Geological CH₄ can be 1448 either microbial or thermogenic, produced throughout diverse geological periods in hydrocarbon source rocks in 1449 sedimentary basins (therefore it is always fossil, ¹⁴C-free). The seepage at the seafloor is typically related to tectonic faults, 1450 sometimes forming mud diapirs and mud volcanoes (Mazzini and Etiope, 2017). Published estimates of geological CH4 1451 submarine emissions range from 3 to 20 Tg yr⁻¹, with a best guess of 7 Tg yr⁻¹ (Etiope and Schwietzke, 2019; Etiope et al., 1452 2019 and references therein).

Here, the diffusive geological CH₄ emissions are estimated at 0.16 (0.11-0.24) Tg CH₄ yr⁻¹ for near-shore (0-50 m), 0.03 (0.02-0.05) Tg CH₄ yr⁻¹ for outer shelf (50-200 m), and 0.02 (0.01-0.03) Tg CH₄ yr⁻¹ for slope (200-2000 m) by calculating the fraction of the Weber et al. (2019) diffusive fluxes that occur within the identified geological seepage regions from Etiope et al. (2019). No geological seepage regions were identified in the open ocean and deep seas (> 2000 m).

1457 In this study, we consider the ebullitive flux as geologically sourced CH4. While modern biogenic CH4 gas production 1458 appears ubiquitous in shallow sediments (Fleischer et al., 2001; Best et al., 2006), no global dataset is currently available to estimate the biogenic ebullitive CH4 flux to the atmosphere. Omission of this flux thus constitutes a significant knowledge 1459 gap in the coastal and oceanic CH4 budget. Global geological CH4 ebullition from continental shelf and slope, referring only 1460 to depths <200 m, were estimated at 5.06 (1.99-8.16) Tg CH₄ yr⁻¹ (Weber et al., 2019). This estimate is based on prior 1461 1462 estimates of the geological flux from the seafloor (Hovland et al., 1993) and bubble transfer efficiency to the ocean surface 1463 (McGinnis et al., 2006). Etiope et al. (2019) estimated a partial fraction of geological emissions in the form of gas bubbles 1464 of 3.9 (1.8-6) Tg CH₄ yr⁻¹, only referring to the sum of published estimates from 15 geological seepage regions, which are 1465 also deeper than 200 m. Global extrapolation including other 16 identified seepage zones (where flux data are not available) was suggested to be at least 7 (3-10) Tg CH₄ yr⁻¹ (Etiope et al., 2019), and this value coincides with the mean emission value 1466 1467 (best guess) derived by combining literature data, see Etiope and Schwietzke (2019) for further details. It is worth noting that the Weber et al. (2019) estimate of 5.06 (1.99-8.16) Tg CH₄ yr⁻¹, which considers only the continental shelf at depths 1468 1469 <200 m, is compatible with the overall submarine emission of 7 (3-10) Tg CH₄ yr⁻¹ (including seeps > 200 m deep) indicated 1470 in Etiope and Schwietzke (2019) and Etiope et al. (2019). Although 300-400 m is considered a general depth limit for Supprimé: (Supprimé:) Supprimé: and Supprimé: (

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efficient transport (with limited oxidation and dissolution) of CH₄ bubbles to the atmosphere (e.g., Judd, 2004; Schmale et
al., 2005; Etiope et al., 2019), in some cases oil coatings on bubbles inhibit gas dissolution so that CH₄-rich bubbles can
reach the atmosphere from depths >500 m (e.g., Solomon et al., 2009). As mentioned above, a fraction of geological CH₄
released in deep seas (such as in the areas with gas-charged sediments inventoried in Fleischer et al., 2001) can also be
transported to the surface by upwelling bottom waters. Further research is needed to better evaluate the atmospheric impact
of such deep seeps.

1482Geological submarine emissions, thus, would amount to 0.21 (0.14-0.32) Tg CH4 yr⁻¹ in the form of a diffusive flux while1483the ebullitive flux would be 5.06 (3.01-7.88) Tg CH4 yr⁻¹, considering only < 200 m deep seepage, and 7 (3-10) Tg CH4 yr</td>1484' considering all data available (Etiope and Schwietzke, 2019). Here, we select the Etiope and Schwietzke (2019) assessment1485in order to account for all potential seepage areas, including those located at water depths > 200m. While we use the estimate1486by Etiope and Schwietzke (2019) estimate, we acknowledge that high uncertainty remains and other studies suggest a lower1487ranges of emissions based on radiocarbon (¹⁴C-CH4) data in ice cores (e.g., Hmiel et al., 2020). The suggested estimate may1488overestimate this source and be part of the top-down bottom-up discrepancy as discussed in Section 5.1.2,

489 As a result, here we report a (rounded) median of 12 Tg CH₄ yr⁻¹ with a range of 6-20 Tg CH₄ yr⁻¹ for all coastal and oceanic 490 sources (Table 3).

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1492 Methane emissions from gas hydrates. Among the different origins of coastal and oceanic CH4, hydrates have attracted a 1493 lot of attention. CH4 hydrates (or clathrates) are ice-like crystals formed under specific temperature and pressure conditions 1494 (Milkov, 2005). Hydrates may host either modern microbial CH₄, containing ¹⁴C and formed *in situ* in shallow sediments 1495 (this type of hydrates is also called "autochthonous") or fossil, microbial or thermogenic CH4, migrated from deeper 1496 sediments, generally from reservoirs in hydrocarbon-rich sedimentary basins (this type of hydrates is also called "allochthonous"; Milkov, 2005; Foschi et al., 2023). The total stock of marine CH4 hydrates is large but uncertain, with 1497 1498 global estimates ranging from hundreds to thousands of Pg CH4 (Klauda and Sandler, 2005; Wallmann et al., 2012). Note 1499 that the highly climate-sensitive subsea permafrost reservoir beneath Arctic Ocean shelves also contributes to the hydrate 1500 inventory (Ruppel and Kassler, 2017).

Concerning more specifically atmospheric emissions from marine hydrates, Etiope (2015) points out that current estimates of CH₄ air–sea flux from hydrates (2–10 Tg CH₄ yr⁻¹ in Ciais et al., 2013, or Kirschke et al., 2013) originate from the hypothetical values of Cicerone and Oremland (1988). No experimental data or estimation procedures have been explicitly described along the chain of references since then (Denman et al., 2007; IPCC, 2001; Kirschke et al., 2013; Lelieveld et al., 1998). It was estimated that ~473 Tg CH₄ has been released into the water column over 100 years (Kretschmer et al., 2015). Those few teragrams per year become negligible once consumption within the water column has been accounted for. While events such as submarine slumps may trigger local releases of considerable amounts of CH₄ from hydrates that may reach

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1509 the atmosphere (Etiope, 2015; Paull et al., 2002), on a global scale, present-day atmospheric CH₄ emissions from hydrates 1510 do not appear to be a significant source to the atmosphere, and at least formally, we should consider 0 (< 0.1) Tg CH₄ yr⁻¹ 1511 emissions.

1512 3.2.7 Terrestrial permafrost

1513 Permafrost is defined as frozen soil, sediment, or rock having temperatures at or below 0°C for at least two consecutive 1514 years (Harris et al., 1988). The total extent of permafrost in the Northern Hemisphere is about 14 million km² or 15% of the 1515 exposed land surface (Obu et al., 2019). As the climate warms, a rise in soil temperatures has been observed across the 1516 permafrost region, and permafrost thaw occurs when temperatures pass 0°C, often associated with melting of ice in the 1517 ground (Biskaborn et al., 2019). Permafrost thaw is most pronounced in southern and spatially isolated permafrost zones, 1518 but also occurs in northern continuous permafrost (Obu et al., 2019). Thaw occurs either as a gradual, often widespread, 1519 deepening of the active layer (surface soils that thaw every summer) or as more rapid localised thaw associated with loss of 1520 massive ground ice (thermokarst) (Turetsky et al., 2020). A total of 1000 ± 200 Pg of carbon can be found in the upper 3 1521 meters of permafrost region soils, or 1400-2000 Pg C for all permafrost (Hugelius et al., 2014; Strauss et al., 2021). 1522 The thawing permafrost can generate direct and indirect CH4 emissions. Direct CH4 emissions are from the release of 1523 CH₄ contained within the thawing permafrost. This flux to the atmosphere is small and estimated to be a maximum of 1 Tg CH4 yr⁻¹ at present (USEPA, 2010b). Increased release of CH4 from deep geogenic sources that occurs as seepage along 1524 525 permafrost boundaries and lake beds may also be considered a direct, and this is estimated to be 2 ± 0.4 Tg CH₄ yr⁻¹ (Walter 1526 Anthony et al., 2012). Indirect CH4 emissions are probably more important. They are caused by 1) methanogenesis induced 1527 when the organic matter contained in thawing permafrost becomes available for microbial decomposition; 2) thaw induced 1528 soil wetting and changes in land surface hydrology possibly enhancing CH4 production (McCalley et al., 2014; Schuur et al., 2022); and 3) the landscape topography changes driven by abrupt thaw processes and loss of ground ice, including the 1529 1530 formation of thermokarst lakes, hill-slope thermokarst, and wetland thermokarst (Turetsky et al., 2020). Such 1531 CH4 production is probably already significant today and is likely to become more important in the future associated with 1532 climate change and strong positive feedback from thawing permafrost (Schuur et al., 2022). However, indirect 1533 CH4 emissions from permafrost thawing are difficult to estimate at present, with very few data to refer to, and in any case 1534 largely overlap with wetland and freshwater emissions occurring above or around thawing areas. In a recent synthesis of 1535 full permafrost region CH4 budgets for the period 2000-2017, Hugelius et al. (2023) compared CH4 budgets from bottomup and top-down (atmospheric inversion models) approaches. They estimate an integrated bottom-up budget of 50 (23, 53; 1536 1537 mean upper and lower 95% CI) Tg CH4 yr⁻¹ while the top-down estimate is 19 (15, 24) Tg CH4 yr⁻¹. The bottom-up estimate 1538 is based on a combination of data-driven upscaling reported by Ramage et al. (2023) and process-based model estimates for 1539 wetland CH4 flux calculated from model ensembles used in Saunois et al. (2020). The top-down estimate is calculated from 1540 ensembles of atmospheric inversion models used in Saunois et al. (2020). Although it is difficult with direct process-

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attribution, fluxes of ca. 20-30 Tg CH₄ yr¹ in the bottom-up budget are caused by land cover types affected by previous permafrost thaw (thermokarst lakes, wetlands, hillslope). Because pre-thaw land cover types often have near neutral CH₄ balances (Ramage et al. 2023), these fluxes can largely be seen as driven by permafrost thaw, however the thaw may have occurred decades, or even centuries, before today.

Here, we choose to report only the direct emission range of 0-1 Tg CH₄ yr⁻¹ (Table 3), keeping in mind that current wetland,

thermokarst lakes and other freshwater methane emissions already likely include a significant indirect contribution originating from thawing permafrost.

1553 3.2.8 Vegetation

1554 Three distinct pathways for the production and emission of CH4 by living vegetation are considered here (see Covey and 1555 Megonigal (2019) and Bastviken et al. (2023) for extensive reviews). Firstly, plants produce CH4 through an abiotic photochemical process induced by stress (Keppler et al., 2006). This pathway was initially questioned (e.g., Dueck et al., 1556 1557 2007; Nisbet et al., 2009), and although numerous studies have since confirmed aerobic emissions from plants and better 1558 resolved its physical drivers (Fraser et al., 2015), global estimates still vary by two orders of magnitude (Liu et al., 2015). 1559 This plant source has not been confirmed in-field however, and although the potential implication for the global CH4 budget 1560 remains unclear, emissions from this source are certainly much smaller than originally estimated in Keppler et al. (2006) 1561 (Bloom et al., 2010; Fraser et al., 2015). Second, and of clearer significance, plant stems, act as "straws", drawing up and 1562 releasing microbially produced CH₄ from anoxic soils (Cicerone and Shetter, 1981; Rice et al., 2010; Nisbet et al., 2009). 1563 For instance, in the forested wetlands of Amazonia, tree stems are the dominant ecosystem flux pathway for soil-produced 1564 CH4, therefore, including stem emissions in ecosystem budgets can reconcile regional bottom-up and top-down estimates 1565 (Pangala et al., 2017; Gauci et al., 2022). Third, the stems of both living trees (Covey et al., 2012) and dead wood (Covey 1566 et al., 2016) provide an environment suitable for microbial methanogenesis. Static chambers demonstrate locally significant 1567 through-bark flux from both soil- (Pangala et al., 2013, 2015), and tree stem-based methanogens (Pitz and Megonigal, 2017; 1568 Wang et al., 2016). A synthesis indicates stem CH4 emissions significantly increase the source strength of forested wetlands, 569 and modestly decrease the sink strength of upland forests (Covey and Megonigal, 2019). Recently, field-work suggested 570 that trees may also act as a CH₄ sink (Machacova et al., 2021; Gorgolewski et al., 2023; Gauci et al., 2024). The scientific 1571 activity covering CH4 emissions in forested ecosystems reveals a far more complex story than previously thought, with an 1572 interplay of productive/consumptive, aerobic/anaerobic, and biotic/abiotic processes occurring between upland/wetland 1573 soils, trees, and atmosphere. Understanding the complex processes that regulate CH4 source-sink dynamics in forests and 1574 estimating their contribution to the global CH4 budget requires cross-disciplinary research, more observations, and new 1575 models that can overcome the classical binary classifications of wetland versus upland forest and of emitting versus uptaking 1576 soils (Barba et al., 2019; Covey and Megonigal, 2019). Although we recognize these emissions are potentially large

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(particularly tree transport from inundated soil), global estimates for each of these pathways remain highly uncertain and/or are currently included here within other flux category sources (e/g. inland waters, wetlands, upland soils).

1583 3.3 Methane sinks and lifetime

1584 CH4 is the most abundant reactive trace gas in the troposphere and its reactivity is important to both tropospheric and 1585 stratospheric chemistry. The main atmospheric sink of CH4 (~90% of the total sink mechanism) is oxidation by the hydroxyl radical (OH), mostly in the troposphere (Ehhalt, 1974). Other losses are by photochemistry in the stratosphere (reactions 1586 with chlorine atoms (Cl) and excited atomic oxygen (O(¹D)), oxidation in soils (Curry, 2007; Dutaur and Verchot, 2007), 1587 1588 and by photochemistry in the marine boundary layer (reaction with Cl; Allan et al. (2007), Thornton et al. (2010)). 1589 Uncertainties in the total sink of CH4 as estimated by atmospheric chemistry models are in the order of 20-40% (Saunois et 1590 al., 2016). It is much less (10-20%) when using atmospheric proxy methods (e.g., methyl chloroform, see below) as in atmospheric inversions (Saunois et al., 2016). In the present release of the global CH4 budget, we estimate bottom-up 1591 1592 CH4 chemical sinks and lifetime mainly based on global model results from the Chemistry Climate Model Initiative (CCMI) 2022 activity (Plummer et al., 2021) and CMIP6 simulations (Collins et al., 2017). 1593

1594 3.3.1 Tropospheric OH oxidation

1595 OH radicals are produced following the photolysis of ozone (O₃) in the presence of water vapour. OH is destroyed by

1596 reactions with carbon monoxide (CO), CH₄, and non-methane volatile organic compounds.

1597 Following the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP), which studied the long-term 1598 changes in atmospheric composition between 1850 and 2100 (Lamarque et al., 2013), a new series of experiments was 1599 conducted by several chemistry-climate models and chemistry-transport models participating in the Chemistry-Climate 1600 Model Initiative (CCMI) (Plummer et al., 2021). Mass-weighted OH tropospheric concentrations do not directly represent 1601 CH4 loss, as the spatial and vertical distributions of OH affect this loss through, in particular, the temperature dependency 1602 and the distribution of CH4 (e.g., Zhao et al., 2019). However, estimating OH concentrations and, spatial and vertical 1603 distributions is a key step in estimating methane loss through OH. Over the period 2000-2010, the global mass-weighted 1604 OH tropospheric concentration is estimated at 13.3 [11.7-18.2] x 10⁵ molecules cm⁻³ by 8 CCMI-2022 models and at 11.8 605 [9.4-13.5] x 10⁵ molecules cm³ by 9 models contributing CMIP6 historical run (Collins et al., 2021) (see supplementary 1606 Table S4). The ranges calculated here are similar to the ones proposed previously in Saunois et al. (2020), where the multi-1607 model mean (11 models) global mass-weighted OH tropospheric concentration was 11.7±1.0 x 10⁵ molecules cm⁻³ (range 1608 9.9-14.4 x 10⁵ molecules cm⁻³, Zhao et al. (2019)) consistent with the previous estimates from ACCMIP (11.7±1.0 x 1609 10⁵ molecules cm⁻³, with a range of 10.3-13.4 x 10⁵ molecules cm⁻³, Voulgarakis et al. (2013) for year 2000) and the 1610 estimates of Prather et al. (2012) of 11.2±1.3 x 10⁵ molecules cm⁻³. Nicely et al. (2017) attribute the differences in OH 1611 simulated by different chemistry transport models to, in decreasing order of importance, different chemical mechanisms,

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1616 various treatments of the photolysis rate of O₃, and modelled O₃ and CO. Besides the uncertainty on global OH 1617 concentrations, there is an uncertainty in the spatial and temporal distribution of OH. Models often simulate higher OH in 1618 the northern hemisphere (NH) than in the southern hemisphere (SH), leading to a NH/SH OH ratio greater than 1 (e.g., Zhao 1619 et al., 2019). However, there is evidence for parity in inter-hemispheric OH concentrations (Patra et al., 2014), which needs 1620 to be confirmed by other observational and model-derived estimates. The analysis of the latest CCMI (Plummer et al., 2021) 1621 and CMIP6 (Collins et al., 2021) model outputs show that structural uncertainties in the atmospheric chemistry models 622 remain large, probably due to inherent biases in OH precursors. Such biases have been highlighted in the OH 3D fields 623 simulated by two atmospheric chemistry models (Zhao et al., 2023), and were corrected using OH precursors observations. 1624 Such corrections, resulted in tropospheric OH mean concentrations lowered by 2. 10⁵ molecules cm⁻³, leading to around 10 1625 x 10⁵ molecules cm⁻³, and a NH/SH OH ratio closer to 1, in better agreement with methyl chloroform (MCF)-based 1626 approaches. This study highlights the need for further improvement of the atmospheric chemistry model. 627 OH concentrations and their changes can be sensitive to climate variability (e.g., Nicely et al., 2018; Anderson et al., 2021). 628 biomass burning (e.g., Anderson et al., 2024), and anthropogenic emissions of precursors (Peng et al., 2022; Stevenson et 629 al., 2020). OH distributions calculated by chemistry climate models show large regional differences and various vertical 630 profiles (Zhao et al., 2019). OH changes present also regional differences over the long term (Stevenson et al., 2020), Despite 631 Jarge regional changes, the global mean OH concentration was suggested to have changed only slightly from 1850 to 1980, 632 but followed by strong (9%) increases up to the present day (Stevenson et al., 2020). This increase simulated by models 633 over 2000-2015 are however not in agreement with observation-based approaches (Thompson et al., 2024; Patra et al., 2020; 634 Nicely et al., 2018; Rigby et al., 2017; Turner et al., 2017) where OH decreases or remain constant over the period. CCMI 635 and CMIP6 models show OH interannual variability ranging from 0.9% to 1.8% over 2000-2010 (Table S4), in agreement 636 with the values of IAV derived from some observationally constrained studies (e.g., Thompson et al., 2024; Montzka et al., 637 2011) but lower than value deduced from methyl chloroform measurements (Patra et al., 2021; Naus et al., 2021). However, 638 chemistry climate simulations consider meteorology variability but not fully emission interannual variability (e.g., from 1639 biomass burning) and thus are expected to simulate lower OH interannual variability than in reality. Using an empirical 1640 model constrained by global observations of O₃, water vapour, CH₄, and temperature as well as the simulated effects of 1641 changing NO_x emissions and tropical expansion, Nicely et al. (2017) found an interannual variability in OH of about 1.3-1642 1.6% between 1980 and 2015, in agreement with methyl chloroform based estimates (Montzka et al., 2011). 1643 Over 2000-2009, the tropospheric loss (tropopause height at 200 hPa) of CH4 by OH oxidation derived from the ten and 1644 CCMI modelling activities (see supplementary Table S5) is estimated at of 546 [446-663] Tg CH₄ yr¹ (Table 3), which is 1645 similar to the one reported previously in Saunois et al. (2020) from CCMI model (553 [476-677] Tg CH4 yr⁻¹) and still

slightly higher than the one from the ACCMIP models (528 [454-617] Tg CH₄ yr⁻¹ reported in Kirschke et al. (2013) and

1647 Saunois et al. (2016).

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For the recent 2010-2019 decade, we report a climatological value based on <u>only</u> five models that contributed to CMIP6 runs (historical run followed by SSP3-7.0 projections starting in 2015, Collins et al. (2021)) to acknowledge the impact of the rise in atmospheric methane on the methane chemical sink. Hence, for 2010-2019, we report the climatological value of 563 [462-663] Tg CH4 yr⁻¹(Table 3).

