Title: Half of global methane emissions come from highly variable aquatic ecosystem sources

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First paragraph:

Atmospheric methane is a potent greenhouse gas that plays a major role in controlling the Earth's climate. The causes of the renewed increase of methane concentration since 2007 are uncertain given the multiple sources and complex biogeochemistry. Here, we present a meta-data analysis of methane fluxes from all major natural, impacted and human-made aquatic ecosystems. Our revised bottom-up global aquatic methane emissions combine diffusive, ebullitive and plant-mediated and/or fluxes from several sediment-water-air interfaces. We emphasize the high variability of methane fluxes within and between aquatic ecosystems and a positively skewed distribution of empirical data, making global estimates sensitive to statistical assumptions and sampling design. We find aquatic ecosystems contribute (median) 41% or (mean) 53% of total global methane emissions from anthropogenic and natural sources. We show that methane emissions increase from natural to impacted aquatic ecosystems, and from coastal to freshwater ecosystems. We argue that aquatic emissions will likely increase due to urbanization, eutrophication and positive climate-feedbacks, and suggest changes in land-use management as potential mitigation strategies to reduce aquatic methane emissions.

Main text:

Methane (CH_4) is the second most important greenhouse gas after carbon dioxide (CO_2), accounting for 16 to 25% of atmospheric warming to date [1,2]. Atmospheric methane nearly tripled since pre-industrial times with a steady rise between 1984 and 2000 (8.4 ± 0.6 ppb yr^{-1}) [3], little or no growth between 2000 and 2006 (0.5 ± 0.5 ppb yr^{-1}) [3], and a renewed growth to present day (2007 to 2020: 7.3 ± 0.6 ppb yr^{-1}) [3–6]. Whether the renewed increase is caused by emissions from anthropogenic or natural sources, or by a decline in the oxidative capacity of the atmosphere, or a combination of all three factors remains unresolved [7–9]. Depending on the approach used, total

methane emissions from natural and anthropogenic sources range between 538-884 Tg CH_4 yr^{-1} [10,11]. However, top-down versus bottom-up estimates of methane sources and sinks do not match, underscoring the incomplete knowledge of global methane dynamics [10,11].

Reducing the uncertainty in methane emission intensities and partitioning emissions to anthropogenic and natural sources is challenging. At the global scale, bottom-up methane emissions from aquatic ecosystems are not well-constrained due to reasons that include the lack of observations, uncertainties associated with surface areas, and the risk of 'double counting' of ecosystem types. In particular, methane emission from small lakes, reservoirs, aquaculture ponds, and coastal wetlands were insufficiently assessed in the IPCC 5th Assessment report [1] and in the most recent global methane budget [11]. Finally, anthropogenic disturbances such as dam construction [12], eutrophication [13], and wetland modification [14], along with climate-feedbacks such as microbial responses to warming [15] and changes in hydrology [16,17] , all lead to an alteration of methane fluxes that are currently difficult to account for at the global scale. A better understanding of the aquatic contribution to global methane emissions is therefore critical to a more robust understanding of atmospheric methane dynamics.

Global aquatic methane emissions

Here we present a meta-data analysis of aquatic methane flux measurements based on inventory, remote sensing and modeling efforts to revise bottom-up estimates of areal methane fluxes (mg CH_4 m^{-2} d^{-1}) (Supplementary Table 1) and global methane emissions (Tg CH_4 yr^{-1}) (Table 1) from rivers and streams, lakes and ponds, reservoirs, estuaries, mangroves, saltmarshes, seagrasses, tidal flats, aquaculture ponds, continental shelves, along with recently published estimates of global methane emissions from freshwater wetlands [11], rice paddies [11], continental slope and open ocean [18]. Our global synthesis reveals median (Q1-Q3) methane emissions from aquatic ecosystems of 269 (202-424) Tg CH_4 yr^{-1} or mean (lower-upper C.I.95%) emissions of

431 (343-519) Tg CH_4 yr^{-1}. Our bottom-up estimates show a larger range with a lower (median) or higher (mean) central tendency than the most recent bottom-up estimate for aquatic ecosystems and wetlands [11] (Table 2). The interquartile range (IQR) (222 Tg CH_4 yr^{-1}) of our global aquatic emissions is larger than the confidence interval (176 Tg CH_4 yr^{-1}), which suggests that methane flux variability is larger than uncertainty. The high variability in data sources is linked to the complexity of how methane is produced, transported, and consumed before reaching the atmosphere, with different transport pathways (i.e., diffusion, ebullition, plant-mediated), physical interfaces (water-atmosphere, sediment-atmosphere), ecosystem conditions (impacted versus natural), and temporal (diel/tidal, seasonal, inter-annual) and spatial scales involved. We find that the statistical distributions of our data sets are ecosystem-specific, and that all aquatic ecosystems have positively skewed distributions (Fig. 1), which greatly affects the results for global upscaling (Fig. 2). If the observational data represent the actual flux distribution, then mean values would be the appropriate measure to scale global emissions. However, our assessment cannot rule out substantial bias in the available flux estimates resulting from limited temporal and spatial coverage and non-random selection of study sites. Under such circumstances, median values provide a more conservative estimate for upscaling.

