GRiMeDB: The global river database of methane concentrations and fluxes

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Abstract. Despite their small spatial extent, fluvial ecosystems play a significant role in processing and transporting carbon in aquatic networks, which results in substantial emission of methane (CH_4) to the atmosphere. For this reason, considerable effort has been put into identifying patterns and drivers of CH_4 concentrations in streams and rivers and estimating fluxes to the atmosphere across broad spatial scales. Yet progress toward these ends has been slow because of pronounced spatial and temporal variability of lotic CH_4 concentrations and fluxes and by limited data availability across diverse habitats and physicochemical conditions. To address these challenges, we present athe first comprehensive database of CH_4 concentrations and fluxes for fluvial ecosystems along with broadly relevant and concurrent physical and chemical data. The Global River Methane database (GriMeDB; https://doi.org/10.6073/pasta/f48cdb77282598052349e969920356efhttps://doi.org/10.6073/pasta/b7d1fba4f9a3e365c9861ac3b58b4a90) includes 24,024 records of CH_4 concentration and 8,205 flux measurements from 5,0290037 unique sites derivedthat were extracted from publications, reports, data repositories, unpublished data sets, and other outlets that became availablepublished between 1973 and 2021. Flux observations are reported as diffusive, ebullitive, and total CH_4 fluxes, and GriMeDB also includes 17,655 and 8,409 concurrent measurements of concentrations and 4,444 and 1,521 of fluxes for CO_2 and nitrous oxide (N_2O) respectively. Most observations are date-specific (i.e., not site averages) and many are supported by data for 1 or more of 12 physicochemical variables and 6 site variables. Site variables include codes to characterize marginal channel types (e.g., springs, ditches) and/or presence of human disturbance (e.g., point source inputs, upstream dams). Overall, observations in GRiMeDB encompass thea broad range of the climatic, biological, and physical conditions that occur among world river basins, although some geographic gaps remain (e.g., arid regions, tropical regions, high latitudes and altitude systems). The global median CH_4 concentration (0.20 μmol L^{-1}) and diffusive flux (0.44 mmol m^{-2} d^{-1}) in GRiMeDB are lower than estimates from priorpast, site-averaged compilations, although ranges (0-456 μmol L^{-1} and -136-4057 mmol m^{-2} d^{-1}) and standard deviations (10.69 and 86.4) are greater forfrom this larger and more temporally -resolved database. Available flux data are dominated by diffusive measurements despite the recognized importance of ebullitive and plant-mediated CH_4 fluxes. NonethelessDespite these limitations, GriMeDB provides a comprehensive and cohesive resource for examining relationships between CH_4 and environmental drivers, estimating the contribution of fluvial ecosystems to CH_4 emissions, and contextualizingto contextualize site-based investigations.

1 Introduction

Despite their small areal extent, running-water (fluvial) ecosystems play a significant role in processing and transporting carbon (C) in and through aquatic networks, including the production, consumption, transport, and evasion of carbon dioxide (CO_2) and methane (CH_4). The profound planetary warming effects of CH_4 in the atmosphere, its erratic but accelerating rate of increase over recent years (NOAA, 2022), the significant contributions of natural sources to the growing atmospheric pool (Turner et al., 2019), and improvements in gas measurement technologies have all contributed to a rapid increase in studies of

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CH_4 dynamics in aquatic environments in general, and fluvial ecosystems in particular. These studies reveal widespread supersaturation of CH_4 in running waters that underlies their larger than expected contribution to the atmospheric pool (Stanley et al., 2016).

Efforts to quantify fluvial CH_4 dynamics at regional, continental, and global scales have been fraught with uncertainty, reflecting the inherent variability of this gas in surface waters combined with a notable limitation in data availability. Sources and sinks of CH_4 are often unevenly distributed over space and time within drainage systems and, as a result, concentrations can vary over 1-~~3-4~~ orders of magnitude over short time periods (sub-daily to sub-weekly; e.g., Natchimuthu et al., 2017; Smith and Böhlke, 2019) or relatively small spatial extents (<10 - <100 m for small streams and large rivers; ~~(~~e.g., Anthony et al., 2012; Crawford et al., 2017; Bretz et al., 2021; Robison et al., 2021). Similarly, several drivers or predictors of CH_4 have been identified in the literature, and these properties also have variable spatial and temporal distributions. Thus, efforts to estimate the total emissions from world rivers have relied on relatively small data sets composed of site-specific values that have been averaged over time, and then used~~have employed~~ upscaling strategies based on Monte Carlo techniques or extrapolations using predictor variables that have little or no significant statistical relationships with large-scale patterns of gas concentrations or fluxes (Hutchins et al. 2020). Consequently, current global scale estimates of riverine emissions are poorly constrained and highly uncertain (Saunois et al., 2020; Rosentreter et al., 2021).~~)~~