1692 3.3.2 Stratospheric loss

In the stratosphere, CH₄ is lost through reactions with excited atomic oxygen $O(^1D)$, atomic chlorine (Cl), atomic fluorine (F), and OH (Brasseur and Solomon, 2005; le Texier et al., 1988). Uncertainties in the chemical loss of stratospheric CH₄ are large, due to uncertain interannual variability in stratospheric transport (Zhang et al., 2023) as well as its chemical interactions and feedbacks with stratospheric O₃ (Morgenstern et al., 2018). Particularly, the fraction of stratospheric loss due to the different oxidants is still uncertain, with possibly 20-35% due to halons, about 25% due to $O(^1D)$ mostly in the high stratosphere and the rest due to stratospheric OH (McCarthy et al., 2003).

In this study, six chemistry climate models that contributed to CMIP6 modelling activities (Table S5) provided estimates of

 $1700 \qquad \text{CH}_4 \text{ chemical loss, including reactions with OH, O(^{1}\text{D}), and Cl; CH_4 \text{ photolysis is also included but occurs only above the}$

stratosphere. Considering a 200 hPa tropopause height, these six CMIP6 simulations suggest an estimate of 34 [10-51] Tg

 $CH_4 \text{ yr}^{-1} \text{ for the } CH_4 \text{ stratospheric sink for the 2000-2009 decade (Table S5), similar to the value derived from the previous}$

CCMI activity reported in Saunois et al. (2020) (31 [12-41] Tg CH₄ yr⁻¹). The lowest estimate provided by a model (10 Tg

704 CH₄ yr⁻¹) is quite unrealistic and would yield a methane stratospheric lifetime of several hundreds of years. As a result, this

oulier is excluded and we prefer to report a mean of 39 Tg CH₄ yr⁻¹ associated with a range of [27-51] for 2000-2009.

For 2010-2019, we report here a climatological range of $\frac{28}{24}$ 43 Tg CH₄ yr⁻¹ associated with a mean value of $3\frac{7}{2}$ Tg CH₄ yr⁻¹

(<u>Table 3</u>) based on five models that contributed to CMIP6 runs (historic followed by SSP3-7.0 projections starting in 2015;
 Table S5).

1709 3.3.3 Tropospheric reaction with Cl

1710 Halogen atoms can also contribute to the oxidation of CH4 in the troposphere. Allan et al. (2005) measured mixing ratios of 1711 methane and δ^{13} C-CH₄ at two stations in the southern hemisphere from 1991 to 2003, and found that the apparent kinetic 1712 isotope effect (KIE) of the atmospheric CH4 sink was significantly larger than that explained by OH alone. A seasonally 1713 varying sink due to Cl in the marine boundary layer of between 13 and 37 Tg CH4 yr⁻¹ was proposed as the explanatory 1714 mechanism (Allan et al., 2007; Platt et al., 2004). This sink was estimated to occur mainly over coastal and marine regions, 1715 where sodium chloride (NaCl) from evaporated droplets of seawater react with NO2 to eventually form Cl2, which then UV-1716 dissociates to Cl. However significant production of nitryl chloride (ClNO2) at continental sites has been recently reported 1717 (Riedel et al., 2014) and suggests the broader presence of Cl, which in turn would expand the significance of the Cl sink in 1718 the troposphere. Recently, Hossaini et al. (2016), Sherwen et al. (2016), and Wang et al. (2019b, 2021b) have made

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1729 significant improvements in tropospheric chemistry modelling and they conclude to an oxidation contribution of 2.6%, 2%, 1730 1% and 0.8%, respectively. These values correspond to a tropospheric CH₄ loss of around 12-13 Tg CH₄ yr⁻¹, 9 Tg CH₄ yr⁻¹ 1731 ¹, 5 Tg yr⁻¹, and 3 Tg CH₄ yr⁻¹ respectively, much lower than the first estimates by Allan et al. (2007). The recent work of 1732 Wang et al. (2021b) is the most comprehensive modelling study and based upon Sherwen et al. (2016) and Wang et al. 1733 (2019b). Both the KIE approach and chemistry transport model simulations carry uncertainties (extrapolations based on 1734 only a few sites and use of indirect measurements, for the former and missing sources, coarse resolution, underestimation 1735 of some anthropogenic sources for the latter). However, Gromov et al. (2018) found that Cl can contribute only 0.23% the tropospheric sink of CH4 (about 1 Tg CH4 yr⁻¹) in order to balance the global ¹³C(CO) budget (see their Table S1). While 1736 tropospheric Cl has a marginal impact on the total CH4 sink (few percents), it influences more significantly the atmospheric 1737 isotopic 8¹³C-CH₄ signal and improved estimates of the tropospheric Cl amount should be used for isotopic CH₄ modelling 1738 1739 studies (Strode et al., 2020; Thanwerdas et al., 2022b).

Each recent Cl estimate suggests a reduced contribution to the methane loss than previously reported by Allan et al. (2007).

As a result, we suggest here to use the mean, minimum and maximum of the last five estimates published since 2016, leading to a climatological value of 6 [1-13] Tg CH₄ yr⁻¹ (Table 3), thus reducing both the magnitude and the uncertainty range compared to Saunois et al. (2020).

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1745 3.3.4 Soil uptake

Unsaturated oxic soils are sinks of atmospheric CH4 due to the presence of methanotrophic bacteria, which consume CH4 as 1746 1747 a source of energy. Dutaur and Verchot (2007) conducted a comprehensive meta-analysis of field measurements of CH4 1748 uptake spanning a variety of ecosystems. Extrapolating to the global scale, they reported a range of 36 ± 23 Tg CH₄ yr¹, but also showed that stratifying the results by climatic zone, ecosystem, and soil type led to a narrower range (and lower mean 1749 1750 estimate) of 22 ± 12 Tg CH₄ yr⁻¹. Modelling studies, employing meteorological data as external forcing, have also produced 1751 a considerable range of estimates. Using a soil depth-averaged formulation based on Fick's law with parameterizations for 1752 diffusion and biological oxidation of CH4, Ridgwell et al. (1999) estimated the global sink strength at 38 Tg CH4 yr¹, with 1753 a range 20-51 Tg CH4 yr⁻¹ reflecting the model structural uncertainty in the base oxidation parameter. Curry (2007) improved on the latter by employing an exact solution of the one-dimensional diffusion-reaction equation in the near-surface soil layer 1754 1755 (i.e., exponential decrease in CH₄ concentration below the surface), a land surface hydrology model, and calibration of the oxidation rate to field measurements. This resulted in a global estimate of 28 Tg CH₄ yr⁻¹ (9-47 Tg CH₄ yr⁻¹), the result 1756 1757 reported by Zhuang et al. (2013), Kirschke et al. (2013) and Saunois et al. (2016). Ito and Inatomi (2012) used an ensemble 1758 methodology to explore the variation in estimates produced by these parameterizations and others, which spanned the range 1759 25-35 Tg CH₄ yr⁻¹. For the period 2000-2020, as part of the wetland emissions modelling activity, JSBACH (Kleinen et al., 1760 2020) and VISIT (Ito and Inatomi, 2012) models compute a global CH4 soil uptake to 18 and 35 Tg CH4 yr⁻¹, respectively.

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1762 Murguia-Flores et al. (2018) further refined the Curry (2007) model's structural and parametric representations of key 1763 drivers of soil methanotrophy, demonstrating good agreement with the observed latitudinal distribution of soil uptake 1764 (Dutaur and Verchot, 2007). Their model (MeMo) simulates a CH4 soil sink of 37.5 Tg CH4 yr⁻¹ for the period 2010-2019 1765 (Fig. S4), compared to 39.5 and 31.3 Tg CH₄ yr⁻¹ using the Ridgwell et al. (1999) and Curry (2007) parameterizations, respectively, under the same meteorological forcing, run specifically for this study. For the 2000s period, the simulations 1766 estimate the soil uptake at 30.4, 36.7 and 38.3 Tg CH₄ yr¹ based on the parameterization of Curry, MeMo, and Ridgwell, 1767 1768 respectively. As part of a more comprehensive model accounting for a range of CH₄ sources and sinks, Tian et al. (2010, 2015, 2016) computed vertically-averaged CH4 soil uptake including the additional mechanisms of aqueous diffusion and 1769 plant-mediated (aerenchyma) transport, arriving at the estimate 30±19 Tg CH4 yr⁻¹ (Tian et al., 2016) for the 2000s. The 1770 1771 still more comprehensive biogeochemical model of Riley et al. (2011) included vertically resolved representations of the 1772 same processes considered by Tian et al. (2016), in addition to grid cell fractional inundation and, importantly, the joint 1773 limitation of uptake by both CH4 and O2 availability in the soil column. Riley et al. (2011) estimated a global CH4 soil sink 1774 of 31 Tg CH₄ yr⁻¹ with a structural uncertainty of 15-38 Tg CH₄ yr⁻¹ (a higher upper limit resulted from an elevated gas 1775 diffusivity to mimic convective transport; as this is not usually considered, we adopt the lower upper bound associated with 1776 no limitation of uptake at low soil moisture). A model of this degree of complexity is required to explicitly simulate situations 1777 where the soil water content increases enough to inhibit the diffusion of oxygen, and the soil becomes a methane source 1778 (Lohila et al., 2016). This transition can be rapid, thus creating areas (for example, seasonal wetlands) that can be either a 1779 source or a sink of methane depending on the season. 1780 The previous Curry (2007) estimate can be revised upward slightly based on subsequent work and the increase in CH₄ 781 concentration since that time. Indeed, Murgia-Flores et al. (2021) estimated that the global soil-uptake doubled between 782 1900 and 2015 and could further increase due to enhanced diffusion of CHe into soil as a result of increases in atmospheric 783 CH₄ mole fraction. Further investigation of the soil uptake is required to better constrain this process at the global scale 784 while it is highly dependent on local scale microbial activity and environmental conditions (e.g., D'Imperio et al., 2023; 1785 Fest et al., 2017).

Considering the latest estimates (based on VISIT, JSBACH, and Memo models, Table S6 in the supplementary) we report
 here a mean estimate of 31 [17-39] Tg CH₄ yr⁻¹ for 2000-2009 and 32 [18-40] for 2010-2019 Tg CH₄ yr⁻¹ (Table 3).

1788 **3.3.5 CH4 lifetime**

1789 The atmospheric lifetime of a given gas in steady state may be defined as the global atmospheric burden (Tg) divided by the

total sink (Tg yr⁻¹) (IPCC, 2001). This value is different from what is called perturbation lifetime. Perturbation lifetime is

1791 used to determine how a one-time pulse emission may decay as a function of time as needed for the calculation of Global

Warming Potentials (GWPs), and as a result is related to a theoretical concept. For CH4, the corresponding perturbation

1793 lifetime that should be used in the GWP calculation is 11.8 ± 1.8 years (Forster et al., 2021). In this section, we discuss the

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global atmospheric lifetime (also called 'burden lifetime' or 'turnover lifetime') that characterises the time required to turn over the global atmospheric burden and defined as the burden divided by the removal flux.

1796 Global models provide an estimate of the loss of the gas due to individual sinks, which can then be used to derive lifetime due to a specific sink. For example, the tropospheric lifetime of CH4 is determined as the global atmospheric CH4 burden 1797 divided by the loss from OH oxidation in the troposphere, sometimes called "chemical lifetime". The total lifetime of 1798 1799 CH4 corresponds to the global burden divided by the total loss including tropospheric loss from OH oxidation, stratospheric chemistry and soil uptake. The CCMI (Plummer et al., 2021) and CMIP6 (Collins et al., 2021) runs estimate the tropospheric 1800 methane lifetime at about 9.2 years (average over years 2000-2009), with a range of 7.5-11 years (see Table S5). This range 1801 agrees with previous values found in ACCMIP and CCMI (9.3 [7.1-10.6] years, Voulgarakis et al. (2013), 9 [7.2-10.1] years, 1802 Saunois et al. (2020)). Adding 31 Tg to account for the soil uptake to the total chemical loss of the CMIP6 and CCMI 1803 1804 models, we derive a total CH₄ lifetime of 8.2 years (average over 2000-2009 with a range of 6.8-9.7 years). The lifetime 1805 calculated over 2010-2019 based on CMIP6 simulations is similar (Table S5). These updated model estimates of total CH4 lifetime agree with the previous estimates from ACCMIP (8.2 [6.4-9.2] years for year 2000, Voulgarakis et al. (2013)) 1806 1807 and Saunois et al. (2020) based CCMI models. Reducing the large spread in CH4 lifetime (between models, and between 1808 models and observation-based estimates) would 1) bring an improved constraint on global total methane emissions, and 2) 1809 ensure an accurate forecast of future climate.

1810 4 Atmospheric observations and top-down inversions

1811 4.1 Atmospheric observations

Systematic atmospheric CH₄ observations began in 1978 (Blake et al., 1982) with infrequent measurements from discrete 1812 1813 air samples collected in the Pacific at a range of latitudes from 67°N to 53°S. Because most of these air samples were from 1814 well-mixed oceanic air masses and the measurement technique was precise and accurate, they were sufficient to establish 1815 an increasing trend and the first indication of the latitudinal gradient of methane. Spatial and temporal coverage was greatly 1816 improved soon after (Blake and Rowland, 1986) with the addition of the Earth System Research Laboratory from US 1817 National Oceanic and Atmospheric Administration (NOAA/GML) flask network (Steele et al. (1987); Lan et al. (2024), Fig. 1), and the Advanced Global Atmospheric Gases Experiment (AGAGE) (Cunnold et al., 2002; Prinn et al., 2018), the 1818 1819 Commonwealth Scientific and Industrial Research Organisation (CSIRO, Francev et al. (1999)), the University of California Irvine (UCI, Simpson et al., 2012) and in situ and flask measurements from regional networks, such as ICOS (Integrated 1820 1821 Carbon Observation System) in Europe (https://www.icos-ri.eu/). The combined datasets provide the longest time series of 1822 globally averaged CH4 abundances. Since the early-2000s, CH4 column-averaged mole fractions have been retrieved through 1823 passive remote sensing from space (Buchwitz et al., 2005a, 2005b; Butz et al., 2011; Crevoisier et al., 2009; Frankenberg et 1824 al., 2005; Hu et al., 2018). Ground-based Fourier transform infrared (FTIR) measurements at fixed locations also provide

time-resolved CH₄ column observations during daylight hours, and a validation dataset against which to evaluate the satellite
 measurements such as the Total Carbon Column Observing Network (TCCON) network (e.g., Pollard et al., 2017; Wunch
 et al., 2011), or Network for Detection of Atmospheric Composition Change (NDACC) (e.g., Bader et al., 2017).

1828 In this budget, in-situ observations from the different networks were used in the top-down atmospheric inversions to estimate

1829 CH₄ sources and sinks over the period 2000-2020. Satellite observations from the TANSO/FTS instrument on board the 1830 satellite GOSAT were used to estimate CH₄ sources and sinks over the period 2010-2020. Other atmospheric data (FTIR, 1831 airborne measurements, AirCore, isotopic measurements, etc.) have been used for validation by some groups, but not 1832 specifically in this study. However, further information is provided in Tables S7, S8, S9, S10, and S11 and a more 1833 comprehensive validation of the inversions is planned to use some of these data.

1834 4.1.1 In situ CH₄ observations and atmospheric growth rate at the surface

We use globally averaged CH₄ mole fractions at the Earth's surface from the four observational networks (NOAA/GML, AGAGE, CSIRO and UCI). The data are archived at the World Data Centre for Greenhouse Gases (WDCGG) of the WMO Global Atmospheric Watch (WMO-GAW) program (https://gaw.kishou.go.jp/), including measurements from other sites that are not operated as part of the four networks. The CH₄ in-situ monitoring network has grown significantly over the last decade due to the emergence of laser diode spectrometers which are robust and accurate enough to allow deployments with low maintenance enabling the development of denser networks in developed countries (Stanley et al., 2018; Yver Kwok et al., 2015), and new stations in remote environments (Bian et al., 2015; Nisbet et al., 2019).