Methane emissions (Q1-Q3) from freshwater wetlands (138-165 Tg CH_4 yr^{-1}) and lakes (23-142 Tg CH_4 yr^{-1}) are the largest aquatic sources, followed by rice cultivation (25-32 Tg CH_4 yr^{-1}), coastal ocean (5-28 Tg CH_4 yr^{-1}, < 200m), reservoir (9-28 Tg CH_4 yr^{-1}), and river and stream emissions (2-21 Tg CH_4 yr^{-1}). While uncertainties for bottom-up (and top-down) global estimates are still high, natural, impacted and human-made aquatic ecosystems including wetlands could be equally important to, or greater than direct anthropogenic emissions [11,19]. Depending on the approach used (median or mean), we find that 41 or 53 % of the global methane emissions can be attributed to aquatic ecosystems, whereas non-aquatic systems contribute the remainder, for example 8 or 6 % to other land sources such as onshore geological, wild animals and termites [11],

and 51 or 41 % to direct anthropogenic activities such as enteric fermentation and manure, landfill and waste, coal mining, gas and oil industry, transport, and biomass and biofuel burning [11] (Table 2).

Our revised global estimates of aquatic ecosystem emissions are mostly higher than previous estimates (Supplementary Table 2). However, the comparison to previous studies is challenging due to the difference in upscaling methods, dissimilar statistical treatment, and uncertainties in surface areas. In brief, our combined lakes, ponds and reservoir emissions are higher than the first mean global estimate for these ecosystems [20], similar to recent estimates based on chlorophyll *a* scaling [21], and lower than recent upscaling from mean values [21] (Supplementary Table 2). The relatively lower emissions we present here are largely the result of an 'ice' correction term, which had not previously been implemented in the computation of global lake and reservoir emissions (Supplementary Table 2). Thus, while our mean annual emissions for lakes, ponds and reservoirs are not higher than recent estimates, mean areal methane fluxes are higher than those recently reported (132 mg CH_4 m^{-2} d^{-1}) [27] (Supplementary Table 2). These higher areal fluxes likely result from our inclusion of recent studies that add smaller waterbodies and whose methods capture ebullitive [22]. The result is a database containing disproportionately more studies from research published since 2015 (205 of 313 lakes or reservoirs; 65%). We find that the smallest lakes are responsible for the largest emission with ~37% of total lake emission coming from lakes <0.001 km^2 regardless of mean or median (Table 1).

Our ice-corrected river and stream emissions are significantly higher than the first reported global mean [1], which used a low surface area, only 21 sites for upscaling, and no data from the tropics. A more recent review [23], using an updated surface area [24] and 385 sites, reported an average diffusive flux that is higher than our global estimate (Supplementary Table 2). Here we increase the number of sites and include ebullitive fluxes to report fluxes from 5 latitudinal bands. Approximately 30% of ice corrected fluxes are from the equatorial latitudes due to the large ice-

free surface area of streams and rivers (Supplementary Table 3). However, the data density of total and ebullitive fluxes are low, particularly for mid to high latitudes.

Our coastal ocean emission estimate is higher than previous mean estimates [11,18,25], which did not include some of the coastal habitats. The large range and uncertainty of coastal methane fluxes that we find in this study are associated with the paucity of data, but also with the high spatial and temporal variability of fluxes in coastal ecosystems driven by e.g. tidal pumping and salinity gradients [26]. More than half of the global coastal ocean emission is attributed to large continental shelf areas, mainly gas seeps (i.e., ebullition) and estuarine plumes (Extended Data Fig.1). However, per area, methane fluxes from continental shelves are much lower compared to those from other coastal ecosystems (Supplementary Table 1). We find particularly high areal methane fluxes from coastal aquaculture ponds that are 7-430 times higher than from non-converted coastal habitats (mangroves, saltmarshes, seagrasses, tidal flats) and nearly 70,000 times higher than from the open ocean [18] (Supplementary Table 1).

Increasing aquatic methane emissions

The renewed increase in atmospheric methane has been attributed to climate-feedbacks on wetlands, increased fossil fuel use, methane production by livestock, and declining removal of methane by tropospheric OH (sinks) [8,9,19]. Our findings complement this picture by highlighting how human alterations of aquatic ecosystems increase methane emissions. The strongest growth in atmospheric methane since 2007 has been reported in the tropics and subtropics (30˚N to 30˚S [8]) with fuel burning and both agricultural and ruminant populations as the major sources [8]. Despite the global coverage of our data, we did not detect clear latitudinal trends of methane emissions from aquatic ecosystems, except for the emissions from coastal wetlands peaking at 30˚N (Extended Data Fig.2). Instead of latitudinal patterns, we found methane emissions increasing from rivers to lakes and wetlands, from natural to impacted and eutrophic ecosystems (Extended Data

Fig.3 and Fig.4), and from coastal marine to freshwater ecosystems (Fig.3). Particularly pronounced is the difference of areal fluxes between aquaculture farms and non-converted coastal and inland water ecosystems (Supplementary Table 1).

As a result of global warming, increased methane emissions are expected across biomes and latitudes because of the higher activities of methanogenic archaea at elevated temperatures relative to methanotrophic microorganisms [27,28]. However, not only archaea but also saprotrophic fungi and cyanobacteria can produced methane under various environmental conditions [29–31]. While the methane flux from these microorganisms is currently poorly constrained, it is intuitive to assume that it increases with increasing eutrophication and temperature. This is indeed supported by a general positive relationship between methane emissions and temperature across biomes [15,27], and the enhanced methane emissions that we found with increasing temperature in coastal wetlands (Extended Data Fig.5). However, the effect of warming and wetting may have synergistic effects on methane emissions from freshwater wetlands. In fact, when freshwater wetlands dry, both the water table level and time of inundation drop, which may foster methane oxidation and thereby reduce emissions [32]. Finally, global warming impacts are predicted to be particularly important at high latitudes [33], therefore, a better understanding of the expected changes in methane emissions from the Arctic, from hydrate deposits in the shallow ocean [34], permafrost soils [35], and melting sea ice [36], for instance, would be required.