The networks differ in their sampling strategies, including the frequency of observations, spatial distribution, and methods 1842 1843 of calculating globally averaged CH4 mole fractions. Details are given in the supplementary material of Kirschke et al. 1844 (2013). The global average values of CH₄ abundances at Earth's surface presented in Fig. 1 are computed using long-term measurements from background conditions with minimal influence from immediate emissions. All measurements are 1845 1846 calibrated against gas standards either on the current WMO reference scale or on independent scales with well-estimate 1847 differences from the WMO scale. The current WMO reference scale, maintained by NOAA/ESRL, WMO-X2004A 1848 (Dlugokencky et al., 2005) was updated in July 2015. NOAA and CSIRO global means are on this scale. AGAGE uses an 1849 independent standard scale (based on work by Tohoku University (Aoki et al., 1992) and maintained at Scripps Institution 1850 of Oceanography (SIO)), but direct comparisons of standards and indirect comparisons of atmospheric measurements show 1851 that differences are well below 5 ppb (Tans and Zwellberg, 2014; Vardag et al., 2014) and the TU-1987 scale used for AGAGE measurements is only 0.5 ppb difference from WMO-X2004A at 1900 ppb level. UCI uses another independent 1852 1853 scale that was established in 1978 and is traceable to NIST (Flores et al., 2015; Simpson et al., 2012), but has not been 1854 included in standard exchanges with other networks so differences with the other networks cannot be quantitatively defined. 1855 Additional experimental details are presented in the supplementary material from Kirschke et al. (2013) and references 1856 therein.

1857 In Fig. 1 (a) globally averaged CH₄ and (b) its growth rate (derivative of the deseasonalized trend curve) through to 2022 1858 are plotted for the four measurement programs using a procedure of signal decomposition described in Thoning et al. (1989). 1859 We define the annual GATM as the increase in the atmospheric concentrations from Jan. 1 in one year to Jan. 1 in the next 1860 year. Agreement among the four networks is good for the global growth rate, especially since ~1990. The large differences observed mainly before 1990 probably reflect the different spatial coverage of each network. The long-term behaviour of 1861 1862 globally averaged atmospheric CH4 shows a positive growth rate (defined as the derivative of the deseasonalized mixing ratio) that is slowing down from the early-1980s through 1998, a near-stabilisation of CH4 concentrations from 1999 to 1863 2006, and a renewed period with positive persistent overall accelerating growth rates since 2007, slightly larger after 2014. 1864 From 1999 to 2006, the annual increase of atmospheric CH₄ was remarkably small at 0.6±0.1 ppb yr⁻¹. After 2006, the 1865 atmospheric growth rate has increased to a level similar to that of the mid-1990s (~5 ppb yr⁻¹), and for 2014 and 2015 even 1866 1867 to that of the 1980s (>10 ppb yr⁻¹). In the two recent years 2020 and 2021, the highest growth rates of 15 ppb yr⁻¹ and 18 1868 ppb yr⁻¹ (see Sect. 6) were unprecedented since the 1980s. On decadal timescales, the annual increase is on average 2.2 ± 0.3 1869 ppb yr⁻¹ for 2000-2009, 7.6±0.3 ppb yr⁻¹ for 2010-2019 and 15.2±0.4 ppb yr⁻¹ for the year 2020 (Table 3). Both climate 870 variability and anthropogenic emission changes are responsible for variations in atmospheric CH4 growth rates. Indeed, 871 climate variation such as El Nino Southern Oscillation induce changes in emissions such as biomass burning or wetland 1872 emission but also impact OH oxidation (e.g., Rowlinson et al., 2019; Zhao et al., 2020b; Peng et al., 2022),

1873 4.1.2 Satellite data of column average CH₄

In this budget, we use satellite data from the JAXA satellite Greenhouse Gases Observing SATellite (GOSAT) launched in 1874 1875 January 2009 (Butz et al., 2011; Morino et al., 2011) containing the TANSO-FTS instrument, which observes in the 1876 shortwave infrared (SWIR). Different retrievals of CH4 based on TANSO-FTS/GOSAT products are made available to the community: from NIES (Yoshida et al., 2013), from SRON (Schepers et al., 2012) and from University of Leicester (Parker 1877 1878 et al., 2020; Parker and Boesch, 2020). The three retrievals are used by the top-down systems (Table 4 and S6). Although 1879 GOSAT retrievals still show significant unexplained biases and limited sampling in cloud covered regions and in the high 1880 latitude winter, it represents an important improvement compared to the first satellite measuring CH4 from space, 1881 SCIAMACHY (Scanning Imaging Absorption spectrometer for Atmospheric CartograpHY) both for random and systematic 1882 observation errors (see Table S2 of Buchwitz et al. (2016)).

1883 Here, as in Saunois et al. (2020), only inversions using GOSAT retrievals are used.

1884 4.2 Top-down inversions used in the budget

1885 An atmospheric inversion is the optimal combination of atmospheric observations, of a model of atmospheric transport and 1886 chemistry, of a prior estimate of CH4 sources and sinks, and of their uncertainties, to provide improved estimates of the **Supprimé:** When a constant atmospheric lifetime is assumed, the decreasing growth rate from 1983 through 2006 may imply that atmospheric CH₄ was approaching steady state, leading to no trend in emissions. The NOAA global mean CH₄ concentration was fitted with a function that describes the approach to a first-order steady state (sindex): $[CH_4](0) = [CH_4]_{arc}(CH_4]_{arc}e(CH_4]_{arc}e(CH_4)_{arc}$

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sources and sinks, and their uncertainty. The theoretical principle of CH₄ inversions is detailed in the Supplementary
Material and an overview of the different methods applied to CH₄ is presented in Houweling et al. (2017).

1904 We consider an ensemble of inversions gathering various chemistry transport models, differing in vertical and horizontal 1905 resolutions, meteorological forcing, advection and convection schemes, and boundary layer mixing. Including these different systems is a conservative approach that allows us to cover different potential uncertainties of the inversion, among 1906 1907 them: model transport, set-up issues, and prior dependency. General characteristics of the inversion systems are provided in 1908 Table 4. Further details can be found in the referenced papers and in the Supplementary Material. Each group was asked to provide gridded flux estimates for the period 2000-2020, using either surface or satellite data, but no additional constraints 1909 1910 were imposed so that each group could use their preferred inversion setup. Two sets of prior emission distributions were 1911 built from the most recent inventories or model-based estimates (see Supplementary Material), but its use was not mandatory 1912 (see Table S8 to S11 for the inversion characteristics). This approach corresponds to a flux assessment, but not to a model 1913 inter-comparison as the protocol was not too stringent. Estimating posterior uncertainty is time and computer resource 1914 consuming, especially for the 4D-var approaches and Monte Carlo methods. Posterior uncertainties have not been requested 1915 for this study, but they were found to be lower than the ensemble spread in Saunois et al. (2020). Indeed, chemistry transport 1916 models differ in inter-hemispheric transport, stratospheric CH4 profiles, and OH distribution, limitations which are not fully 1917 considered in the individual posterior uncertainty. As a result, we report the minimum-maximum range among the different 1918 top-down approaches.

1919 Seven atmospheric inversion systems using global Eulerian transport models were used in this study; they contributed to the 1920 previous budgets that included eight atmospheric inversion systems in Saunois et al. (2016) and nine in Saunois et al. (2020). 1921 Each inversion system provided one or several simulations, including sensitivity tests varying the assimilated observations 1922 (surface or satellite), the OH interannual variability, or the prior fluxes ensemble. This represents a total of 24 inversion runs 1923 with different time coverage: generally, 2000-2020 for surface-based observations, and 2010-2020 for GOSAT-based 1924 inversions (Table 4 and Table S7). In poorly observed regions, top-down surface inversions may rely on the prior estimates and bring little or no additional information to constrain (often) spatially overlapping emissions (e.g., in India, China). Also, 1925 1926 we recall that many top-down systems solve for the total fluxes at the surface only or for some categories that may differ 1927 from the GCP categories. When multiple sensitivity tests were performed the mean of this ensemble was used not to 1928 overweight one particular inverse system. It should also be noticed that some satellite-based inversions are in fact combined 1929 satellite and surface inversions as they use surface-based inversions to correct the latitudinal bias of the satellite retrievals 1930 against the optimised atmosphere measurements to correct for errors in the transport model especially in the stratosphere 1931 (e.g., Segers et al., 2022; Maasakkers et al., 2019). Nevertheless, these inversions are still referred to as satellite-based 1932 inversions. Most of the top-down models use the OH distribution from the TRANSCOM experiment (Patra et al., 2011) 1933 either as fixed over the period or with the interannual variability derived by Patra et al. (2021).

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Each group provided gridded monthly maps of emissions for both their prior and posterior total and for sources per category (see the categories Sect. 2.3). Results are reported in Sect. 5. Atmospheric sinks from the top-down approaches have been provided for this budget, and are compared with the values reported in Saunois et al. (2020). Not all inverse systems report their chemical sink; as a result, the global mass imbalance for the top-down budget is derived as the difference between total

1940 sources and total sinks for each model when both fluxes were reported.

1941 5 Methane budget: top-down and bottom-up comparison

1942 5.1 Global methane budget

943 5.1.1 Global total methane emissions,

944 Top-down estimates. At the global scale, the total annual emissions inferred by the ensemble of 24 inversions is 575 [553-945 586] Tg CH₄ yr¹ for the 2010-2019 decade (Table 3), with the highest ensemble mean emission of 608 [581-627] Tg CH₄ 946 yr¹ for 2020. Global emissions for 2000-2009 (543 Tg CH₄ yr¹) are consistent with Saunois et al. (2016, 2020) and the 1947 range for global emissions, 526-558 Tg CH₄ yr⁻¹ falls within the range in Saunois et al. (2016) (535-569) and Saunois et al. 1948 (2020) (524-560), although the ensemble of inverse systems contributing to this budget is different from Saunois et al. (2016, 2020). Changes in ensemble members contributing to the different budgets are a feature of each new GMB release and, 1949 1950 therefore, introduce a source of variation (Table S7). The range reported gives the minimum and maximum values among 1951 studies and does not reflect the individual full uncertainties. In addition, most of the top-down models use the same OH 1952 distribution from the TRANSCOM experiment (Patra et al., 2011), which introduces less variability to the global budget 953 than is likely justified, and so contributes to the rather low range (10%) compared to bottom-up estimates (see below). We 954 recall here that Zhao et al. (2020a) found an uncertainty of about 17% in global methane emissions (518 to 611 Tg CH₄ yr 1955 ¹ for the early 2000s) due to changes in OH burden and distribution (OH ranging from 10.3 to 12.6 10⁵ molec cm³)

Bottom-up estimates. The bottom-up estimates considered here differ substantially from the top-down results, with annual 1956 1957 global emissions being about 15% larger at 669 [512-849] Tg CH4 yr¹ for 2010-2019 (Table 3). Yet, thanks to the double 1958 counting corrections in this budget, bottom-up and top-down budgets are in better agreement compared to previous GMB 1959 releases. For the period 2000-2009, the discrepancy between bottom-up and top-down was about 30% of the top-down 1960 estimates in Saunois et al. (2016, 2020) (167 and 156 Tg CH₄ yr⁻¹, respectively), a value that has been reduced significantly 1961 in this budget (now 95 Tg CH4 yr⁻¹ (<17%) for the same 2000-2009 period). This reduction is due to improvements from an 1962 important decrease in the estimate of emissions from natural and indirect anthropogenic emissions from bottom-up approaches, and more specifically inland freshwater emissions. From the previous budget, the estimate for inland freshwater 1963 1964 emissions (lakes, ponds, reservoirs, rivers, and streams) has decreased from 159 Tg CH₄ yr⁻¹ to 112 Tg CH₄ yr⁻¹ (47 Tg 1965 decrease). Then, 23 Tg have been removed in the total freshwater ecosystem emissions due to double counting between 1966 vegetated wetlands and mostly small ponds and lakes (Sect. 3.2.2). As a result, the combined wetland and inland freshwater

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emissions are estimated to be 242 Tg CH₄ yr¹ for 2000-2009 (Table 3), compared with 306 Tg CH₄ yr¹ in Saunois et al. (2020).

1973 This budget is the first that reconciles bottom-up and top-down total emissions within the uncertainty ranges. However, the

1974 uncertainty in the global budget remains high because of the large range reported for emissions from freshwater systems.

1975 Still, the upper bound of global emissions from bottom-up approaches is not consistent with top-down estimates that rely

1976 on OH burden constrained by methyl chloroform atmospheric observations and is still likely overestimated.

1977 5.1.2 Global methane emissions per source category

1987

1978 The global CH₄ emissions from natural and anthropogenic sources (see Sect. 2.3) for 2010-2019 are presented in Fig. 6, Fig. 1979 7, and Table 3. Top-down estimates attribute about 65% of total emissions to anthropogenic activities (range of 55-70%), 1980 and 35% to natural emissions. Bottom-up estimates attribute 57% of emissions to direct anthropogenic and the rest to natural 1981 plus indirect anthropogenic emissions. A current predominant role of direct anthropogenic sources of CH4 emissions is 1982 consistent with and strongly supported by available ice core and atmospheric CH4 records. These data indicate that 1983 atmospheric CH₄ varied around 700 ppb during the last millennium before increasing by a factor of 2.6 to ~1800 ppb since 1984 pre-industrial times. Accounting for the decrease in mean-lifetime over the industrial period, Prather et al. (2012) estimated 1985 from these data a total source of 554±56 Tg CH4 in 2010 of which about 64% (352±45 Tg CH4) was of direct anthropogenic 1986 origin, consistent with the range in our top-down estimates.

1988Natural and indirect anthropogenic emissions. Although smaller than in previous Global Methane Budget releases, the1989main remaining discrepancy between top-down and bottom-up budgets is found for the natural and indirect anthropogenic1990emission total (105 Tg), with 311 [183-462] Tg CH4 yr⁻¹ for bottom-up and only 206 [188-225] Tg CH4 yr⁻¹ for top-down1991over the 2010-2019 decade (Table 3). In the bottom-up estimates, this discrepancy comes first from the estimates in both1992inland freshwater sources (64 Tg) and second from other natural sources (20 Tg from geological sources, termites, oceans,1993and permafrost). The top-down approaches may be biased due to missing fluxes (mainly inland freshwaters) in their prior1994estimates.

1995 For 2010-2019, the top-down and bottom-up derived estimates for wetlands emissions of 165 [145-214] Tg CH4 yr⁻¹ and 1996 159 [119-203] Tg CH₄ yr⁻¹ (Table 3), respectively, are comparable within their range. Based on diagnostic wetland area 1997 values (see notes in Table 3), bottom-up mean wetland emissions for the 2000-2009 period are smaller in this study than 1998 those of Saunois et al. (2016) but larger than in Saunois et al. (2020). The changes in wetland emissions from bottom-up 1999 models may be related to updates on the wetland extent data set (WAD2M), the use of two different meteorological forcings for this study and a different set of models (see Sect. 3.2.1). Conversely, the current 2000-2009 mean top-down wetland 2000 2001 estimates are lower than those of Saunois et al. (2016) and Saunois et al. (2020) (Table 3). In the bottom-up estimates, the 2002 amplitude of the range of emissions of 116-189 is roughly similar to Saunois et al. (2016) (151-222) and Saunois et al.

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2005 (2020) (102-179) for 2000-2009. Here, the larger range in bottom-up estimates of wetland emissions is due to the use of 2006 GSWP3-W5E5 and greater sensibilities of some models to the climate parameters, as discussed in Sect. 3.2.1. Bottom-up 2007 and top-down estimates for wetland emissions agree better in this study (~5 Tg yr¹ for 2000-2009) than in Saunois et al. 2008 (2016, 2020) (~17 Tg yr⁻¹ and ~30 Tg yr⁻¹, respectively). Natural emissions from inland freshwater systems were not included in the prior fluxes used in the top-down approaches, due to unavailable or uncertain gridded products at the start 2009 2010 of the modelling activity. However, emissions from these inland freshwater systems may be implicitly included in the 2011 posterior estimates of the top-down models, as these two sources are close and probably overlap at the rather coarse 2012 resolution of the top-down models. This is the reason why the 'wetland emissions' in the top-down budget in fact better correspond to the sum of combined wetland and inland freshwaters emissions in the bottom-up budget. The double-counting 2013 of 23 Tg CH₄ reduces the bottom-up budget for combined wetland and inland freshwaters from 271 Tg CH₄ yr⁻¹ to 248 Tg 2014 2015 CH₄ yr⁻¹ (Sect. 3.2.2). Comparing the 2000-2009 decadal emissions from wetlands and inland freshwater ecosystems 2016 estimated by the bottom-up approaches across the last three Global Methane Budgets shows an upward and then a downward 2017 revision with 305 (183+122) Tg CH₄ yr⁻¹, 356 (147+209) Tg CH₄ yr⁻¹ and 248 (159+112-23) Tg CH₄ yr⁻¹ (respectively from 2018 Saunois et al. (2016, 2020) and this work; the sum in bracket corresponds to the sum of vegetated wetland emissions and 2019 inland water emissions estimated through the different budgets). The combined wetland and inland freshwater emissions 2020 discrepancy between bottom-up and top-down approaches amount to 105 Tg CH₄ yr⁻¹ for the 2010-2019 decade. From a 2021 top-down point of view, the sum of all the natural sources is more robust than the partitioning between wetlands, inland 2022 waters, and other natural sources. Including all known spatio-temporal distributions of natural emissions in top-down prior 2023 fluxes would be a step forward to consistently compare natural versus anthropogenic total emissions between top-down and 2024 bottom-up approaches.

In the top-down budget, wetlands represent 28% on average of the total methane emissions but only 24% in the bottom-up budget (because of higher total emissions inferred) (see Table 3). Given the large uncertainties, neither bottom-up nor topdown approaches included in this study point to significant changes in wetland emissions between the two decades 2000-2009 and 2010-2019 at the global scale.

2029 For the 2010-2019 decade, top-down inversions infer "Other natural emissions" (Table 3) at 43 [40-46] Tg CH4 yr⁻¹, whereas 2030 the sum of the individual bottom-up emissions is 63 [24-93] Tg CH₄ yr⁻¹ contributing to a 20 Tg discrepancy between 2031 bottom-up and top-down approaches. Atmospheric inversions infer the same amount over the decade 2000-2009 as over 2032 2010-2019, which is almost half of the value reported in Saunois et al. (2016) (68 [21-130] Tg CH4 yr⁻¹). This reduction in 2033 magnitude and uncertainty is due to 1) a more consistent way of considering other natural emissions in the various inverse 2034 systems (same prior estimate as in this budget) and 2) a difference in the ensemble of top-down inversions reported here 2035 compared to previous releases. It is worth noting that, most of the top-down models include about the same ocean and 2036 onshore geological emissions and termite emissions in their prior scenarios. However, none include freshwater or 2037 permafrost emissions in their prior fluxes, and thus in their posterior estimates.

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2041 Geological emissions are associated with relatively large uncertainties, and marine seepage emissions are still widely 2042 debated (Thornton et al., 2020). However, summing up all bottom-up fossil-CH4 related sources (including anthropogenic 2043 emissions) leads to a total of 165 [135-190] Tg CH₄ yr⁻¹ in 2010-2019, which is about 29% of the top-down global 2044 CH4 emissions, and 25% of the bottom-up total global estimate. These results agree with the value inferred from ¹⁴C 2045 atmospheric isotopic analyses of 30% contribution of fossil-CH4 to global emissions (Etiope et al., 2008; Lassev et al., 2046 2007b). This total fossil fuel emissions from bottom-up approaches agrees well with the ¹³C-based estimate of Schwietzke et al. (2016) of 192 ± 32 Tg CH₄ yr⁻¹. In the bottom-up budget, the larger total emissions (due to uncertainties in bottom-up 2047 estimates of natural emissions) leads to a lower fossil fuel contribution compared to Lassey et al. (2007b). 2048

2049 Anthropogenic direct emissions. Total anthropogenic direct emissions for the period 2010-2019 were assessed to be statistically consistent between top-down (369 Tg CH₄ yr⁻¹, range 350-391) and bottom-up approaches (358 Tg CH₄ yr⁻¹, 2050 2051 range 329-387), albeit top-down approaches infer direct anthropogenic emissions larger by 11 Tg CH₄ yr⁻¹ on average 2052 compared to bottom-up approaches (Table 3). The partitioning of anthropogenic direct emissions between agriculture and 2053 waste, fossil fuels extraction and use, and biomass and biofuel burning, also shows good consistency between top-down and 2054 bottom-up approaches, though top-down approaches still suggest less fossil fuel and more agriculture and waste emissions 2055 than bottom-up estimates (Table 3 and Fig. 6 and 7). For 2010-2019, agriculture and waste contributed an estimated 2056 228 [213-242]Tg CH4 yr⁻¹ in the top-down budget and 211 [195-231]Tg CH4 yr⁻¹ in the bottom-up budget. Fossil fuel 2057 emissions contributed 115 [100-124] Tg CH4 yr⁻¹ in the top-down budget and 120 [117-125] Tg CH4 yr⁻¹ in the bottom-up 2058 budget. Biomass and biofuel burning contributed 27 [26-27] Tg CH4 yr¹ in the top-down budget and 28 [21-39]Tg CH4 yr 2059 ¹ in the bottom-up budget. Biofuel CH₄ emissions rely on very few estimates currently (Wuebbles and Hayhoe, 2002). 2060 Although biofuel is a small source globally (~12 Tg CH₄ vr⁻¹), more estimates are needed to allow a proper uncertainty 2061 assessment. Overall for top-down inversions the global fraction of total emissions for the different source categories is 40% 2062 for agriculture and waste, 20% for fossil fuels, and 5% for biomass and biofuel burning. With the exception of biofuel 2063 emissions, the uncertainty associated with global anthropogenic emissions appears to be smaller than that of natural sources 2064 but with an asymmetric uncertainty distribution (mean significantly different than median). The relative agreement between 2065 top-down and bottom-up approaches may indicate a limited capability of the inversion to separate emissions and a 2066 dependency to their prior fluxes; this agreement should therefore be treated with caution. Indeed, in poorly observed regions, 2067 top-down inversions rely on the prior estimates and bring little or no additional information to constrain (often) spatially overlapping emissions (e.g., in India, China). Also, as many top-down systems solve for the total fluxes at the surface or for 2068 2069 some categories that may differ from the GCP categories, their posterior partitioning relies on the prior ratio between 2070 categories that are prescribed using bottom-up inventories.

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2078 5.1.3 Global budget of total methane sinks

2079 Top-down estimates. The annual CH₄ chemical removal from the atmosphere is estimated to be 521 Tg CH₄ yr⁻¹ averaged 2080 over the period 2010-2019, with an uncertainty of about $\pm 2\%$ (range 485-532 Tg CH₄ yr⁻¹) (Table 3). All the inverse models 2081 account for CH₄ oxidation by OH and O(¹D), and some include stratospheric Cl oxidation (Table S8 to S11). Most of the 2082 top-down models use the OH distribution from the TRANSCOM experiment (Patra et al., 2011) either as fixed over the 2083 period or including interannual variability from Patra et al. (2021). This study shows no trend in OH and IAV below ±4%, 2084 in agreement with Thompson et al. (2024) (no significant OH trend and IAV \leq 2%). As a result, the range of the top-down 2085 sink estimates is rather low compared to bottom-up estimates (see below). Differences between transport models affect the 2086 chemical removal of CH4, leading to different chemical loss rates, even with the same OH distribution. However, 2087 uncertainties in the OH distribution and magnitude (around $\pm 10\%$ at the global scale, Zhao et al., 2019) are not considered 2088 in our study, while they could contribute to a significant change in the chemical sink, and then in the derived posterior 2089 emissions through the inverse process ((Zhao et al., 2020), around $\pm 17\%$ at the global scale, much larger than the model 2090 spread derived here. The chemical sink represents more than 90% of the total sink, the rest being attributable to soil uptake $(35 [35-36] \text{Tg CH}_4 \text{vr}^1)$. The rather narrow range is due to the use of the same climatological soil sink provided within the 2091 modelling protocol which is based on Murgia-Flores et al. (2018). This sink estimate used as prior in the inversions is a bit 2092 2093 higher than the mean estimate of the soil sink calculated by bottom-up models (30 Tg CH₄ yr⁻¹, Sec. 3.3.4).

Bottom-up estimates. The total chemical loss for the 2010s reported here is $602 \text{ Tg CH}_4 \text{ yr}^{-1}$ with an uncertainty of 21% (~125 Tg CH₄ yr⁻¹). Differences in chemistry schemes in the models (especially in the stratosphere) and in the volatile organic compound treatment probably explain most of the discrepancies among models (Zhao et al., 2019).

2097 5.2 Latitudinal and regional methane budgets

2098 The latitudinal and regional breakdown of the bottom-up budget is based on crude assumptions that we acknowledge here. 2099 Natural and indirect anthropogenic emissions are based on wetland gridded products from land surface models and the 2100 combination of the maps from lakes and ponds from Johnson et al. (2022), reservoirs from Johnson et al. (2022) and streams 2101 and rivers from Rocher-Ros et al. (2023), the sum of those three scaled to 89 Tg CH_4 yr⁻¹ (shown in Fig. 5) to artificially include the double counting (estimated only at the global scale) and match the global estimate. However, we acknowledge 2102 2103 that this procedure distributes the double counting relatively to the final emission distribution and not according to the 2104 freshwater ecosystems where the double counting probably occurs. Wild animal and permafrost maps do not exist and are 2105 missing from the calculation, leading to at least 3 Tg CH4 yr¹ of discrepancy. However, as aforementioned (Sections 3.2.5 2106 and 3.2.7) this 3 Tg CH4 yr⁻¹ estimate is probably underestimated in the bottom-up budget. Geological and ocean sources 2107 are based on Etiope et al. (2019) and Weber et al. (2019) gridded products scaled to 50 Tg CH₄ yr¹ to be consistent to the 2108 reported global values. Finally, we use the termite emission map produced for this budget and used in the global budget.

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The latitudinal budget does not include the estimates from FAO and USEPA for the direct anthropogenic emissions as they are only provided at country scale.

2114 5.2.1 Latitudinal budget of total methane emissions

2115 The latitudinal breakdown of emissions inferred from atmospheric inversions reveals a dominance of emissions in the 2116 latitudinal band 90°S-30°N of 364 [337-390] Tg CH4 yr 1 representing 64% of the global total (Table 5 and 6). As emissions 2117 in the Tropics (30°S-30°N) dominate this latitudinal contribution, we may refer to 90°S-30°N as the Tropics in the following 2118 32% of the emissions are from the mid-latitudes (187 [160-204] Tg CH4 yr¹) and 4% from high latitudes (above 60°N). 119 While the amounts of emissions depend on the surface area of the regions, the relative contribution of the emissions is much 120 larger (12 points of percent) than the relative importance of the surface areas for the 90°S-30°N region, on the contrary the 2121 boreal regions (60°N-90°N) emissions contribute significantly less than the relative importance of their surface areas (9 2122 points of percent). The ranges around the mean latitudinal emissions are larger than for the global CH₄ sources. While the 2123 top-down uncertainty is less than $\pm 5\%$ at the global scale, it increases to $\pm 7\%$ for the tropics, to $\pm 12\%$ the northern mid-2124 latitudes and to more than ±20% in the northern high-latitudes (for 2010-2019, Table 5). Both top-down and bottom-up 2125 approaches consistently show that CH4 decadal emissions have increased by +21-27 Tg CH4 yr⁻¹ in the tropics, and by +5-2126 16 Tg CH₄ yr⁻¹ in the northern mid-latitudes between 2000-2009 and 2010-2019 using the mean ensemble estimate. 2127 Over 2010-2019, at the global scale, satellite-based inversions infer almost identical emissions to ground-based inversions

2128 (difference of +1 [-3-9] Tg CH₄ yr⁻¹, with GOSAT based inversion a bit higher than surface measurements-based inversions), 2129 when comparing consistently surface versus satellite-based inversions for each system, similar to Saunois et al. (2020). This 2130 difference is much lower than the range derived between the different systems (range of 20 Tg CH₄ yr⁻¹ using surface- or 2131 satellite-based inversions). This result reflects that differences in atmospheric transport among the systems probably have 2132 more impact on the estimated global emissions than the types of observations assimilated.

2133 As expected, considering the different coverage of observation datasets, regional distributions of inferred emissions differ 2134 depending on the nature of the observations used (satellite or surface). The largest differences (satellite-based minus surface-2135 based inversions) are observed over the tropical region, between -10 and +43 Tg CH₄ yr⁻¹ (90°S to 30°N), and the northern 2136 mid-latitudes (between -36 and -2 Tg CH4 yr⁻¹). Satellite data provide stronger constraints on fluxes in tropical regions than 2137 surface data, due to a much larger spatial coverage. It is therefore not surprising that differences between these two types of 2138 observations are found in the tropical band, and consequently in the northern mid-latitudes to balance total emissions, thus affecting the north-south gradient of emissions. However, the regional patterns of these differences are not consistent 2139 2140 through the different inverse systems. Indeed, some systems found higher emissions in the tropics when using GOSAT instead of surface observations, while others found the opposite. This difference between inversion systems may depend on 2141 2142 whether or not a bias correction is applied to the satellite data based on surface observations, and also on the modelled 2143 horizontal and vertical transports, in the troposphere and in the stratosphere.

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2147 5.2.2 Latitudinal methane emissions per source category

The analysis of the latitudinal CH₄ budget per source category (Fig. 8 and Table 6) can be performed both for bottom-up and top-down approaches but with limitations. Bottom-up estimates of natural and indirect anthropogenic emissions are based on assumptions as specified at the beginning of this section 5.2. For top-down estimates, as already noted, the partitioning of emissions per source category has to be considered with caution. Indeed, using only atmospheric CH₄ observations to constrain CH₄ emissions makes this partitioning largely dependent on prior emissions. However, differences in spatial patterns and seasonality of emissions can be utilised to constrain emissions from different categories by atmospheric methane observations (for those inversions solving for different sources categories, see Sect. 2.3).

2155 Agriculture and waste are the largest sources of CH₄ emissions in the tropics and southern hemisphere (140 [121-150] Tg 2156 CH₄ yr⁻¹ in the bottom-up budget and 150 [135-168] Tg CH₄ yr⁻¹ in the top-down budget, about 40% of total CH₄ emissions 2157 in this region) (Table 6). However, combined wetland and inland freshwater emissions are nearly as large with 151 [85-234] 2158 Tg CH₄ yr⁻¹ in the bottom-up budget and 128 [112-155] Tg CH₄ yr⁻¹ in the top-down budget (Table 6). Anthropogenic 2159 emissions dominate in the northern mid-latitudes, with the highest contribution from agriculture and waste emissions (40% 2160 of total emissions in the top-down budget), closely followed by fossil fuel emissions (32% of total emissions, top-down budget). Boreal regions are largely dominated by inland freshwater emissions (41% and 54% of total emissions, top-down 2161 2162 and bottom-up budget, respectively) (Table 6).

The largest discrepancies between the top-down and the bottom-up budgets are found in the mid-latitudes and boreal regions from the natural and indirect sources with bottom-up estimates twice as large as the top-down ones, especially in the inland freshwater category.

The uncertainty for wetlands and inland freshwater emissions is larger in the bottom-up models than in the top-down models (mostly wetlands), while uncertainty in anthropogenic emissions is larger in the top-down models than in the bottom-up inventories. The large uncertainty in tropical inland freshwater emissions (mostly wetlands) of $\pm 44\%$ results from large regional differences between the bottom-up land-surface models. Although they are using the same forcings, their responses in terms of flux density show different sensitivities to temperature, water vapour pressure, precipitation, and radiation.

2171 5.2.3 Regional budget for total emissions

The regional breakdown of emissions is provided for 18 continental regions (see map in Fig. S3 and Table S1 with the country aggregation in the supplementary materials).

At the regional scale and, for the 2010-2019 decade <u>(Table 7)</u>, total methane emissions are dominated by South East Asia with 63 [52-71] Tg CH₄ yr⁻¹, China with 57 [37-72] Tg CH₄ yr⁻¹, and South Asia with 52 [43-60] Tg CH₄ yr⁻¹ (top-down budget). These top three emitters contribute 30% of total global CH₄ emissions. The following high emitting regions are Brazil 47 [41-58] Tg CH₄ yr⁻¹, Equatorial Africa 47 [39-59] Tg CH₄ yr⁻¹, USA 38 [32-46] Tg CH₄ yr⁻¹, Southwest South America 38 [30-48] Tg CH₄ yr⁻¹, Russia 36 [27-45] Tg CH₄ yr⁻¹, Europe 31 [24-36] Tg CH₄ yr⁻¹, Middle East 31 [24-39] Tg

2179 CH₄ yr⁻¹, Northern Africa 25 [23-29] Tg CH₄ yr⁻¹, and Canada 20 [17-24] Tg CH₄ yr⁻¹. Other regions contribute less than 2180 20 Tg CH₄ yr⁻¹.

2181 5.2.4 Regional budget per source category

2182 Natural and indirect anthropogenic emissions versus direct anthropogenic emissions. In agreement with Stavert et al. 2183 (2021), natural and indirect anthropogenic emissions are dominated by Brazil, Canada, Russia, Equatorial Africa and Southeast Asia, contributing 126 Tg CH4 yr⁻¹ in the bottom-up and 105 Tg CH4 yr⁻¹ in the top-down budget (Table 7), i.e., 2184 47% and 50% of the global natural and indirect anthropogenic emissions in these budgets, respectively. At regional scale 2185 2186 also, the range of uncertainty in natural and indirect anthropogenic emissions are much larger in the bottom-up budget than 2187 in the top-down budget (Fig. S5). Except for 4 regions (Canada, Brazil, Northern South America, Southwest South America), 2188 direct anthropogenic emissions contribute more than half of the total regional emissions. Due to the large uncertainty and discrepancies in natural and indirect emissions estimates, the regional direct anthropogenic fractions may differ between the 2189 2190 bottom-up and top-down budgets. However, in absolute values, the highest direct anthropogenic emitters are the same in 2191 the two budgets with China and South Asia being the top two by far, contributing 56 [51-66] Tg CH₄ yr⁻¹ and 45 [44-47] Tg 2192 CH4 yr⁻¹, respectively (bottom-up values, Fig. 9 and Table 7). These two regions contribute 28% (26%) of the global direct 2193 anthropogenic emissions in the bottom-up (top-down) budget. The ranks of direct anthropogenic emitters are similar to those 2194 presented in the last budget (Stavert et al., 2021). Southeast Asia, United States of America, Middle East, Europe, Equatorial 2195 Africa, and Russia emit between 32 Tg CH4 yr⁻¹ and 23 Tg CH4 yr⁻¹ as direct anthropogenic emissions (bottom-up values, 2196 Fig 8). Brazil, Northern Africa, and Southwest South America emit between 10 CH₄ yr⁻¹ and 20 CH₄ yr⁻¹, while the rest of 2197 the regions emit less than 10 CH4 yr⁻¹ direct anthropogenic emissions (Table 7 and Fig. S5).

2198

Sectoral emissions. The sectoral partitioning at the regional scale has been derived from both bottom-up and top-down approaches. However, the top-down budget has more limitations, as the sectoral partitioning is usually based on the prior fluxes fractions at the pixel scale, and assimilating only total methane observations does not allow to disentangle the different source sectors overlapping in a pixel grid. However, differences in spatial patterns and seasonality of emissions can still be constrained by atmospheric CH4 observations for those inversions solving for different sources categories (see Sect. 2.3).

Bottom-up approaches allow deeper sectorial splitting, especially in terms of direct anthropogenic emissions (Fig. 9). Table
7, Fig. 9 and Fig. 10 present the estimations of CH4 emissions on average over 2010-2019. Fig. 10 presents the budgets for
three main categories (Combined wetland and inland freshwaters, Fossil fuels and Agriculture & Waste), a more detailed
figure and table including the five categories is available in the supplementary material (Fig. S6 and Table S13 to S18).
Values for each individual data-set for the decades 2000-2009, 2010-2019, and the last year 2020 are made available in a

2209 spreadsheet (see Data Availability).

2210 For most regions, "Combined wetland and inland freshwater emissions" are the most uncertain in the bottom-up budget, 2211 and generally their range is larger than in the top-down budget. In the top-down budget for 2010-2019 (Table 7), this category 2212 contributes the most to the regional emissions in Brazil 24 [20-33] Tg CH₄ yr⁻¹, Southeast Asia 24 [14-29] Tg CH₄ yr⁻¹ 2213 (though similar to their Agriculture and Waste emissions 24 [21-31] Tg CH4 yr⁻¹), Equatorial Africa 22 [19-28] Tg CH4 yr⁻¹ 2214 ¹, Southwest South America 22 [14-33] Tg CH₄ yr⁻¹, Canada 12 [9-18] Tg CH₄ yr⁻¹, Northern South America 8 [6-10] Tg CH4 yr⁻¹, Southern Africa 7 [4-9] Tg CH4 yr⁻¹. Agriculture and Waste emissions dominates in South Asia 39 [33-43] Tg CH4 2215 yr¹, China 30 [13-37] Tg CH₄ yr¹, Europe 19 [16-23] Tg CH₄ yr¹, United States of America 13 [9-16] Tg CH₄ vr¹, Northern 2216 2217 Africa 13 [12-14] Tg CH4 yr¹, Central America 9 [8-10] Tg CH4 yr¹, and Korea and Japan 3 [3-4] Tg CH4 yr¹. Fossil fuel emissions dominate in the Middle East 18 [11-24] Tg CH4 yr⁻¹ and Russia 14 [8-23] Tg CH4 yr⁻¹ (close to their combined 2218 wetland and inland freshwater emissions of 11 [8-13] Tg CH₄ yr⁻¹). 2219

The four largest contributors to the Fossil Fuel sector remain China, the Middle East, Russia, and the United States of America. Altogether they contribute 67 (64) Tg CH₄ yr⁻¹ in the bottom-up (top-down) budget, around 55% of the global fossil fuel emissions. The bottom-up and top-down approaches generally agree in terms of ensemble mean, except for China for which the top-down estimates suggest lower emissions than the inventories. While Chinese fossil fuel emissions occur mainly through coal mining activity (88%), the Middle East, Russia and the USA extract mainly oil and gas (100%, 80%, 72%).