Human alterations (e.g. damming, rice cultivation) have increased the surface area of perennial and seasonal freshwater ecosystems by 94,000 km^2 and ~230,000 km^2, respectively, between 1984 and 2014 [17]. This areal expansion of inland waters compounds our finding of areal methane fluxes from aquatic ecosystems. It also indicates that total methane fluxes from aquatic ecosystems will most likely increase due to habitat expansion and/or transformation.

Uncertainties in aquatic methane sources

196 Methane emissions from individual sources are challenging to measure given the large
197 spatial and temporal variation in net emissions from production, consumption, transport pathways,
198 and due to mixing in atmosphere [37]. Ebullition often constitutes a significant, albeit highly variable,
199 fraction of the total aquatic methane flux. While many ecosystems have a large proportion of
200 emissions driven by ebullition (e.g. some lakes and reservoirs), other ecosystems may have
201 negligible ebullition rates (e.g. seagrasses). Furthermore, different physical interfaces need to be
202 considered when estimating whole-ecosystem emissions, in particular in coastal ecosystems,
203 where methane can be released by exposed (sediment-air flux) or inundated (water-air flux)
204 sediments following the tidal cycle [26]. Plant-mediated methane fluxes can be important in aquatic
205 ecosystems dominated by plants, but the relative contribution of plant-mediated and tree fluxes to
206 total emissions is highly uncertain at global scale [38]. It is also likely that there is a bias in site
207 selection, but the direction of this bias is unknown. Very high or very low values can also be related
208 to inadequate sampling methods, incorrect data analysis or reporting.
209 Another challenge lies within the statistical comparison of different upscaling methods.
210 For instance, a recent estimate of global methane emissions from freshwater wetlands was based
211 on an ensemble of 13 biogeochemical models constrained with remote sensing of surface
212 inundation and inventory-based wetland area [39]. These wetland models use standardized climate,
213 atmospheric CO_2, and dynamic wetland area, but to operate at global scales and across multiple
214 wetland types, the models generalize fundamental processes of methane production, oxidation, and
215 transport to the atmosphere. In contrast, here, we upscale data-driven methane fluxes from streams,
216 rivers, lakes, and reservoirs using a Monte Carlo approach, and relationships between methane
217 fluxes and either latitudinal band (streams and rivers) (Supplementary Table 3), size bin (lakes and
218 small reservoirs) or chlorophyll *a* concentration (large reservoirs) (Supplementary Table 4). For
219 coastal ecosystems, where fewer data were available, we multiplied rates by surface area.

There are uncertainties associated with surface areas of aquatic ecosystems and the risk of 'double counting' due to issues in clearly identifying different ecosystem types. In particular, small ponds and lakes, streams and rivers, and coastal wetlands are difficult to separate from freshwater wetlands using coarse-to-moderate spatial resolution optical and radar remote sensing. Recent wetland area mapping aims to reduce the problem of double counting by explicitly removing inland-waters from remote-sensing based surface inundation data [40]. However, there remains a need for finer spatial resolution approaches that would permit better mapping and counting of both small ponds and streams to partition these from vegetated wetlands.

When we combine our median bottom-up aquatic methane emissions with emissions from thermogenic, pyrogenic and other methane producing sectors [11], we find a discrepancy of about 26 Tg CH_4 yr^{-1} compared with bottom-up sinks [11], which is similar to the difference of +20 Tg CH_4 yr^{-1} required to account for the change in atmospheric concentrations since 2007 (7.3 ppb yr^{-1})[3] (Table 2). Our median bottom-up total source estimate exceeds the top-down sinks [11] by 95 Tg CH_4 yr^{-1}, which is close to the source-sink imbalance of 112 Tg CH_4 yr^{-1} reported in the global methane budget [11]. However, our mean bottom-up global source estimate exceeds bottom-up and top-down global sinks by 188-257 Tg CH_4 yr^{-1} [11]. While we are unable to explain such high source-sink imbalances, they are consistent with the large uncertainties related to bottom-up and top-down global sinks [11,19,41]. In particular, global OH concentrations are difficult to measure, and thus atmospheric chemistry models are used to simulate these concentrations, which vary by 10-20% [42,43]. The upland soil methanotrophic sink is equally uncertain and known only via numerical modeling estimates and sparse observations made by soil chambers and flux towers [11,44]. Given these uncertainties, there may be room for large aquatic sources of methane to the atmosphere that we identify in our study.