2226 The three largest contributors to the Agriculture and Waste sector remain South Asia, China, and Southeast Asia. Together 2227 they contribute 88 (92) Tg CH4 yr⁻¹ in the bottom-up (top-down) budget, around 40% of the global agriculture and Waste 2228 sector (Table 7). While the ensemble means tend to agree between bottom-up and top-down budgets, the uncertainty derived 2229 from the top-down approaches is larger, especially for these three regions. CH₄ emissions due to rice cultivation originate 2230 mostly from these same three regions (South East Asia, China and South Asia). Livestock management emissions occurs 2231 mainly in South Asia 20 [18-22] Tg CH₄ yr⁻¹, Brazil 12 [11-13] Tg CH₄ yr⁻¹, China 11 [8-16] Tg CH₄ yr⁻¹, and Europe 11 2232 [10-12] Tg CH₄ yr⁻¹ (bottom-up estimates, Table 7). The United States of America, Equatorial Africa, Northern Africa and 2233 Southwest South America emit between 7 Tg CH4 yr⁻¹ and 10 Tg CH4 yr⁻¹ in this sub-sector. Other regions emit less than 4 2234 Tg CH₄ yr⁻¹ in the livestock management sector. The Waste sector emissions are dominated by three regions: China 11 [6-2235 14] Tg CH₄ yr⁻¹, South Asia 9 [4-11] Tg CH₄ yr⁻¹, and Europe 8 [6-12] Tg CH₄ yr⁻¹ (bottom-up estimates, Table 7). These 2236 three regions contribute around 40% of the global emissions of the Waste sector. It is worth noting that the uncertainty in 2237 the inventory estimates at the regional scale is around 40% (from the min-max range of the estimate, not including the 2238 uncertainty from each inventory).

6 Insights on the methane cycle from 2020-2022 during which there has been unprecedented high growth rates of methane emissions

The mean emissions estimate for the last year of the budget (2020) was 608 [581-627] Tg CH₄ yr⁻¹ (Top-down).) with 65% 2241 2242 of the emissions from direct anthropogenic sources. This is 65 Tg CH₄ vr^{-1} higher (11%) than the mean emissions of the 2243 2000-2009 decade and 6% higher than 2010-2019. In Jackson et al. (2024), we estimated that total methane emissions 2244 increased by around 20% between the early 2000s (2000-2002) and the late 2010s (2018-2020). 2020 was a second highest 2245 year in terms of atmospheric CH4 growth rate (+15.2 ppb/yr) since systematic measurements began in the late 1980s, coming in just behind the highest in 2021 at 17.97 ppb/yr. A few studies analysed the large growth rate increase between 2019 (+9.7 2246 2247 ppb/yr) and 2020 (+15.2 ppb/yr) of +5.4 ppb/yr (corresponding to +14.4 \pm 2.0 Tg CH₄ yr⁻¹) (Peng et al., 2022; Stevenson 2248 et al., 2022). Peng et al. (2022) estimated that the 2019-2020 growth rate change was almost equally due to an increase in wetland emissions $(6.9 \pm 2.1 \text{ Tg CH}_4 \text{ yr}^{-1})$ and a decrease of the OH chemical loss $(7.5 \pm 0.8 \text{ Tg CH}_4 \text{ yr}^{-1})$ due to reduced 2249 OH precursor emissions during the COVID lockdown (Laughner et al., 2021). The COVID19 lockdown resulted in 2250 2251 decreased NO_x emissions and reduced fossil fuel related CH₄ emissions (Thorpe et al., 2023), leading to less OH production. At the global scale, Feng et al. (2023) calculated an emission increase of 27 Tg CH_4 vr⁻¹ between 2019 and 2020 considering 2252 constant OH, and a smaller increase of 21 Tg CH₄ yr⁻¹ when including a 1.4% decrease of OH. Increased emissions were 2253 mainly found in the northern tropics. Qu et al. (2022) also inferred a 31 Tg CH₄ yr⁻¹ increase of emissions, mostly in the 2254 2255 tropics, half of it in Africa. Furthermore, Niwa et al. (2024) suggested emission increases by 10-18 Tg CH4 vr-1 in 15°S-2256 10°N and by 20 Tg CH4 yr-1 in 10-35°N from 2016-2019 to 2020-2022. Such a result is compatible with wetland driven 2257 abnormal emissions during a consecutive 3-year La Nina event spanning from 2020 to 2022 (Zhang et al., 2023; Nisbet et 2258 al., 2023). The difference in terms of methodology and approaches between these three studies make it difficult to compare 2259 them quantitatively but provide a robust understanding on the possible causes. Importantly, all the studies indicate, in various 2260 proportions, increasing CH4 emissions in the tropics and in the boreal region, potentially driven by microbial emission from wetlands due to wetter and warmer climate , and a significant contribution of reduced OH concentrations due to COVID 2261 2262 lockdown. Based on our ensemble of data, we find that top-down approaches infer a much larger change in CH4 emissions (median 2263

[Q1-Q3] at +23 [10-31] Tg CH₄ yr⁻¹) than bottom-up approaches (-1 [-5-3] Tg CH₄ yr⁻¹) between 2019 and 2020 (Fig. S7). Bottom-up approaches suggest a very small increase in wetland emissions (around (+1 [0-3] Tg CH₄ yr⁻¹), while top-down approaches suggest on average a larger increase for wetlands of +8 [5-11] Tg CH₄ yr⁻¹, mainly in the tropics and midlatitudes. It is worth noting that large uncertainties exist for a given year and that the interannual variability is much lower than the ensemble spread. While bottom-up approaches suggest almost constant fossil fuel emissions and slight increase in agriculture and waste (+3 Tg CH₄ yr⁻¹), top-down approaches tend to derive higher emissions changes (+6 Tg CH₄ yr⁻¹ from the fossil fuel sector and +11 Tg CH₄ yr⁻¹ from agriculture and waste as the median over the ensemble). Biomass

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burning emissions decreased using both approaches by about 5 Tg CH₄ yr⁻¹ in agreement with Peng et al. (2022). Some inversions were run with IAV of OH from Patra et al. (2021) and others with constant OH. However, the inferred OH IAV in 2019 and 2020 are rather low (0.3% and 0.15% on yearly average) in Patra et al. (2021), leading to a small impact in terms of emissions changes between 2019-2020, with +22 [9-31] (median [Q1-Q3]) based on the inversions with constant OH and 19 [7-28] based on the inversions with varying OH (Fig S8).

This first analysis based on our ensemble shows how challenging it is to attribute CH₄ emissions changes to a specific sector or region between two years, because related uncertainties remain much larger than the targeted signal to explain. This calls again for further improvement of both approaches.

2280 NOAA estimates of 2021 and 2022 methane atmospheric growth rates 17.8.0±0.5 ppb/yr and 14.0±0.8 ppb/yr, respectively

2281 (Lan et al., 2024). They show a continuation of very high growth rates, challenging again our understanding of the methane

budget. The very high values of CH_e growth rate over 2020-2022 have also been accompanied by a sharp decline in the stable isotopic signal, $\delta^{13}C_{CH4}$, which suggest that this recent increase of methane growth rate is at least partly explained by increased emissions from microbial sources such as those found in wetlands, inland waters, agriculture and waste systems

- $\frac{(\text{Nisbet et al., 2023; Michel et al., 2024). However, it is worth noting that almost all published top-down studies}{\text{aforementioned include constraints only on CH}_{\underline{d}}, and do not discuss the consistency with the atmospheric isotopic signal.}$
- As of the time of submission of this manuscript, bottom-up estimates for anthropogenic emissions for 2021 and 2022 are only available from the EDGARv8 data set (https://edgar.jrc.ec.europa.eu/dataset_ghg80; EDGAR, 2023). This research inventory suggests that anthropogenic emissions continued to increase from 2020 (374 Tg CH₄ yr⁻¹) to 2021 (379 Tg CH₄ yr⁻¹) and 2022 (386 Tg CH₄ yr⁻¹) with around 62% of the increase due to the fossil fuel sources, 23 % from the Waste sector, and 14% from the agriculture sector (Table S19). The bottom-up estimate of wetland emissions for 2021-2023, derived from a single wetland model, indicates positive anomalies of 26 Tg CH₄ yr⁻¹ in 2020, 23 Tg CH₄ yr⁻¹ in 2021, and

21 Tg CH₄ yr⁻¹ 2022 relative to the 2000-2006 baseline (<u>https://earth.gov/ghgcenter/data-catalog/lpjwsl-wetlandch4-grid-</u>
 21 Tg CH₄ yr⁻¹ 2022 relative to the 2000-2006 baseline (<u>https://earth.gov/ghgcenter/data-catalog/lpjwsl-wetlandch4-grid-</u>
 2294 v1; Zhang et al., 2023).

2295 7 Future developments, missing elements, and remaining uncertainties

In this budget, robust features and uncertainties on sources and sinks estimated by bottom-up or top-down approaches have been highlighted as well as discrepancies between the two budgets. Limitations of the different approaches have also been highlighted. Four shortcomings of the CH₄ budget were already identified in Kirschke et al. (2013) and Saunois et al. (2016, 2020) and are revisited below pointing to key research areas. Although much progress has been made, they are still relevant, and actions are needed. However, these actions fall into different timescales and actors. Here, we revisit the four shortcomings of the contemporary methane budget and discuss how each weakness has been addressed since Saunois et al. (2020). Each section ends by discussing remaining research needs with a list of suggestions, from higher to lower priority.

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Shortcoming 1: Towards a decrease of the high uncertainty in the amount of methane emitted by wetland and inland
 water systems, and a weakened double counting issue.

- 2309 This first shortcoming has probably received the largest interest in the last few years with significant improvements. First a community effort has been made based on more studies, documenting, or modelling more inland freshwater systems and 2310 2311 synthesising emissions from the complex and heterogeneous ensemble of emitting areas: wetlands, ponds, lakes, reservoirs, 2312 streams, rivers, estuaries, and marine systems. The range of wetland and inland water emissions has been narrowed down 2313 with improved wetland extent and refined estimates for inland freshwater systems. Double counting between inland 2314 freshwater systems has been estimated for the first time and accounted for in this budget. All these improvements decreased the discrepancy between top-down and bottom-up estimate of combined wetland and inland freshwater emissions from 2315 156 Tg CH₄ yr⁻¹ in Saunois et al. (2020) down to 85 Tg CH₄ yr⁻¹ in this update for the 2000-2009 decade. Gridded maps 2316 for lakes, ponds, reservoirs, and streams and rivers freshwater emissions have been produced over the past years (Johnson 2317
- et al., 2021, 2022; Rocher-Ros et al., 2023) making the spatial distribution of CH₄ sources almost complete for the first time
- and allowing better description of prior emissions in future top-down inversions.
- 2320 Next steps include on the short term from highest to lowest priority include:

- (i) integration of spatial distribution of inland waters in atmospheric inversion models to reach a full description of priormethane sources and sinks.
- 2323 (ii) refinement of double counting estimation and its possible reduction with more precise spatial and temporal distributions
- 2324 of the different systems contributing to inland freshwater emissions by using very high-resolution satellite data (down to
- 2325 metre resolutions) to properly separate them. The development of a dynamical global high-resolution (typically few metres)
- classification of saturated soils and inundated surfaces based on satellite data (visible and microwave), surface inventories,
 and expert knowledge.
- 2328 (iii) continuation of ongoing efforts to calibrate and evaluate land surface models for wetland emissions against in-situ
- observations such as FLUXNET-CH4 (Knox et al., 2019; Delwiche et al., 2021) or BAWLD-CH4 (Kuhn et al., 2021) for
- 2330 boreal regions and avoid dependence on top-down estimates. It is still critical to increase the limited number of tropical
- 2331 observations and to assimilate them in the inverse systems to help address the issue (e.g., Kallingal et al., 2023).
- 2332 (iv) continuation of ongoing efforts to develop a diversity of modelling approaches (among them process-based model or
- 2333 machine learning approaches) to estimate wetland and inland freshwater CH4 emissions, including lateral fluxes, and
- reducing upscaling issues, as done by e.g. Zhuang et al. (2023) for lakes.
- $2335 \qquad (v) \ \text{continuous integration of collected flux measurements such as in the FLUXNET-CH_4 \ activity (Knox et al., 2019;$
- Delwiche et al., 2021) or in BAWLD-CH4 data set (Kuhn et al., 2021) to provide global flux maps based on machine
 learning approaches or other approaches (Peltola et al., 2019, McNicol et al., 2023).
 - 65

2338	Over the long run, developing measurement systems will help to improve estimates of the diversity of wetland and inland
2339	freshwater sources, and further reduce uncertainties:
2340	- More systematic measurements of CH4 fluxes and their isotopic signatures from sites reflecting the diversity of
2341	environment of wetlands and inland waters, complemented with environmental meta-data (e.g., soil temperature
2342	and moisture, vegetation types, water temperature, acidity, nutrient concentrations, NPP, soil carbon density for
2343	wetlands, lake morphologies) will allow us to better understand and estimate the processes of production and
2344	transport to the atmosphere (diffusive, ebullitive, plants mediated) and to better constrain methane fluxes and
2345	their isotopic signatures in the different modelling approaches (Glagolev et al., 2011; Turetsky et al., 2014).
2346	
2347	2. Shortcoming 2: Towards a better assessment of uncertainties for global methane sinks in top-down and bottom-up
2348	budgets.
2349	The inverse systems used here have similar caveats than those described in Saunois et al. (2016, 2020) (same OH field, same
2350	kind of proxy method to optimise it) leading to quite constrained atmospheric sink and therefore total global CH4 sources.
2351	Although we have used the latest release of CCMI-2022 (Plummer et al., 2021) and CMIP6 simulations (Collins et al.,
2352	2017), the uncertainty of derived CH4 chemical loss from the chemistry climate models remains at the same (large) level
2353	compared to the previous intercomparison project ACCMIP (Lamarque et al., 2013). The causes of uncertainties on the
2354	CH4 loss and the differences between the different OH fields derived from Chemistry Transport Models (CTM) and Climate
2355	Chemistry Models (CCM) have been widely discussed (e.g., Nicely et al., 2017; Zhao et al., 2019, 2020a). These results
2356	emphasise the need to first assess, and then improve, atmospheric transport and chemistry models, especially vertically, and
2357	to integrate robust representation of OH fields in atmospheric models. Recently, numerous efforts based on satellite data
2358	have been made to constrain OH distribution, variability and trends Ge.g. Anderson, 2023, 2024; Pimlott et al. 2022; Zhao
2359	et al., 2023; Zhu et al., 2022). Finally, soil uptake estimates rely on very few studies, and interannual variations remain
2360	underconstrained.
2361	Next steps, in the short term, could include developments by the modelling community in:
2362	- Estimating the soil uptake with different land surface models (creating an ensemble) and discussing its variations
2363	over the past decade.
2364	- Assessing the impact of using updated and varying soil uptake estimates, especially considering a warmer climate
2365	in the top-down approach. Indeed, for top-down models resolving for the net flux of CH4 at the surface integrating
2366	a larger estimate of soil uptake would allow larger emissions, and then reduce the uncertainty with the bottom-up
2367	estimates of total CH ₄ sources.

Further studying the reactivity of the air parcels in the chemistry climate models and defining new diagnostics to
 assess modelled CH₄ lifetimes such as in Prather et al. (2023),

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Supprimé: the latter, Zhao et al. (2023) have proposed a new approach based on OH precursor observations and a chemical box model to improve the 3D distributions of tropospheric OH radicals obtained from atmospheric chemistry models.

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2376	-	Developing benchmarking of CTM and CCM regarding simulated OH distribution and variability (as in Zhao et
2377		al. (2019) for example) to increase efforts to assess biases and improve atmospheric chemical schemes in CTM and
2378		<u>CCM.</u>
2379	-	Developing methods to better constrain, OH. Numerous have been proposed: satellite CH4 observations (Zhang et
2380		al., 2018; Anderson et al., 2023; 2024) could afford this but strategy is needed (see Duncan et al., 2024 and
2381		references therein); using halogenated compounds beyond methyl chloroform (MCF), such as done in box models
2382		(Thompson et al., 2024) to derive a 3D dynamical OH, Such methods should be able to reach very low uncertainty
2383		for OH burden and trends (<2%) in order to really better constrain the CH ₄ budget. Duncan et al. (2024) discuss
2384		the existing satellite-based methods and propose a strategy to constrain OH from space-based approaches.
2385	-	Integrating the aforementioned different potential OH chemical fields, including also interannual variability, to
2386		assess the impact on the methane budget following Zhao et al. (2020).

2387 Over the long run, other parameters should be (better) integrated into top-down approaches, among them:

2392

- The magnitude of the CH₄ loss through oxidation by tropospheric Cl, a process debated in the recent literature.
 More modelling (e.g., Thanwerdas et al., 2022b) and instrumental studies should be devoted to reducing the
 uncertainty of this potential additional sink before integrating it in top-down models. This would be especially
 critical if inversions using ¹³C-CH4 observations are included in GMB in the future.
- Shortcoming 3: Towards a better partitioning of methane sources and sinks by region and process using top-down
 models

In this work, we report inversions assimilating satellite data from GOSAT, which bring more constraints than provided by surface stations alone, especially over tropical continents. However, we still found that satellite- and surface-based inversions, and the different inversion systems do not consistently infer the same regional flux distribution.

The estimates contributing to the Global Methane budget are further used in more specific studies focusing on the comparison of the estimates from bottom-up and top-down approaches at national (Deng et al., 2022) and regional scales, including efforts from the GCP-REgional Carbon Cycle Assessment and Processes (RECCAP2) (Petrescu et al., 2021; 2023; Tibrewal et al., 2024; Lauerwald et al., 2023b; and other RECCAP-2 publications to come, see https://www.globalcarbonproject.org/reccap/publications.htm).

- 2403 Next steps, in the short term, could integrate developments to be made by the top-down community:
- Including GOSAT 2 retrievals (Noël et al., 2022; Imasu et al., 2023) for the GOSAT-based inversions and
- 2405 considering TROPOMI-based inversions (as done in Tsuruta et al. (2023), Shen et al. (2023), Chen et al. (2022)
- 2406 Qu et al. (2021) or Yu et al. (2023)) in the next releases once at least 8 years of data are available to provide a
- 2407 decadal estimate and biases are reduced for global scale use (Lorente et al., 2023; Balasu et al., 2023). Indeed,

Supprimé: strategy Supprimé: constrained Supprimé: Applying Zhao et al. (2023) recipe to several CTM used for top-down inversions in order to increase consistency between source and sink estimates in individual approaches. Supprimé: ¶ Supprimé: Developing 3D inverse methods to optimise OH using CH4 satellite data (Zhang et al., 2018) or Supprimé: field or machine learning methods using satellite data to constrain OH (Anderson et al., 2023) Supprimé: . Mis en forme : Indice Supprimé: -

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- recent satellite developments have provided higher temporal and spatial resolutions of CH₄ observations in regions
 with poor in-situ measurements (Figure S9, such as TROPOMI observations in North Africa).
- Integrating the newly available updated gridded products for the different natural sources of CH₄ in their prior
 fluxes (e.g. inland freshwaters) to reach a full spatial description of sources and sinks, and to be able to better
 compare the top-down budget with the bottom-up budget.
- Integration of the newly developed 4D variational inversion systems using isotopic species in the top-down budget
 (Basu et al., 2022; Thanwerdas et al., 2024; Drinkwater et al. 2023; Mannisenaho et al., 2023).
- Improving the availability of in-situ data at high temporal resolution for the scientific community, especially ones
 covering poorly documented regions such as China (Liu et al., 2021b; Guo et al., 2020), India (Nomura et al., 2021;
 Lin et al., 2015; Tiwari and Kumar, 2012) and Siberia (Sasakawa et al., 2010, 2017; Fujita et al., 2020; Winderlich
- 2433 et al., 2010), which are not delivered so far to international databases, or only at poor temporal resolution.
- Integrating the information from imagery satellites (e.g., TROPOMI, Carbon Mapper, Methane Sat, GHG Sat.) of
 high to super-emitters to improve prior fluxes of anthropogenic emissions in terms of quantity and locations for
 each covered sector.
- 2437 Over the long run, integrating more measurements and regional studies will help to improve the top-down systems, and 2438 further reduce the uncertainties:
- Extending the CH4 surface networks to poorly observed regions (e.g., Tropics, China, India, high latitudes) and to
 the vertical dimension: aircraft regular measurements (e.g., Filges et al., 2015; Brenninkmeijer et al., 2007; Paris
 et al., 2010; Sweeney et al., 2015); Aircore campaigns (e.g., Andersen et al., 2018; Membrive et al., 2017); TCCON
 observations (e.g., Wunch et al., 2011, 2019) remains critical to complement satellite data that do not observe well
 in cloudy regions and at high latitudes, and also to evaluate and eventually correct satellite biases (Buchwitz et al.,
 2016).
- Extending and developing continuous isotopic measurements of CH₄ to help partitioning methane sources and to
 be integrated in 4D variational isotopic inversions (e.g., Yacovitch et al., 2021).
- Integrating global data from future satellite instruments with intrinsic low-bias, such as active LIDAR techniques
 with MERLIN (Ehret et al., 2017), that are promising to overcome issues of systematic errors (Bousquet et al.,
 2018) and should provide measurements over the Arctic, contrary to the existing and planned passive missions.
- Other co-emitted species such as radiocarbon for fossil/non-fossil emissions (Lassey et al., 2007a, 2007b; Petrenko et al., 2017), CO (e.g., Zheng et al., 2019) for biomass burning emissions, and ethane for fugitive emissions (e.g., Ramsden et al., 2022) could bring additional information for partitioning emissions.
- 2453

2454 4. Shortcoming 4: Towards reducing uncertainties in the modelling of atmospheric transport in the models used in the
 2455 top-down budget

2456 The TRANSCOM experiment synthesised in Patra et al. (2011) showed a large sensitivity of the representation of 2457 atmospheric transport on CH4 abundances in the atmosphere. In particular, the modelled CH4 budget appeared to depend 2458 strongly on the troposphere-stratosphere exchange rate and thus on the model vertical grid structure and circulation in the 2459 lower stratosphere. Also, regional changes in the CH4 budget depend on the characteristics of the atmospheric transport models used in the inversion (Bruhwiler et al., 2017; Locatelli et al., 2015). This axis of research is demanding important 2460 2461 development from the atmospheric modelling community. Waiting for future improvements (finer horizontal and vertical 2462 resolutions, more accurate physical parameterization, increase in computing resources...), assessing atmospheric transport error and the impact on the top-down budget remain crucial and mostly rely on the use of an ensemble of models. 2463 2464 Methodology changes that could be integrated into the next methane budget releases include:

2465 Evaluating more deeply the inversions provided against independent measurements such as aircraft regular campaigns available through for example the CH4 GLOBALVIEWplus v6.0 ObsPack (Schuldt et al., 2023), the 2466 2467 IAGOS data portal (https://iagos.aeris-data.fr/download/), the NIES portal (https://db.cger.nies.go.jp/ged/en/datasetlist/index.html) for CONTRAIL (e.g., Machida et al., 2008) and Siberian 2468 measurements (e.g., Sasakawa et al., 2017), the WDCGG data portal (https://gaw.kishou.go.jp/) for additional 2469 flights over three other Japanese airports and Orléans, France ; Aircore campaigns data set can be downloaded 2470 2471 through the NOAA Global Monitoring Laboratory website (https://gml.noaa.gov/ccgg/arc/?id=144, Baier et al., 2472 2021) and the French AIrCore Program for atmospheric sampling (https://aircore.aeris-data.fr, Membrive et al., 2473 2017); TCCON observations (https://tccondata.org; e.g., Wunch et al., 2011, 2019), and use this evaluation to weight the different models used in the CH4 budget. 2474

2475 Next steps, in the short term, could include some development to be addressed by the top-down community to reduce 2476 atmospheric transport errors:

- 2477 Developing further methodologies to extract stratospheric partial column abundances from observations such as
 2478 TCCON data (Saad et al., 2014; Wang et al., 2014), Aircore (e.g. Andersen et al., 2018; Membrive et al., 2017) or,
 2479 ACE-FTS (De Mazière et al., 2018) or MIPAS (Glatthor et al., 2023) satellite data.
- Combining SWIR and TIR measurements from space to better constrain the tropospheric column, from TROPOMI
 and IASI for example in the MethanePlus ESA project (<u>https://methaneplus.eu/#docs</u>, Buchwitz etal., 2023) or
 GOSAT (Kuze et al., 2020).
- Porting transport models codes to run on Graphics processing Units (GPU) to achieve sub-degrees resolution global
 inversions (Chevallier et al., 2023).
- In the long run, developments within the dynamical core of the atmospheric transport models through the implementation

phase of hexagonal-icosaedric grid with finer resolution (Dubos et al., 2015; Niwa et al., 2017, 2022; Lloret et al., 2023), and

- improvements in the simulated boundary layer dynamics or troposphere-stratosphere exchanges are promising to reduce
- 2488 atmospheric transport errors.

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2491 8 Conclusions

2492 We have built an updated global methane budget by using and synthesising a large ensemble of published methods and new 2493 results using a consistent, transparent, and traceable approach, including atmospheric observations and inversions (top-down 2494 models), process-based models for land surface emissions and atmospheric chemistry, and inventories of anthropogenic 2495 emissions (bottom-up models and inventories). For the 2010-2019 decade, global CH₄ emissions are 575 Tg CH₄ yr¹ (range of 553-586 Tg CH₄ yr¹), as estimated by top-down inversions. About 65% of global emissions are anthropogenic (range of 2496 2497 63-68%). Bottom-up models and inventories suggest larger global emissions (669 Tg CH₄ yr⁻¹ [512-849]) mostly because 2498 of larger and more uncertain natural emissions from inland freshwater systems, natural wetlands, and geological seepage, 2499 and likely some unresolved double counting of these sources. It is also likely that some of the individual bottom-up emission 2500 estimates are too high, leading to larger global emissions from the bottom-up approach than the atmospheric constraints 2501 suggest. However, the important progress in this update is that for the first time, the bottom-up and top-down budgets agree 2502 within their uncertainty ranges. This is substantial progress toward defining more accurate global methane emissions. 2503 The latitudinal breakdown inferred from the top-down approach reveals a dominant role of tropical emissions (~64%) 2504 compared to mid (~32%) and high (~4%) northern latitudes (above 60°N) emissions. 2505 Our results, including an extended set of atmospheric inversions, are compared with the previous budget syntheses of 2506 Kirschke et al. (2013) and Saunois et al. (2016; 2020). They show overall good consistency when comparing the same 2507 decade (2000-2009) at the global and latitudinal scales. The magnitude and uncertainty of most natural or indirect 2508 anthropogenic sources have been revised and updated. In particular, this new budget benefits from large efforts and collaborations from the research community to provide improved estimates of the magnitude and uncertainty of the different 2509 2510 freshwater sources and helps reduce the potential double counting at the global scale. Of note, newly available gridded 2511 datasets for lakes, ponds, reservoirs, streams, and rivers allow building latitudinal and regional estimates for all these sources 2512 for the first time in these estimates. In the next review, we hope to be able to reduce uncertainties in emissions from inland 2513 freshwater systems by better quantifying the emission factors of each contributing sub-systems (streams, rivers, lakes, 2514 ponds) and estimating double counting at regional scale or avoiding double counting by better defining the surface areas of 2515 each ecosystem. Another important priority for improvements is the uncertainty on the chemical loss of CH4 which still 2516 needs to be better assessed in both the top-down and the bottom-up budgets. Building on the improvement of the points 2517 detailed in Sect. 7, our aim is to update this budget synthesis as a living review paper regularly (~every three or four years). 2518 Each update will produce a more recent decadal CH4 budget, highlight changes in emissions and trends, and incorporate 2519 newly available data and model improvements.

2520

It is still under debate why exactly there is a sustained increase of atmospheric CH₄ (more than +5 ppb yr^{-1}) since 2007 (Nisbet et al., 2019; Nisbet et al., 2023; Turner et al., 2019). Some likely explanations, already introduced by Saunois et al.

(1000 ct da., 200) <u>(1000 ct da., 2002</u>, rando ct da., 200) Some inter studies, include, include of stations et al. (2017) and further investigated by Jackson et al. (2020; 2024) and other studies, include, by decreasing order of certainty: Supprimé: leaks

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2526 1) a positive contribution from microbial and fossil sources (e.g., Nisbet et al., 2019; Nisbet et al., 2023; Schwietzke et al., 2527 2016; Jackson et al., 2020), a negative contribution from biomass burning emissions before 2014 (Giglio et al., 2013; 2528 Worden et al., 2017); 2) a negligible role of Arctic emission changes (e.g., Nisbet et al., 2019; Saunois et al., 2017); and 3) 2529 a tropical dominance of the increasing emissions (e.g., Saunois et al., 2017; Jackson et al., 2020; Wilson et al., 2021; 2530 Drinkwater et al., 2023). Although the accelerated atmospheric methane growth rate in 2020 (15.2 ppb/yr) has found some 2531 explanation with the impact of the world Pandemia in 2020, the sustained observed growth rates in 2021 (17.8 ppb/yr) and 2532 2022 (14 ppb/yr) still challenge our understanding of the global methane cycle. While in Jackson et al. (2020; 2024), the 2533 increase in CH4 emissions over the last two decades is almost attributed entirely to direct anthropogenic emissions, the 2534 uncertainty range from the GMB ensemble is large, and the contribution from natural emissions (wetlands) is still largely 2535 uncertain. Besides the decadal change in CH4 emissions, large interannual variability can occur from these natural emissions. 2536 The recent high record of CH4 growth rate highlights the potential of large variations from natural emissions from one year 2537 to another, in particular wetland emissions (e.g., Peng et al., 2022; Feng et al., 2023). These remain the challenges to be 2538 overcome in better quantifying global methane emissions. 2539 Further investigation is needed in follow-up studies to (1) compare these results to the official UNFCCC declarations and 2540 to important assessment (as those of IEA) as done previously for example in Deng et al. (2022; 2024) or more specifically 541 for fossil fuel emissions in Tibrewal et al. (2024) and (2) further discuss the trend and interannual variability of CH4 sources 542 and sinks at sectoral and regional scales as in Jackson et al. (2020, 2024), Stavert et al. (2021) or RECCAP-2 related 543 publications (e.g., Petrescu et al., 2021; 2023; Lauerwald et al., 2023b), and discuss the compatibility of the budget against 2544 the atmospheric isotopic signal such as in Saunois et al. (2017). The next budgets will be critical to assess whether the Global 2545 Methane Pledge is successful and assess methane mitigation efforts. 2546 The GCP will continue to support and coordinate the development of improved flux estimates for all budget components 2547 and new underlying science to support improved modelling, acquisition of observations, and data integration. At regular 2548 intervals (3-4 years), we will continue to bring all flux components together to produce an improved and updated global

9 Data availability

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2551 The data presented here are made available in the belief that their dissemination will lead to greater understanding and new

CH4 budget, and provide a global benchmark for other CH4 products and assessments.

- scientific insights on the methane budget and changes to it, and help to reduce its uncertainties. For research projects, if the
- data used are essential to the work to be published, or if the conclusion or results largely depend on the data, co-authorship
- should be considered. Full contact details and information on how to cite the data are given in the accompanying database.

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The accompanying database includes a netcdf file defining the regions used, an archive with the maps of prior fluxes used in the top-down activity, an archive with data corresponding to Fig. 3 and 5, and one Excel file organised in the following spreadsheets.

2561 The file Global Methane Budget 2000-2020 v1.0.xlsx includes (1) a summary, (2) the methane observed mixing ratio and growth rate from the four global networks (NOAA, AGAGE, CSIRO and UCI), (3) the evolution of global anthropogenic 2562 2563 methane emissions (including biomass burning emissions) used to produce Fig. 2, (4) the global and latitudinal budgets over 2000-2009 based on bottom-up approaches, (5) the global and latitudinal budgets over 2000-2009 based on top-down 2564 approaches, (6) the global and latitudinal budgets over 2010-2019 based on bottom-up approaches, (7) the global and 2565 2566 latitudinal budgets over 2010-2019 based on top-down approaches, (8) the global and latitudinal budgets for year 2020 2567 based on bottom-up approaches, (9) the global and latitudinal budgets for year 2020 based on top-down approaches, and (10) the list of contributors to contact for further information on specific data. 2568

This database is available from ICOS Carbon Portal (https://doi.org/10.18160/GKO9-2RHT, Martinez et al., 2024).

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2571 Author contributions.

- 2572 MS, AM, and JT gathered the bottom-up and top-down data sets and performed the post processing and analysis.
- MS, BP, PB, PeC, and RJ coordinated the global budget. MS, BP, PB, PeC, RJ, PP and PCi contributed to the update of the
 full text and all coauthors appended comments. AM, ED, and XL produced the figures. DJB, NG, PH, AI, AJ, TK, TL, XL,
 KMcD, JMe, JMu, SP, CP, WR, HT, YY, WZ, ZZ, Qing Z, Qiuan Z and Qianlai Z performed surface land model simulations
 to compute wetland emissions. GA, DB, SC, BRD, GE, MAH, GH, MSJ, RL, SN, GRR, JAR, EHS, PRa, PRe, and TSW
 provided data sets useful for natural emission estimates and/or contributed to text on bottom-up natural emissions. LHI, SJS,
 TNF, GRvW, and MC provided anthropogenic data sets and contributed to the text for this section. AM, JT, PP, DBe, RJ,
 YN, AS, AT, and BZ performed atmospheric inversions to compute top-down methane emission estimates for sources and

sinks. EJD, XL, DRB, PBK, JM, RJP, MR, MS, DWo, and YYo are PI of atmospheric observations used in top-down inversions and/or contributed the text describing atmospheric methane observations. FD, MS, and JT contributed to the bottom-up chemical sink section by providing data sets, processing data and/or contributing to the text. FMF provided data for the soil sink.

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2585 **Competing interests.** At least one of the (co-)authors is a member of the editorial board of Earth System Science Data.

2586

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sector at breakuown this dataset was not used in Table 5.							
B-U models and inventories	Contribution	Time period (resolution)	Gridded	References			
CEDS (country based)	Fossil fuels, Agriculture and waste, Biofuel	1970-2019 (yearly)	no	Hoesly et al. (2018)			
CEDS (gridded)*	Fossil fuels, Agriculture and waste, Biofuel	1970-2020 (monthly)	0.5x0.5°	Hoesly et al. (2018) O'Rourke et al (2021)			
EDGARv6	Fossil fuels, Agriculture and waste, Biofuel	1990-2018^ (yearly, monthly for some sectors)	0.1x0.1°	Oreggioni et al. (2021), Crippa et al. (2021)			
EDGARv7	Fossil fuels, Agriculture and waste, Biofuel	1990-2021 (yearly)	0.1x0.1°	Crippa et al. (2023)			
IIASA GAINS v4.0	Fossil fuels, Agriculture and waste, Biofuel	1990-2020 (yearly)	0.5x0.5°	Höglund-Isaksson et al., (2020)			
USEPA	Fossil fuels, Agriculture and waste, Biofuel, Biomass Burning	1990-2030 (10-yr interval, interpolated to yearly)	no	USEPA (2019)			
FAO-CH4	Agriculture, Biomass Burning	1961-2020 1990-2020 (Yearly)	no	Federici et al. (2015) ; Tubiello et al. (2013); Tubiello (2019)			
FINNv2.5	Biomass burning	2002-2020 (daily)	1km resolution	Wiedinmyer et al. (2023)			
GFASv1.3	Biomass burning	2003-2020 (daily)	0.1x0.1°	Kaiser et al. (2012)			
GFEDv4.1s	Biomass burning	1997-2020 (monthly)	0.25x0.25°	Giglio et al. (2013); van der Werf et al (2017)			
QFEDv2.5	Biomass burning	2000-2020 (daily)	0.1x0.1°	Darmenov and da Silva (2015)			

Table 1: Bottom-up (BU) models and inventories for anthropogenic and biomass burning used in this study. *Due to its limited sectoral breakdown this dataset was not used in Table 3.

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Table 2: Biogeochemical models that computed wetland emissions used in this study. Model runs were performed with two climate
inputs, CRU and GSWP3-W5E5. Models were run with prognostic (using their own calculation of wetland areas) and/or diagnostic
(using WAD2M (Zhang et al., 2021b)) wetland surface areas (see Sect 3.2.1).
(using wAD2wi (Zhang et al., 20210)) weuland surface areas (see Sect 3.2.1).