Aquatic methane management interventions

Reducing methane emissions from aquatic ecosystems is an effective pathway to mitigate climate change, particularly those targeting freshwater wetlands, which account for 35-55% of aquatic emissions (Table 1). Salinities of ~10-15 are an important tipping point for biogeochemical processes in wetlands [45], as sulfate-reducing bacteria, favored by more saline waters, can outcompete methanogens. Hence, converting freshwater wetlands back to saltmarshes by restoring tidal flows is a promising strategy to reduce methane emissions [14], while increasing carbon sequestration. Converting aquaculture ponds and salinized rice paddies back to saltmarsh and mangrove habitats may also achieve order of magnitude reductions in methane fluxes because mangrove and saltmarshes have lower fluxes than aquaculture ponds (Supplementary Information). Reducing nutrient inputs to freshwater wetlands, lakes and reservoirs can help reduce methane emissions [13,21] (Extended Data Fig.4). Reservoirs and constructed ponds can be designed to reduce methane emissions through their placement within the watershed [46] and their depth [47], and in the case of reservoirs, by withdrawing water from depths with lower methane concentrations [48]. In rivers and streams, methane emissions can be reduced if the benthic environments of the streams are restored and organic loadings are decreased [49]. In coastal environments, reduction of eutrophication should lead to significant decrease of methane emissions as suggested by the comparison of fluxes from impacted versus natural sites (Extended Data Fig.3). Thus, land use and management choices may significantly reduce methane emissions and lessen the impact on future atmospheric methane.

Because of their prominent contribution to global emissions, actions to reduce aquatic methane emissions through the management of land use, nutrient and organic inputs, and hydrological flows are a promising avenue to mitigate methane emissions. These actions will be particularly effective when targeting the ecosystems with the greatest contributions to aquatic methane emissions, primarily freshwater wetlands, lakes, reservoirs, and rice paddies. This requires an effort to integrate existing knowledge across disciplines, from the microbial processes

that cycle methane, to the biogeochemical constraints that favor and inhibit these processes, to spatial and hydrological planning and management to create the conditions conducive to the lowest fluxes, while preserving ecosystem function and biodiversity.

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Competing financial interests:
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Author contributions:

J.A.R. did the synthesis for mangroves, saltmarshes, seagrasses and tidal flats, and produced all figures in the main manuscript; A.V.B. did the synthesis for estuaries and continental shelves; A.V.B. and J.A.R. did the synthesis for aquaculture ponds; C.S. compiled the data for streams, rivers, lakes, and reservoirs with help from S.L.; B.R.D. and M.A.H. updated the compiled data for lakes and reservoirs and analyzed the data with input from J.M.; C.S., S.L., G.H.A. and P.A.R. analyzed the data for streams and rivers; G.H.A. determined zonal estimates of river surface area and stream and lake ice corrections; B.D.E. and J.A.R. conceived the project; J.A.R. drafted the first manuscript and all authors reviewed and edited the manuscript and approved the final version.

Figure captions:

Figure 1. Natural log-transformed (*ln*) areal methane fluxes. The violin plots include boxplots showing the median and interquartile range of methane fluxes from streams and rivers, lakes, reservoirs, aquaculture (coastal and freshwater), estuaries, coastal wetlands, tidal flats and continental shelves compiled in this study. All data sets (non-log transformed) are positively skewed (skewness coefficient >1, range 1.1-9.8).

Figure 2. Global aquatic methane emissions in comparison to other methane sources and sinks. Cumulative bottom-up (BU) mean (IQR) and median (± C.I.95%) aquatic methane emissions estimated in this study compared to other BU methane sources versus BU and top-down (TD) methane sinks from Saunois et al. [11] (Table 2). The coastal and open ocean estimate includes emissions from estuaries, saltmarshes, mangroves, seagrasses, tidal flats, coastal aquaculture ponds, continental shelves, slope and the open ocean [18]. Error estimates for freshwater wetland and rice emissions are based on inventory and biogeochemical modelling efforts, therefore show comparably low variability and uncertainty.

Figure 3. Conceptual diagram showing global aquatic methane emissions from headwater streams to the open ocean. Numbers are in Tg CH_4 yr^{-1}. The relative importance of the factors controlling methane distribution and emissions vary along the land-ocean aquatic continuum.

Table 1

Annual methane emissions from aquatic ecosystems. We present median, first (Q1) and third (Q3) quartile, mean and 95% confidence intervals of bottom-up global methane emissions. Although two decimal places imply more accuracy than the methods provide, this was done to avoid losing the emission estimates from ecosystems with < 1 Tg CH_4 yr^{-1}.

Aquatic ecosystem	Bottom-up global methane emission (Tg CH_4 yr^{-1})		Reference
	Median (Q1-Q3)	Mean (± C.I. 95%)	
Rivers (ice-corrected)	5.8 (1.8-21.0)	30.5 ± 17.1	*This study*
Lakes (ice-cover, ice-melt corrected)			
< 0.001 km^2	21.2 (9.1-53.5)	54.5 ± 48.5	*This study*
0.001 – 0.01 km^2	13.2 (5.6-33.1)	31.1 ± 23.7	*This study*
0.01 – 0.1 km^2	4.4 (1.4-16.7)	22.4 ± 18.4	*This study*
0.1 – 1 km^2	3.0 (1.1-8.0)	9.9 ± 7.0	*This study*
> 1 km^2	14.0 (6.0-31.0)	33.0 ± 45.0	*This study*
All lakes	55.8 (23.3-142.3)	150.9 ± 73.0	*This study*
Reservoirs (ice-cover, ice-melt corrected)			
< 1 km^2	0.4 (0.1-1.3)	2.4 ± 4.7	*This study*
> 1 km^2	14.7 (8.7-27.1)	22.0 ± 6.4	*This study*
All reservoirs	15.1 (8.8-28.4)	24.3 ± 8.0	*This study*
Freshwater wetlands	150.1 (138.3-164.6)	148.6 ± 15.2	Saunois et al. [11] (A)
Freshwater aquaculture ponds	4.4 (0.4-7.9)	14.0 ± 18.8	*This study*
Rice cultivation	29.9 (24.9-32.1)	29.8 ± 6.7	Saunois et al. [11] (B)
Total inland waters	**261.0 (197.5-396.2)**	**398.1 ± 79.4**	***This study***
Estuaries	0.23 (0.02-0.91)	0.90 ± 0.29	*This study*
Coastal wetlands			
Saltmarshes	0.18 (0.02-0.89)	2.00 ± 1.51	*This study*
Mangroves	0.21 (0.06-0.77)	1.46 ± 0.91	*This study*
Seagrasses	0.13 (0.07-0.21)	0.18 ± 0.19	*This study*
Tidal flats	0.17 (0.04-2.7)	4.2 ± 4.9	*This study*
Coastal aquaculture ponds	0.62 (0.01-1.0)	5.9 ± 15.1	*This study*
Continental shelves	5.7 (3.6-20.4)	17.2 ± 34.0	*This study*
Slope (200-2000m)	0.30 (0.23-0.40)	0.36 ± 0.93	Weber et al. [18] (C)
Open ocean (> 2000 m)	0.91 (0.75-1.1)	1.0 ± 1.7	Weber et al. [18] (C)
Total coastal and open ocean	**8.4 (4.8-28.4)**	**33.2 ± 37.6**	***This study***
Total aquatic	**269.4 (202.3-424.6)**	**431.3 ± 87.9**	***This study***

(A) Based on 13 biogeochemical models for wetland emissions, bottom-up estimate year 2008-2017
(B) Based on 5 inventory models for rice cultivation, bottom-up estimate year 2008-2017
(C) Based on 2 machine-learning methods; confidence interval is mean of the lower and upper bound of the 95% level

453 **Table 2**
454
455 Bottom-up (BU) global aquatic methane sources compared to other BU natural and
456 anthropogenic methane sources and BU and top-down (TD) methane sinks.
457

	Average methane emissions ($Tg\ CH_4\ yr^{-1}$)	Range of methane emissions ($Tg\ CH_4\ yr^{-1}$)	Period of time	Reference
Aquatic sources				
BU (A)	352	253-455	(2008-2017)	Saunois et al. [11]
BU (median, Q1-Q3)	269	202-424	(1978-2019)	*This study*
BU (mean, lower- upper C.I.95%)	431	343-519	(1978-2019)	*This study*
Non-aquatic sources				
Natural sources (BU)				
Geological (onshore)	38	13-53	(2008-2017)	Saunois et al. [11]
Wild animals	2	1-3	(2008-2017)	Saunois et al. [11]
Termites	9	3-15	(2008-2017)	Saunois et al. [11]
Anthropogenic sources (BU)				
Agriculture and waste				
Enteric fermentation & manure	111	106-116	(2008-2017)	Saunois et al. [11]
Landfills & waste	65	60-69	(2008-2017)	Saunois et al. [11]
Fossil fuels				
Coal mining	42	29-60	(2008-2017)	Saunois et al. [11]
Oil & Gas	79	66-92	(2008-2017)	Saunois et al. [11]
Industry	3	0-7	(2008-2017)	Saunois et al. [11]
Transport	4	1-12	(2008-2017)	Saunois et al. [11]
Biomass & biofuel burning				
Biomass burning	17	14-26	(2008-2017)	Saunois et al. [11]
Biofuel burning	12	10-14	(2008-2017)	Saunois et al. [11]
Total methane sources				
BU (B)	737	594-881	(2008-2017)	Saunois et al. [11]
BU (median, Q1-Q3)	651	505-892	(1978-2019)	*This study*
BU (mean, lower- upper C.I.95%)	813	646-986	(1978-2019)	*This study*
Total methane sinks				
BU	625	500-798	(2008-2017)	Saunois et al. [11]
TD	556	501-574	(2008-2017)	Saunois et al. [11]

458 (A) Includes estimates for wetlands, freshwater, oceanic sources, permafrost and rice cultivation
459 (B) Differences of 3 $Tg\ CH_4\ yr^{-1}$ compared to the sum of aquatic and non-aquatic sources (BU) (2008-2017) of 734
460 $Tg\ CH_4\ yr^{-1}$ in this table are due to rounding errors [11]
461

462

Methods:

We use the term 'natural' to describe less impacted and less disturbed study sites or ecosystems, whereas 'impacted' refers to highly impacted, modified, polluted or eutrophied study sites or ecosystems.

Our datasets were compiled from peer-reviewed publications. Temporal (annual, seasonal, diel) or spatial data were averaged to a single flux per study site. If 'site' was not obvious, we set a criterion of 10 km distance to distinguish between sites. An exception was the river and stream data set, where measurements with the exact same coordinates were treated as one site, and fluxes with different coordinates were treated as many sites to account for variable fluxes of low and high stream orders. Values under detection limit were set to "0" and included in the statistical analysis. Sites were classified as 'natural' or 'impacted' if clearly identified in the literature, or based on the authors' knowledge.

Rivers and streams

We compiled peer reviewed studies until March 2019 using the Boolean search string "(CH_4 OR methane) AND (concentration OR flux OR emission) AND (river OR stream)" in the Web of Science Core Collection (http://isiknowledge.com) and China Knowledge Resource Integrated database (https://www.cnki.net/). In our river and stream database, we only included georeferenced methane concentrations or fluxes. If exact coordinates were not provided but site description was sufficient, we obtained approximate coordinates from Google Maps. We excluded non-river data by either referring to the original site descriptions or by overlying the measurement locations with a map of global open inland waters [50].