Model	Institution	Prognostic		Diagnostic		References
		CRU	GSWP3-W5E5	CRU	GSWP3-W5E5	
CH4MOD _{wetland}	Institute of Atmospheric Physics, CAS	n	n	у	у	Li et al. (2010)
CLASSIC	Environment and Climate Change Canada	у	y*	у	y*	Arora et al. (2018); Melton and Arora (2016)
DLEM	Boston College	у	у	У	у	Tian et al. (2015, 2023)
ELM-ECA	Lawrence Berkeley National Laboratory	у	у	у	у	Riley et al. (2011)
ISAM	University of Illinois, Urbana- Champaign	у	у	у	у	Shu et al. (2020) Xu et al. (2021)
JSBACH	MPI	у	у	у	у	Kleinen et al. (2020, 2021, 2023)
JULES	UKMO	у	у	у	у	Gedney et al. (2019)
LPJ-GUESS	Lund University	n	n	у	у	McGuire et al. (2012)
LPJ-MPI	MPI	у	у	у	у	Kleinen et al. (2012)
LPJ-WSL	NASA GSFC	у	у	у	у	Zhang et al. (2016)
LPX-Bern	University of Bern	у	у	у	у	Spahni et al. (2011), Stocker et al. (2014)
ORCHIDEE	LSCE	у	у	у	у	Ringeval et al. (2011)

SDGVM	University of Birmingham/ University of Sheffield	у	у	у	у	Beerling & Woodward (2001), Hopcroft et al. (2011, 2020)
TEM-MDM	Purdue University	n	n	у	у	Zhuang et al. (2004)
TRIPLEX-GHG	UQAM	n	n	У	у	Zhu et al. (2014, 2015)
VISIT	NIES	у	У	у	У	Ito and Inatomi (2012)

¹4 *CLASSIC uses GSWP3-W5E version 2 that covers the time period till 2016. All other models use GSWP-W5E5 version 3.

Table 3: Global methane emissions by source type in Tg CH₄ yr⁻¹ from Saunois et al. (2020) (left column pair) and from this work using bottom-up and top-down approaches. Because top-down models cannot fully separate individual processes, only five categories of emissions are provided (see text). Uncertainties are reported as [min-max] range of reported studies. The mean, minimum and maximum values are calculated while discarding outliers, for each category of source and sink. As a result, discrepancies may occur when comparing the sum of categories and their corresponding total due to differences in outlier detections. Differences of 1 Tg CH₄ yr⁻¹ in the totals can also occur due to rounding errors. Compared to Saunois et al. (2020), emissions are split between "direct anthropogenic" emissions and "natural and indirect anthropogenic" sources. We also propose an estimate of the double-counting between bottom-up wetland and inland freshwater ecosystems emissions.

		t al. (2020)			This		T				
Period of time	2000-2009			-2009		-2019		020			
Approaches	bottom-up	top-down	bottom-up	top-down	bottom-up	top-down	bottom-up	top-down			
NATURAL & indirect anthropogenic SOURCES											
Combined	306 [229-	180	242	158 [145-	248	165 [145-	251 [171-	175 [151-			
wetlands and	391]	[153-196]	[156-355]	172]	[159-369]	214]	364]	229]			
inland	-										
freshwaters											
Wetlands	147	180	153	158 [145-	159 [119-	165 [145-	161	175 [151-			
	[102-179]	[153-196]	[116-189] (***)	172]	203] (***)	214]	[131-198] (***)	229]			
Inland freshwaters	159		112 [49-		112		112 [49-				
a a	[117-212]		202]		[49-202]		202]				
	[]		1		[==]		1				
Double counting ^b	NA		-23 [-9		-23 [-9		-23 [-9				
			36]		36]		36]				
Other natural	63	35	63	44	63	43	63	44			
sources	[26-94]	[21-47]	[24-93]	[40-46]	[24-93]	[40-46]	[24-93]	[40-47]			
Land sources	50 [17-72]		51 [18-73]								
Geological	38 [13-53]		38 [13-53]								
(onshore)											
Wild animals	2 [1-3]		2 [1-3]								
Termites	9 [3-15]		10 [4-16]								
Wildfires	(**)		(**)								
Permafrost soils (direct)	1 [0-1]		1 [0-1]								
Vegetation	(*)		(*)								
Coastal and	13 [9-22]		12 [6-20]								
Oceanic sources ^e	15[9-22]		12 [0-20]								
Biogenic	6 [4-10]		5 [3-10]								
Geological	7 [5-12]		7 [5-12]								
(offshore)											
TOTAL											
NATURAL &	369	215	305	204 [189-	311	206 [188-	314	216 [193-			
INDIRECT	[245-485]	[176-243]	[180-448]	223]	[183-462]	225]	[195-457]	241]			
SOURCES											
		-			<u>IC SOUR</u>						
Agriculture and	192	202	194	210	211	228	211	245			
waste	[178-206]	[198-219]	[181-208]	[197-223]	[195-231]	[213-242]	[204-216]	[232-259]			
Agriculture	132 [NA]		134 [125- 142]		143 [132- 155]		147 [143- 149]				
Enteric ferm. &	104		142		112		149]				
manure	[93-109]		[100 -110]		[107 -118]		[114 -124]				
Rice cultivation	28 [23-34]		30 [24-34]		32 [25-37]		32 [29-37]				
Landfills and waste	60 [55-63]		61 [52-71]		69 [56-80]		71 [60-84]				
Fossil fuels	110	101	105 [97-	105 [88-	120 [117-	115 [100-	128 [120-	122 [101-			
G 1 · · ·	[94-129]	[71-151]	123]	115]	125]	124]	133]	133]			
Coal mining	32 [24-42]		(****)	I	(****)		(****)	I			

	Saunois e	t al. (2020)			This	work		
Period of time	riod of time 2000-2009		2000-2009		2010-2019		2020	
Oil & Gas Industry Transport	73 [60-85] 2 [0-6] 4 [1-11]		30 [26-32] 65 [63-71] 4 [1-8] 3 [1-8]		40 [37-44] 67 [57-74] 5 [1-9] 2 [1-3]		41 [38-43] 74 [67-80] 5 [1-8] 2 [1-3]	
Biomass & biof. burn. Biomass burning Biofuel burning	31 [26-46] 19 [15-32] 12 [9-14]	29 [23-35]	30 [22-44] 19 [14-29] 11 [8-14]	26 [22-29]	2 [1-3] 28 [21-39] 17 [12-24] 11 [8-14]	27 [26-27]	27 [20-41] 17 [13-27] 10 [7-14]	26 [22-27]
TOTAL DIRECT ANTHROPOGENI C SOURCES	334 ^d [321-358]	332 [312-347]	333 ^d [305-365]	341 [319-355]	358 ^d [329- 387]	369 [350- 391]	372 ^d [345-409]	392 [368-409]
			S	INKS				
Total chemical loss	595	505	585	504 ^e	602 [496-	521°	602 [496-	538°
Tropospheric OH	[489-749] 553 [476-677]	[459-516]	[481-716] 546 [446-663]	[496-511]	747] 563 [462- 663]	[485-532]	747] 563 [462- 663]	[503-554]
Stratospheric loss Tropospheric Cl	31 [12-37] 11 [1-35]		<u>37 [27-51]</u> 6 [1-13]		<u>37 [28-43]</u> 6 [1-13]		<u>37 [28-43]</u> 6 [1-13]	
Soil uptake	30 [11-49]	34 [27-41]	30 [11-49]	34 [34-34]	31 [11-49]	35 [35-35]	31 [11-49]	36 [35-36]
TOTAL SINKS	625 [500-798]	540 [486-556]	615 [492-765]	538 [530- 545] ^e	633 [507-796]	554 [520- 567] ^e	633 [507-796]	575 [566- 589] ^e
		SOUF	RCES – SI	NKS IME	BALANCE	2		
TOTAL SOURCES	703 [566-842]	547 [524-560]	638 [485- 813]	543 [526- 558]	669 [512- 849]	575 [553- 586]	685 [540- 865]	608 [581-627]
TOTAL SINKS	625 [500-798]	540 [486-556]	615 [492-765]	538 [530- 545] ^e	633 [507-796]	554 [550- 567] ^e	633 [507-796]	575 [566- 589]°
IMBALANCE	78	3 [-10-38]	23	5 [-4-13]e	36	21 [19-33]e	52	32 [15-38]
ATMOSPHERIC		5.8		6.1		20.9		41.8

Supprimé: 34	
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Supprimé: 10-51	

 A INDSTITURE
 5.6
 0.1
 20.7
 41.6

 GROWTH '
 [4.9-6.6]'
 [5.2-6.9]'
 [20.1-21.7]'
 [40.7-42.9]'

 (*) uncertain but likely small for upland forest and aerobic emissions, potentially large for forested wetland, but likely included elsewhere

 (**) Wes top reporting this value to avoid potential double counting with satellite-based products of biomass burning (see Sect. 3.1.5)

 (***) Here the numbers are from prognostic runs. To ensure a fair comparison with previous budgets (Saunois et al., 2020), the numbers are 163[117-195] for
 2000-2009 from diagnostic runs with CRU/CRU-JRA-55 climate inputs (see Sect. 3.2.1).

(****) Up to 8 Tg of additional emissions could account for ultra emitters (Lauvaux et al., 2022), as in Tibrewal et al. (2024), that are fully or partly missed in

regular anthropogenic inventories a: Freshwater includes lakes, ponds, reservoirs, streams and rivers, part of it is due to anthropogenic disturbances estimated in Sect.3.2.2 b: The double counting estimate is discussed in Sect. 3.2.2

c: includes flux from hydrates considered at 0 for this study, includes estuaries

d: Total anthropogenic emissions are based on estimates of full anthropogenic inventory and not on the sum of "Agriculture and Waste", "Fossil fuels" and "Biofuel and biomass burning" categories (see Sect. 3.1.2)

f: Some inversions did not provide the chemical sink. These values are derived from a subset of the inversion ensemble. f: Atmospheric growth rates are given in the same unit Tg CH₄ yr¹, based on the conversion factor of 2.75 Tg CH₄ ppb⁻¹ given by Prather et al. (2012) and the atmospheric growth rates provided in the text in ppb yr-1.

33

Model	Institution	Observation used	Time period	Number of inversions	2000- 2009	2010- 2019	2020	References
arbon Tracker- Europe CH ₄	FMI	Surface stations	2000-2020	4	у	у	у	Tsuruta et al. (2017)
LMDz-CIF	LSCE/CE A	Surface stations	2000-2020	4	у	у	у	Thanwerdas et al. (2022a)
LMDz-PYVAR	LSCE/CE A/THU	GOSAT Leicester v9.0	2010-2020	4	n	у	у	Zheng et al. (2018a, 2018b, 2019)
MIROC4-ACTM	JAMSTEC	Surface stations	2000-2020	5	у	у	у	Patra et al. (2018); Chandra et al. (2021)
NISMON-CH4	NIES/MRI	Surface stations	2000-2020	2	у	у	у	Niwa et al. (<u>2022;</u> <u>2024)</u>
NIES-TM- FLEXPART (NTFVAR)	NIES	Surface stations	2000-2020	2	у	у	у	Maksyutov et al. (2020); Wang et al. (2019a)
NIES-TM- FLEXPART (NTFVAR)	NIES	GOSAT NIES L2 v02.95	2010-2020	1	n	у	у	Maksyutov et al. (2020); Wang et al. (2019a)
TM5-CAMS	TNO/VU	Surface stations	2000-2020	1	у	у	у	Segers et al. (2022)
TM5-CAMS	TNO/VU	GOSAT ESA/CCI v2.3.8 (combined with surface observations)	2010-2020	1	n	у	у	Segers et al. (2022)
	L	Total nu	mber of runs	24	18	24	24	

Table 4: Top-down studies used here with their contribution to the decadal and yearly estimates noted. For decadal means, top down studies must provide at least 8 years of data over the decade to contribute to the estimate. Details on each inverse system and inversions are provided in Table 58 to \$11 in the Sunnlementary Material.

.5

Table 5: Global and latitudinal total methane emissions in Tg CH₄ yr⁻¹, as decadal means (2000-2009 and 2010-2019) and for the year 2020 from this work using bottom-up and top-down approaches. Global and latitudinal emissions for 2000-2009 are also compared with Saunois et al. (2016, 2020) for top-down and bottom-up approaches when available. Uncertainties are reported as [min-max] range. The mean, minimum and maximum values are calculated while discarding outliers, for each category of source and sink. As a result, discrepancies may occur when comparing the sum of categories and their corresponding total due to differences in outlier detections. Differences of 1 Tg CH₄ yr⁻¹ in the totals can also occur due to rounding errors. For the latitudinal breakdown, bottom-up anthropogenic estimates are based only on the gridded products (see Table 1). As a result, the total from the latitudinal breakdown (line called "This work (gridded BU products only") is slightly different from the values provided in Table 3 and recalled in the line "This work (all BU products)". <u>BU stands for bottom-up</u>.

Period	2000	-2009	2010	-2019	2020		
Approach	Bottom-up	Top-down	Bottom-up	Top-down	Bottom-up	Top-down	
			Global				
This work (all BU products)	638 [485-813]	543 [526-558]	669 [512-849]	575 [553-586]	685 [540-865]	608 [581-627]	
This work (gridded BU products only)	642 [501-809]		676 [526-845]		691 [565-862]		
S2020	703 [566-842]	547 [524-560]	-	-	-	-	
S2016	719[583-861]	552[535-566]	-	-	-	-	
90°S-30°N							
This work	367 [254-487]	337 [311-361]	388 [275-503]	364 [337-390]	395 [292-521]	386 [353-425]	
S2020	408 [322-532]	346 [320-379]	-	-	-	-	
S2016	-	356 [334-381]	-	-	-	-	
			30°N-60°N				
This work	234 [169-335]	182 [162-197]	250 [184-345]	187 [160-204]	256 [186-356]	197 [170-215]	
S2020	252 [202-342]	178 [159-199]	-	-	-	-	
S2016	-	176[159-195]	-	-	-	-	
			60°N-90°N		•	•	
This work	42 [22-79]	26 [22-33]	38[17-73]	24 [18-29]	39 [17-74]	25 [20-32]	
S2020	42 [28-70]	23 [17- 32]	-	-	-	-	
S2016	-	20 [15-25]	-	-	-	-	

27	
28	Table 6: Latitudinal methane emissions in Tg CH ₄ yr ⁻¹ for the last decade 2010-2019, based on top-down and bottom-up approaches.
29	Uncertainties are reported as [min-max] range of reported studies. The mean, minimum, and maximum values are calculated while
30	discarding outliers, for each category of source and sink. As a result, discrepancies may occur when comparing the sum of categories
31	and their corresponding total due to differences in outlier detections. Differences of 1 Tg CH ₄ yr ⁻¹ in the totals can also occur due to
32	rounding errors. For bottom-up approaches, natural and indirect anthropogenic sources are estimated based on available gridded
33	data sets (see text Sect 5.2). As some emissions are missing gridded products (wild animals, permafrost, and hydrates), discrepancies
34	may occur in terms of totals proposed in Table 3. Bottom-up direct anthropogenic estimates are based only on the gridded products
35	(see Table 1).

Latitudinal band	90°S- 30°N		30°N-60°N		60°-90°N	
Approach	Bottom-up	Top-Down	Bottom-up	Top-Down	Bottom-up	Top-Down
Natural and indirect anthropogenic Sources	178 [95-276]	148 [133-164] 100 [43-188]		42 [36-50]	28 [9-53]	14 [10-21]
Combined wetland and Inland freshwaters	151 [85-234]	128 [112-155]	73 [32-147]	27 [20-42]	24 [9-53]	9 [7-17]
Other natural	27 [11-42]	22 [20-29]	27 [10-41]	19 [16-22]	4 [2-6]	3 [1-5]
Anthropogenic direct sources	210 [180-227]	215 [191-238]	151 [142-157]	144 [121-162]	10 [6-14]	10 [6-16]
Agriculture & Waste	140 [121-150]	150 [135-168]	81 [77-84]	77 [56-88]	1 [1-2]	2 [2-2]
Fossil Fuels	52 [44-65]	46 [36-62]	65 [61-71]	61 [50-69]	7 [4-10]	7 [3-13]
Biomass & biofuel burning	22 [18-30]	19 [16-21]	7 [4-10]	6 [2-7]	1 [0-1]	1 [1-2]
Sum of sources	388 [275-503]	364 [337-390]	250 [184-345]	187 [160-204]	38 [7-73]	24 [18- 29]

38 39

Table 7: Regional methane emissions (regions ranked by continent) in Tg CH₄ yr⁻¹ for the last decade 2010-2019, based on top-down and bottom-up approaches. Uncertainties are reported as [min-max] range of reported studies. Differences of 1 Tg CH₄ yr⁻¹ in the totals can occur due to rounding errors. For bottom-up approaches, natural and indirect anthropogenic sources are estimated based on available gridded data sets (see text Sect 5.2). As some emissions are missing gridded products (wild animals, permafrost, and hydrates), discrepancies may occur in terms of totals proposed in Table 3. Bottom-up direct anthropogenic estimates are based on all products (gridded and per country).