Our efforts identified a total of 2,601 records with either a methane concentration or a flux measurement. Our primary analysis showed that methane fluxes calculated from concentration versus gas transfer velocity (k) poorly predicted the literature reported fluxes. Therefore, we only

included reported methane fluxes from publications. This resulted in a collection of 652 methane fluxes from 74 publications including one unpublished data set (provided by T.I.B). The total number of records included 187 total fluxes, 590 diffusive fluxes and 126 ebullitive methane fluxes. We refer to the total methane flux as either the sum of diffusive and ebullitive fluxes or the total flux without differentiation between diffusion and ebullition. For example, a properly designed chamber can catch both diffusive and ebullitive methane fluxes [51,52]. If the original studies clearly identify chamber fluxes as diffusive + ebullitive fluxes, we included these as total fluxes. If the original studies identified chamber fluxes as diffusive (e.g., because of low observed ebullition) or reported calculated fluxes from concentration and *k*, we included these as diffusive fluxes. If the original studies measured methane fluxes with bubble traps or invert funnels, we included these as ebullitive fluxes.

We used a Monte Carlo approach to upscale river methane fluxes at the global scale and to estimate uncertainties. We performed simulations for five latitudinal bands (0-10^{o}, 10-25^{o}, 25-40^{o}, 40-60^{o}, 60-90^{o}) and at the global scale for diffusive, ebullitive and total fluxes. Because the data were skewed, we natural-log (*ln*) transformed all fluxes prior to simulations. For each simulation, we generated a total of 1,000 random values from a normal distribution centered around means of the *ln* methane fluxes and with deviations confined by those of the *ln* methane fluxes (R package mc2d). Values generated from the simulations were then back-transformed to raw fluxes before calculation of any statistics. Global methane emissions were calculated as the products of the recently-developed Global River Widths from Landsat (GRWL) surface area [53] and the post-simulation methane fluxes for each latitudinal band (Supplementary Table 4). Finally, we corrected our latitudinal methane emissions for ice coverage periods by excluding GRWL surface areas [53] with an atmospheric temperature below -4 °C for each month in each latitudinal band and at the global scale [54].

Lakes and reservoirs

We conducted a literature search until May 2019 using "(CH_4 OR methane) AND (concentration OR flux OR emission) AND (lake OR pond OR reservoir)" in the same search engines used for rivers and streams. Overall, the 84 publications provided 243 and 116 total methane fluxes for 227 lakes and 86 reservoirs, respectively. In our freshwater lakes, ponds and reservoir database, we included studies that provided both diffusive and ebullitive fluxes from the open water surface either separately (e.g. via bubble traps or acoustic surveys for ebullition and via thin boundary layer modelling or floating chambers for diffusion) or together (e.g. via floating chamber or eddy covariance methods). We categorized 'site' as either a lake or a reservoir, wherein a reservoir was defined as a system whose primary outflow was dammed. The lake category was largely comprised of natural lakes, and a small subset (n=23) of artificial lakes [55,56]. The total methane flux refers either to the total emission estimate (diffusive + ebullitive) of the whole water body reported by the authors; or was estimated by us using the mean of all reported areal fluxes (diffusive and ebullitive) or the mean of the range of reported fluxes (diffusive and ebullitive). We excluded studies that estimated only diffusive or ebullitive methane fluxes, but not both. We only include studies where both diffusive and ebullitive fluxes were estimated because the extent to which each contributes to total emissions is variable (ranging from negligible to most the flux) [57]. We further excluded methane fluxes that were made solely during mixing events, and fluxes from adjacent marsh and drawdown zones of reservoirs because they should be accounted for in freshwater wetland emissions. Finally, we also excluded methane fluxes from beaver ponds and river reaches upstream of weirs to avoid potential overlap with river and stream emissions. If original studies used an $r^2 > 0.85$ as a cut-off for linear gas accumulation in floating chambers, these fluxes were considered diffusive only and excluded from the dataset (unless accompanied by an independent estimate of ebullition).

Given previous evidence that chlorophyll *a* and ecosystem productivity are predictors of total lentic methane emissions [21,22], we used total phosphorus (TP) to model missing chlorophyll *a* data and assigned trophic statuses [22] . If no estimates of chlorophyll *a* or TP were reported by the primary study, we mined the literature for other studies of the same site that reported TP and/or chlorophyll *a* within a +/- five-year time period of the primary study.

We upscaled lake and reservoir methane fluxes separately to a global scale. For each water body type, we used a Monte Carlo analysis (R package mc2d) that allowed for uncertainty in both surface area and areal methane fluxes. Only sites with surface area information were included in the Monte Carlo analysis (n = 198 lakes, n = 78 reservoirs). Because the data were skewed, we *ln* transformed all total fluxes prior to Monte Carlo analysis. Our approach for binning Monte Carlo analyses differed for lakes versus reservoirs due to different correlates with methane emissions [57]. In a parallel study, we show that morphometric features better predicted methane emission in lakes, whereas chlorophyll *a* was a better predictor in reservoirs [57].

For small lakes < 1 km^2, we upscaled methane fluxes based on logarithmic size classes. However, for lakes > 1 km^2, our low sample size (n=20) precluded this approach, and we lumped together all lakes > 1 km^2. We ran a Monte Carlo analysis with 1,000 iterations for each size-classes of small lakes and for large lakes as one category. Each iteration randomly selected a methane flux from a normal distribution surrounding the mean and standard deviation for that size class. Simulations also selected for a surface area estimate of lakes in each size class using a uniform distribution based on estimates from [58–60]. Because Verpoorter et al.[60] report combined lake and reservoir surface area, we subtracted reservoir areas using estimates of reservoir surface area for each size class from Lehner et al. [61]. Because surface area estimates for lakes < 0.01 km^2 are highly uncertain, we extrapolated the data from Verpoorter et al.[60] to estimate the lower bound [62].