Region Total emissions				and indirect genic emissions	Direct anthro	Direct anthropogenic emissions	
	Bottom-up	Top-down	Bottom-up	Top-down	Bottom-up	Top-down	
USA	49 [27-77]	38 [32-46]	24 [7-43]	12 [7-22]	26 [19-34]	25 [16-31]	
Canada	38 [14-71]	20 [17-24]	32 [11-63]	14 [11-22]	6 [3-8]	7[5-9]	
Central America	18 [10-28]	17 [14-19]	8 [3-17]	5 [2-6]	10 [8-12]	12 [11-13]	
Northern South America	19 [9-35]	16 [13-20]	10 [3-17]	9 [7-11]	9 [6-17]	7 [6-8]	
Brazil	51 [26-79]	47 [41-58]	32 [11-57]	26 [22-36]	19 [16-22]	21 [17-26]	
Southwest South America	34 [16-51]	38 [30-48]	21 [6-35]	24 [16-34]	13 [10-16]	14 [12-17]	
Europe	42 [29-57]	31 [24-36]	17 [6-30]	7 [5-9]	25 [22-27]	24 [20-31]	
Northern Africa	24 [18-33]	25 [23-29]	7 [2-13]	6 [6-8]	18 [16-20]	19 [17-21]	
Equatorial Africa	47 [28-83]	47 [39-59]	23 [10-49]	24 [20-30]	24 [19-34]	23 [19-29]	
Southern Africa	21 [5-43]	19 [16-24]	11 [2-29]	8 [7-10]	10 [3-14]	11 [10-12]	
Russia	48 [24-83]	36 [27-45]	25 [9-47]	14 [11-18]	23 [15-36]	21 [14-29]	
Central Asia	15 [6-29]	10 [8-13]	8 [2-19]	1 [0-2]	8 [4-10]	9 [7-11]	
Middle East	35 [21-47]	31 [24-39]	9 [3-15]	4 [1-6]	26 [18-31]	28 [20-34]	
China	71 [55-99]	57 [37-72]	15 [4-33]	4 [3-7]	57 [51-66]	53 [34-66]	
Korean-Japan	6 [4-12]	5 [4-6]	3 [1-7]	1 [1-1]	4 [3-5]	4 [3-5]	
South Asia	58 [49-72]	52 [43-60]	13 [5-25]	6 [5-6]	45 [44-47]	45[37-49]	
Southeast Asia	64 [42-93]	63 [52-71]	32 [19-54]	27 [20-34]	32 [23-39]	35 [31-46]	
Australasia	16 [9-26]	13 [10-17]	10 [4-19]	6 [4-7]	7 [6-7]	7 [6-7]	

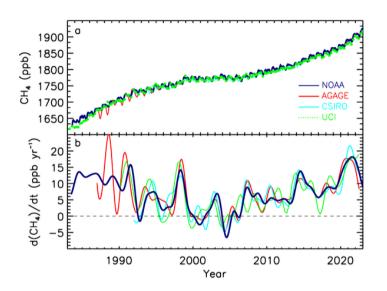
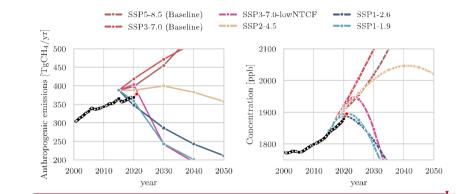


Figure 1: Globally averaged atmospheric CH₄ concentrations (ppb) (a) and annual growth rates G_{ATM} (ppb yr⁻¹) (b) between 1983 and 2022, from four measurement programs, National Oceanic and Atmospheric Administration (NOAA), Advanced Global Atmospheric Gases Experiment (AGAGE), Commonwealth Scientific and Industrial Research Organisation (CSIRO), and University of California, Irvine (UCI). Detailed descriptions of methods are given in the supplementary material of Kirschke et al. (2013).



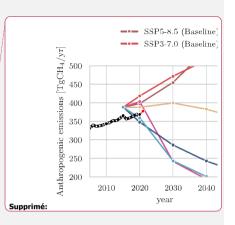


Figure 2: Left: Global anthropogenic methane emissions (including biomass burning) over 2005-2050 from historical inventories (black line and grey shaded area) and future projections (colored lines) (in Tg CH₄ yr⁻¹) from selected scenarios harmonized with historical emissions (CEDS) for CMIP6 activities (Gidden et al., 2019). Historical mean emissions correspond to the average of anthropogenic inventories listed in Table 1 added to the GFEDv4.1s (van der Werf et al., 2017) biomass burning historical emissions. Right: Global atmospheric methane concentrations for NOAA surface site observations (black) and projections based on SSPs (Riahi et al., 2017) with concentrations estimated using MAGICC (Meinshausen et al., 2017, 2020). Red dots show the last year available (2022 for observations).

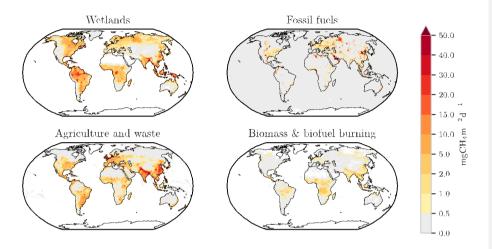


Figure 3: Methane emissions from four source categories: natural wetlands (excluding lakes, ponds, and rivers), biomass and biofuel burning, agriculture and waste, and fossil fuels for the 2010-2019 decade in mg CH₄ m⁻² day⁻¹. The wetland emission map represents the mean daily emission average over the 16 biogeochemical models listed in Table 2 and over the 2010-2019 decade. Fossil fuel and Agriculture and Waste emission maps are derived from the mean estimates of gridded CEDS, EGDARv6, EDGARv7 and GAINS models. The biomass and biofuel burning map results from the mean of the biomass burning inventories listed in Table 1 added to the mean of the biofuel estimate from CEDS (O'Rourke et al., 2021), EDGARv6 (Crippa et al., 2021), EDGARv7 (Crippa et al., 2023) and GAINS (Höglund-Isaksson et al., (2020)) models.

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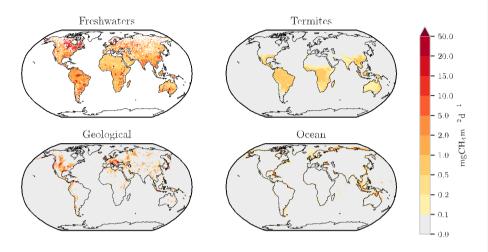


- 81 Figure 4: Estimation of wetland and inland freshwater emissions over the 2010-2019 decade in Tg CH4 yr⁻¹. The
- 82 fluxes related to voluntary (such as through reservoirs or farm ponds) or involuntary (land use or eutrophication-
- 83 related), perturbations of the methane cycle are shown here in pink. They are accounted for into the "natural and 84 indirect anthropogenic" sources in the Table 3 budget and depicted as "natural and indirect anthropogenic" sources
- 85 (darker green and pink hatches) in Fig. 7.





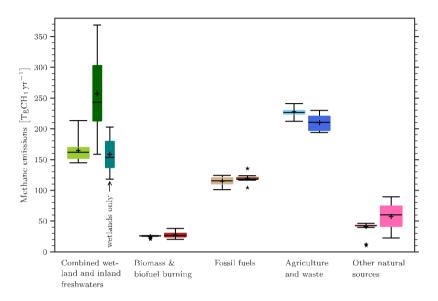




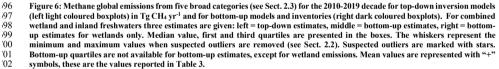
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Figure 5: Methane emissions (mg CH₄ m⁻² day⁻¹) from four natural and indirect anthropogenic sources: inland freshwaters (includes lakes, ponds (Johnson et al., 2022), reservoirs (Johnson et al., 2021) and stream and rivers (Rocher-Ros et al., 2023) with a global total scaled to 89 Tg yr⁻¹), geological (Etiope et al., 2019), termites (this study) and oceans (Weber et al., 2019). 91 i92 i93









'03

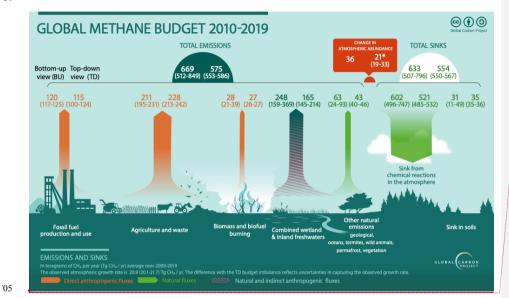
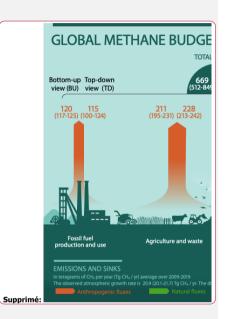


Figure 7: Global Methane Budget for the 2010-2019 decade. Both bottom-up (left) and top-down (right) estimates are provided for each emission and sink category in Tg CH₄ yr⁻¹, as well as for total emissions and total sinks. <u>Combined wettam and inland</u>

freshwaters are depicted as natural and indirect anthropogenic sources (darker green and pink hatches) to recall Figure 4 (Sect.



Supprimé: Biomass and biofuel burning emissions are depicted here as both natural and anthropogenic emissions while they are fully included in anthropogenic emissions in the budget tables and text (Sect. 3.1.5).

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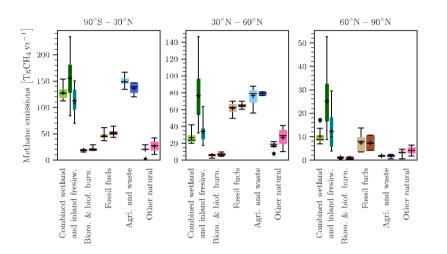
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Figure 8: Methane latitudinal emissions from five broad categories (see Sect. 2.3) for the 2010-2019 decade for top-down inversion models (left light coloured boxplots) in Tg CH4 yr⁻¹ and for bottom-up models and inventories (right dark coloured boxplots). For combined wetland and inland freshwaters three estimates are given: left = top-down estimates, middle = bottom-up estimates for wetlands only. Median value, first and third quartiles are presented in the boxes. The whiskers represent the minimum and maximum values when suspected outliers are removed (see Sect. 2.2). Suspected outliers are marked with stars. Bottom-up quartiles are not available for bottom-up estimates, except wetland emissions. Mean values are represented with "+" symbols, these are the values reported in Table 6.

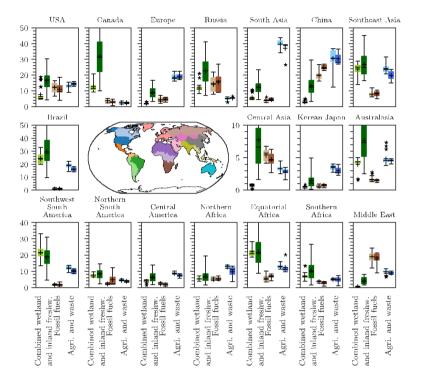
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	Total anthropogenic sources	Enteric ferm. & manure	Rice cultivation	Landfills& waste	Oil & gas	Coal mining	Industry & Transports	Biomass & biofuel burning
China	56.6	11.4	9.1	11.1	2.0	21.5	0.9	2.2
South Asia	43.7	20.1	8.1	8.7	1.3	2.1	0.8	2.7
Southeast Asia	32.0	1.2	9.4	5.9	2.6	3.8	0.7	4.6
Middle East	26.1	3.2	0.2	5.3	15.8	0.2	0.1	0.3
USA	25.9	8.7	0.5	5.0	7.9	2.5	0.8	0.7
Europe	24.9	10.9	0.1	8.3	1.9	1.9	0.5	1.0
Equatorial Africa	23.9	7.4	1.6	3.1	4.9	0.5	1.1	5.1
Russia	22.7	2.0	0.0	3.6	12.1	2.9	1.2	1.7
Brazil	18.7	12.1	0.3	3.5	0.4	0.1	0.3	1.9
Northern Africa	16.3	7.2	0.4	2.9	1.7	0.2	0.1	1.1
Southwest South America	12.9	7.5	0.4	1.8	1.3	0.1	0.1	1.1
Southern Africa	10.1	2.0	0.3	2.3	1.2	1.0	0.3	2.8
Central America	9.7	4.0	0.1	3.1	1.3	0.2	0.2	0.7
Northern South America	9.0	2.6	0.2	1.0	3.4	0.5	0.1	0.3
Central Asia	7.6	2.1	0.0	0.6	3.3	1.0	0.2	0.3
Australasia	6.7	3.7	0.0	0.9	0.3	1.1	0.1	0.7
Canada	5.7	1.2	0.0	1.1	2.3	0.1	0.1	0.7
Korean Japan	3.8	0.9	0.9	1.2	0.1	0.4	0.2	0.1
	0 50	0 20	0 10	0 10	0 20	0 20	0 1	0 5

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'32Figure 9: Regional anthropogenic emissions for the 2010-2019 decade from bottom-up estimates in Tg CH4 yr⁻¹. Regions are ranked'33by their total anthropogenic emissions. Note that each category has its own emission scale.

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'36 '37 '38 '39 Figure 10: Regional emissions for three broad main emissions categories for the 2010-2019 decade: Combined wetland and inland freshwaters, fossil fuel and agriculture & waste from top-down estimates (left box-plots- and bottom-up estimates (right boxplots). The inner map shows the region's distribution (see also Supplementary material, Table S1 and Fig. S3). More categories are

presented in the Supplementary Material in Figure S6.

'40 Table A1. Comparison of terminologies used in this study and previous reports for methane sources.

Table A1. Comparison of	terminologies used in t	his study and previous re	ports for methane sources.	
GCP terminology (This study)		IPCC AR6 (Canadell et al., 2021)	National GHG inventories (used by UNFCCC according to IPCC (2006) and IPCC (2019))	IPCC (2006, 2019) Source sector numbering
Anthropogenic Sources			L	<u>,</u>
	Coal Mining	Coal Mining	Fugitive emissions from Fuels / Solid fuels	1B1
	Oil and gas	Oil and gas	Fugitive emissions from Fuels / Oil and natural gas	1B2
Fossil fuels	Transport	Transport	Transport	1A3
	Industry	Industry	Mineral, chemical, metal industry and others	2A, 2B, 2C, 2D, 2E
	industry		Energy/fuel Combustion activities	1A except 1A3 + 1B3
Agriculture	Enteric fermentation and manure management	Enteric fermentation and manure management	Livestock	3A
	Rice cultivation	Rice cultivation	Rice cultivation	3C7
Waste	Landfills and waste	Landfills and waste	Waste	4
Biofuel and biomass burning	Biofuel burning	Biofuel burning	Biofuel burning	1A4b
	Biomass burning	Biomass burning	Biomass burning	3C1
Natural and indirect so	irces			
Wetlands	Wetlands	Wetlands		
Inland freshwaters	Reservoirs	included in Inland freshwaters	Land (incl Reservoirs)	in 3B
	Lakes, ponds, and rivers	incl in Inland freshwaters	only canal, ditches and ponds for human uses	in 3B
Other natural sources	Oceans	Oceans		
	Termites	Termites		

Geological sources	Geological sources	

'43 '44 '45 '46 Table A2. Summary of methodological changes since the previous budget (Saunois et al., 2020). No significant changes have been applied to the vegetation (Sect. 3.2.8), wild animal (Sect. 3.2.5) and terrestrial permafrost and hydrates (Sect 3.2.7) estimates, though litterature has been expanded and/or updated.

	Saunois et al. (2020)	This study	
Regions definition (Table S1, Fig S3)	18 continental regions + ocean	same regions except the last region including only Australia and New- Zealand and called Australasia	
Anthropogenic global inventories (See Table 1, Sect 3.1.1)	CEDS, EDGARv4.3.2, USEPA (2012), FAO and GAINS ECLIPSE v6	CEDS, EDGARv6 and v7, USEPA (2019), FAO, IIASA GAINS v4 Add estimate of ultra emitters from Lauvaux et al. (2022)	
Biomass burning data sets	FINNv1.5, GFASv1.3, GFEDv4.1s, QFEDv2.5	FINNv2.5, GFASv1.3, GFEDv4.1s, QFEDv2.5	
Estimate of wetland emissions (See Tables 2 and S3 and Section 3.2.1)	13 land surface models involved, runs with either prescribed areas or based on Hydrological scheme, single meteorological forcing	16 land surface models involved, runs with either prescribed areas or based on Hydrological scheme, two sets of meteorological forcings	
Estimate of reservoirs emissions (Sect.3.2.2)	based on Deemer et al. (2016)	based on Johnson et al. (2021), Rosentreter et al. (2021) and Harrison et al. (2021)	Mis en forme : Français
Estimate of lakes and ponds emissions (Sect.3.2.2)	based on Bastviken et al. (2011), Wik et al. (2016b) and Tan and Zhuang (2015)	Jakes > .1km2 : based on Rosentreter et al. (2021), Zhuang et al. (2023) and Johnson et al. (2022) lakes and ponds < 0.1 km2 : based on Rosentreter et al. (2021), and Johnson et al. (2022)	Mis en forme : Français
Estimates of stream and river emissions (Sect.3.2.2)	From Stanley et al. (2016)	based on Rosentreter et al. (2021) and Rocher-Ros et al. (2023)	
Estimates of the anthropogenic perturbation component of inland freshwater emissions (Sect.3.2.2)		based on several individual studies on the effect of eutrophication on emissions from lakes, and ponds (See text in Sect. 3.2.2)	
Estimate of the double counting in the aquatic systems (Sect.3.2.2)		due to the accounting of small lakes and ponds (<0.1km2) in the vegetated wetlands areas used in land surface models and to lateral transport from vegetated wetland to rivers.	
		•	

Geological sources (Sect 3.2.3) - onshore and offshore	based on Etiope and Schwiezke et al. (2019)	same as in Saunois et al. (2020)	
Termite emissions (Sect. 3.2.4)	GPP : Zhang et al. (2017) termite biomass: Jung et al. (2011) EF : Kirshke et al. (2013) and Fraser et al., 1986)	GPP: Wild et al. (2022) termite biomass: based on different studies depending on regions (see text) EF: Sugimoto et al. (1998) Applied a correction factor for mound from Nauer et al. (2018)	Mis en forme : Français
Oceanic sources (Sect 3.2.6)	modern biogenic: based on Wuebbles and Hayhoe (2002), Laruelle et al. (2013) and Rosentreter et al. (2018); geological: based on Etiope (2019)	modern biogenic: based on Rosentreter et al. (2021;2023) and Laruelle et al. (2025) geological: based on Etiope (2019)	Supprimé: Supprimé: 23
Tropospheric OH oxidation (Sect 3.3.2) and stratospheric loss (Sect 3.3.3) (See Supplementary Table S4)	based on results from 11 models contributing to the Chemistry Climate Model Initiative (Morgenstern et al., 2017)	based on results from 11 models contributing to the Chemistry Climate Model Initiative 2022 (Plummer et al., 2021) and the CMIP6 simulations (Collins et al., 2017)	
Tropospheric reaction with Cl	based on Hossaini et al. (2016), Wang et al. (2019b) and Gromov (2018)	based on Hossaini et al (2016), Sherwenn et al. (2016), Wang et al (2019b, 2021b) and Gromov (2018)	Mis en forme : Français
Soil uptake (See Table S6)	based on Tian et al. (2016)	based on VISIT, JSBACH en MeMo surface models.	
Estimates through top-down approaches (See table S7 and S8 to S11)	9 inverse systems contributing, prior fluxes based on EDGARv4.2 or v4.3.2 for most inversions. Most inversion used constant OH.	7 inverse systems contributing, runs with constant and varying OH, prior fluxes based on either EDGARv6 or GAINS	

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'50 '51 '52

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GOSAT retrievals

CSIRO flask network Kennaook/Cape Grim AGAGE,

CSIRO flask network

CSIRO flask network AGAGE calibrations and

American Samoa, and

Kennaook/Cape Grim

CSIRO flask network

Barbados

Mace Head

Cape Matatula

Kennaook/Cape Grim AGAGE, CSIRO flask network

Kennaook/Cape Grim AGAGE

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Trinidad Head, Mace Head, Barbados,

AGAGE station operations at

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GOSAT data. Robert Parker

GOSAT data, Robert Parker

GOSAT data, Robert Parker

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CSIRO flask network

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