For reservoirs, we upscaled methane fluxes for small (< 1 km^2) and large (> 1 km^2) reservoirs. For small reservoirs, where sample size was low (n=16), we used the same scaling approach as with large lakes. For large reservoirs, where estimates of the global distribution of lake and reservoir chlorophyll *a* were available [63], we upscaled methane fluxes based on the positive log-linear relationship between chlorophyll *a* and areal methane fluxes [57] and reservoir surface area estimates [58,61]. We generated 1,000 Monte Carlo simulations of reservoir surface area based on a uniform distribution ranging between the surface area estimates by Downing et al. [58] and Lehner et al. [61]. We also allowed for uncertainty in the relationship between chlorophyll *a* and methane flux by generating 1,000 Monte Carlo simulations of slope and intercept terms based on a normal distribution around the standard error of these terms. We then estimated areal methane fluxes by applying reservoir surface areas across 20 chlorophyll *a* bins (with each bin spanning 5 µg L^{-1} from 0-100 µg L^{-1}), then calculating total methane emissions from each bin, and finally summing across the 20 bins. The global distributions of chlorophyll *a* concentrations were generated using MERIS OC4 satellite imagery via the MTRI method which is based on 300 m resolution inputs [63].

To account for the impact of ice on lake and reservoir emissions, we excluded surface areas [61] with an average atmospheric temperature of 0 °C or less for each month [54]. For lakes and reservoirs that freeze, we scaled upwards the ice-corrected emissions by 127% [64] to account for an ice-melt pulse in emissions. Both the ice cover and ice melt corrections were applied after the Monte Carlo upscaling by adjusting the estimated annual flux by the size class specific fraction of emission expected based on both ice cover and ice melt (fractions ranged from 0.60 to 0.98, Supplementary Table 4). Combined corrections for both ice cover and ice melt reduced overall annual methane emissions to 66% of their uncorrected values. We do not account for potential diel effects on lentic methane emissions. A further uncertainty is our small sample size for large lakes

(>1 km^2), and that half of the large lakes were shallow (≤ 3 m mean depth), and only 3 were >100 km^2 suggesting emissions may be overestimated from this size class.

Freshwater aquaculture ponds

We conducted a literature search using "(CH_4 OR methane) AND (aquaculture pond OR aquaculture farm) AND (shrimp OR fish)". For freshwater aquaculture ponds, we built on the database from Yuan et al. [65] and added three new studies of diffusive and ebullitive methane fluxes since 2018. Total freshwater aquaculture pond fluxes in the database were derived mainly from carp and mixed shrimp-fish ponds. We scaled areal freshwater aquaculture methane fluxes to global emissions using to the surface area estimated by Verdegem and Bosma [66] (Supplementary Table 5), which is likely an underestimate assuming an increase of freshwater aquaculture farms since 2009.

Coastal ocean

For each coastal ocean ecosystem, we performed a literature review until December 2019 using 'Scopus' by Elsevier (https://www.scopus.com/) and 'Google Scholar' (https://scholar.google.com/) databases. Additionally, we scanned the reference lists of publications. When methane fluxes were only presented in figures, we used a manual data extraction tool (WebPlotDigitizer) to estimate the values. We included methane fluxes with identifiable coordinates (latitude/longitude) derived from the original studies or from Google Earth based on site description. Coastal wetlands were distinguished from inner estuaries by accounting for methane fluxes solely from studies that were conducted in clearly vegetation-dominated marsh, seagrass or mangrove sites in opposition to spatial surveys over salinity gradients in estuarine open waters. For each coastal ocean ecosystem, we upscaled combined spatial and temporal methane fluxes to a global scale using recent surface area estimates (Supplementary Table 5).

610

611 ***Estuaries***

612 We conducted a literature search using "(CH_4 OR methane) AND estuary", which resulted
613 in 53 publications containing 137 averaged water-air methane fluxes for 124 sites. In our estuary
614 database, we included methane fluxes over full salinity gradients (freshwater to seawater). We
615 excluded methane fluxes from coastal wetlands and from incomplete coverage of salinity
616 gradients. Most studies reported diffusive methane fluxes computed from concentration gradients
617 and *k* parameterizations. A minority (n=3) of the studies measured diffusive fluxes with floating
618 chambers, and only one study reports measurements with eddy-covariance. If the original studies
619 estimated fluxes using several *k* parameterizations, we chose the value corresponding to the
620 parameterization most accepted (e.g., we chose the Wanninkhof [67] over the Liss and Merlivat [68]
621 model).

622

623 ***Saltmarshes***

624 We conducted a literature search using "(CH_4 OR methane) AND (saltmarsh OR salt-
625 marsh OR tidal marsh)", which resulted in 75 publications containing 89 averaged methane fluxes
626 for 60 sites. In our saltmarsh database, we included methane flux estimates and measurements for
627 saltmarsh and tidal marsh. We excluded methane fluxes from freshwater marsh (salinity <0.5) that
628 should be accounted for in freshwater wetland emissions. Saltmarsh methane fluxes were grouped
629 into three salinity classes: oligohaline (0.5-5), mesohaline (5-18), and polyhaline (>18) [69]. Most
630 studies (n=49) reported diffusive methane fluxes from the sediment-air interface during low tide
631 using static chambers. Several other studies (n=33) determined sediment-water-air fluxes during
632 exposed and inundated periods using either static dynamic chambers or eddy covariance. Few
633 studies (n=7) were available that determined the water-air methane flux, either computed based on
634 *k* parameterization or using the floating chamber approach.

Mangroves

We conducted a literature search using "(CH_4 OR methane) AND (mangroves OR mangrove forest)", which resulted in 56 publications containing 79 averaged methane fluxes for 59 sites. Our global mangrove methane emission estimate is an update of the review by Rosentreter et al. [26]. In our mangrove database, we included sediment-water fluxes from core incubations (n = 2), sediment-air (n=45) and sediment-water-air fluxes (n=8) using static chambers, and water-air fluxes (n=22) using floating chambers or based on *k* parameterizations. Our revised global estimate includes mainly diffusive sediment-air and water-air fluxes, but also plant-mediated fluxes (through pneumatophores, roots, stems, leaves), and fluxes over sediments with crab burrows. No estimates of ebullitive fluxes from mangroves were available. We excluded fluxes estimated from methane concentrations in gas bubbles that were actively stirred up from mangrove sediments, as they cannot be accounted for *in situ* ebullition.

Seagrasses

We conducted a literature search using "(CH_4 OR methane) AND (seagrasses OR seagrass beds OR seagrass meadows)", which resulted in 11 publications containing 18 averaged methane fluxes for 18 sites. In our seagrass database, we included plant-mediated and diffusive sediment-water fluxes (n=14) from submerged seagrass meadows and few available water-air methane fluxes (n=4) over seagrass meadows. The majority of studies reported sediment-water fluxes from core incubation and benthic chambers. One study used a dynamic flux chamber, which allowed flux measurements during exposed and submersed conditions, hence includes sediment-air fluxes [70]. No estimates of ebullitive fluxes from seagrass sites were available.

Tidal flats

660 We conducted a literature search using "(CH_4 OR methane) AND (tidal flat OR mud flat
661 OR sand flat)", which resulted in 23 publications containing 25 averaged methane fluxes for 16
662 sites. Tidal flat ecosystems were classified as tidal mudflats (unconsolidated fine-grain sediments),
663 tidal sand flats (unconsolidated coarse-grain sediments), and wide tidal rock-platforms
664 (consolidated sediments, organic material or rocks) [71] and distinguished from coastal wetlands
665 through the absence of vegetation. Because tidal flats comprise at least a global distribution of
666 127,921 km^2 [71], which is similar to that of mangrove forests, and are characterized by frequent
667 tidal inundation, we included tidal flats in our coastal ocean emission estimates. Our tidal flat data
668 set is biased towards tidal mudflats in China, with a few data from North America and Europe. We
669 included diffusive and ebullitive fluxes from coastal bare sediments of the inter-tidal zone (salinity
670 > 0.5) measured with static chambers or core incubations, which resulted in 16 sediment-air fluxes,
671 8 sediment-water-air fluxes and one water-air flux. We excluded freshwater bare sediments and
672 sites where the salinity region was unclear.
673
674 ***Coastal aquaculture ponds***
675 We conducted a literature search using "(CH_4 OR methane) AND (aquaculture pond OR
676 aquaculture farm) AND (shrimp OR fish)", which resulted in 10 publications containing 18
677 methane fluxes for 5 sites. Most methane fluxes (n = 10) were from studies conducted in coastal
678 aquaculture ponds near the Min River estuary in China. In our coastal aquaculture database, we
679 included diffusive and ebullitive fluxes mainly from shrimp ponds, with the residual measurements
680 from mixed fish-shrimp, mixed shrimp-sea cucumber, drained and undrained coastal aquaculture
681 farms.
682
683 ***Continental shelves***

Continental shelves were subdivided into 'estuarine plumes', 'seep areas' and 'upwelling areas', if identified as such in the literature, or based on the authors' knowledge. We conducted a literature search using "(CH_4 OR methane) AND (shelf OR coastal) AND (Arctic ocean OR upwelling OR river plume)", which resulted in 77 publications providing 9 methane fluxes for estuarine plumes, 19 for seep areas (diffusion), 3 for seep areas (ebullition), 12 for upwelling areas, and 57 for other continental shelf areas. In our continental shelf database, we included methane water-air flux estimates or measurements for continental shelf environments. We excluded studies that reported only the dissolved methane concentrations without computing fluxes. We summed our upscaled emissions from estuarine plumes, seep areas (diffusion + ebullition), upwelling areas, the East Siberian Arctic Shelf, and other continental shelves to total global continental shelf methane emissions.

Statistical analysis

We use the interquartile range (IQR) to describe methane flux variability, and the 95% confidence intervals (C.I.95%) (using the population standard deviation (σ) and sample size (n) assuming Student-T distribution and a confidence level of $\alpha = 1 - 0.95 = 0.05$) to estimate uncertainties of mean methane fluxes. For global estimates, we combined the confidence intervals by taking the square root of the sum of the variances. We applied the function 'skewness' from the R package e1071 (R Core Team, RStudio, version 1.2.5019) to compute the skewness coefficient of each data set. We did not conduct an assessment for publication bias.

Data availability:

The datasets that support the findings of this study are available in the "figshare" repository, 10.6084/m9.figshare.13611296.

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