The combination of rapidly increasing atmospheric concentrations of CH_4, the significant role of fluvial systems in emitting this gas, and, critically, current difficulties in explaining or predicting concentrations and fluxes with reasonable certainty inspired the central goal of this paper: to assemble a comprehensive database of CH_4 concentrations and fluxes for fluvial ecosystems that includes broadly relevant concurrent physical and chemical data. This effort expands upon a prior compilation of CH_4 and CO_2 data (MethDB; Stanley et al., 2015) that was constructed to emphasize among-site differences and included 1,496 concentration records and 532 flux records from 1,080 sites. In this more comprehensive Global River Methane database (GRiMeDB), most data are date-specific (i.e., not averaged over time), the breadth of site types is expanded to include marginal fluvial habitats as well as disturbed and artificial waterways, and CH_4 data are supported by a broad suite of site-specific physical and chemical attributes along with concurrent measurements of CO_2 and N_2O where available. Given the more finely resolved scale of the data and the growth of the field in the past decade, GRiMeDB represents a significant expansion beyond MethDB. Building GRiMeDB with greater detail and breadth of data was done with the intent of increasing opportunities to identify and predict spatial and temporal variation in CH_4, to test hypotheses related to greenhouse gas dynamics, and to reduce uncertainty in future upscaled estimates of gas emissions. In this paper, we (1) provide a detailed description of the components of the database and its construction; (2) summarize some basic patterns of gas concentrations and fluxes from GRiMeDB; and (3) highlight critical data gaps and possible future research opportunities for improving current understanding of CH_4 dynamics in streams and rivers.

2 Database components and assembly

GRiMeDB is composed of four tables that contain information related to (1) data sources, (2) sites, (3) gas concentrations and supporting physicochemical data, and (4) gas fluxes. All tables are linked by unique data source identifiers, and all concentration and flux observations are also linked to unique site numbers (Fig. 1). Data included in GRiMeDB were gathered from scientific journals, government reports, public data repositories, theses, dissertations, and unpublished data sets provided by individual investigators. Sources were discovered via searches of bibliographic databases and data repositories (Web of Science, Google Scholar, ProQuest Dissertations & Theses Global, China National Knowledge Infrastructure, Environmental Data Initiative, USGS ScienceBase, Natural Environment Research Council (NERC) Environmental Information Data Centre, Arctic Data Center, and PANGAEA, Zenodo) using the keywords: methane and stream* or river* or ditch* or canal*, and searches were repeated numerous times prior to December 2021 for completeness. We also used informal 'word of mouth' approaches to discover additional, often unpublished data sets.

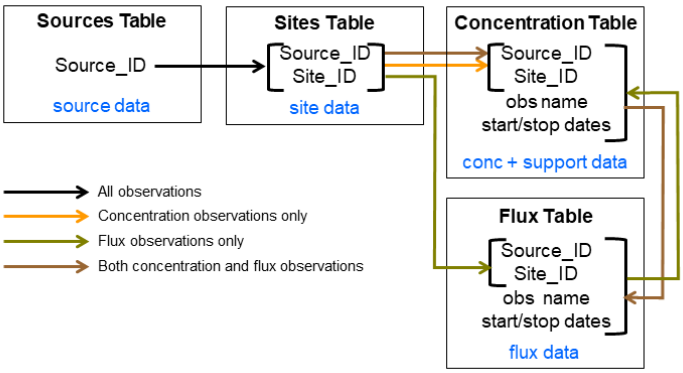

Figure 1: General structure of the GRiMe database and connections between its four tables. Information flow began with entering information about each data source into the Sources Table and assigning a unique Source_ID. Site information for each site within a data source was then entered into the Sites table. The site was given a unique Site_ID and linked to its data source by the Source_ID. Source_IDs and Site_IDs were carried over to all concentration and flux observations in their respective tables. Methane (CH_4) observations include site-date combinations with only concentration data (orange), only flux data (green), or both concentration and flux data (brown). Concentrations and available supporting data (described in Sect. 2.3) were entered into the Concentration Table, and each observation was given a unique observation (obs) name. For site-date combinations that had both concentration and flux observations, the Source_ID, Site_ID, observation name, and date information were copied to the Flux Table for data entry. Site-date combinations with flux data only were entered into the Flux Table and given a unique observation name. If a flux observation had associated supporting data, the Source_ID, Site_ID, observation name, and date information were copied to the Concentration Table for supporting data entry. However, if there were no supporting data, matching rows were not added to the Concentration Table.

All potential data sources were first screened to determine their appropriateness for inclusion in GRiMeDB. Several criteria were established *a priori* to ensure the usability of the data and that it was derived from inland running water systems. Coastal sites with

>1 ppt salinity were considered estuarine and thus were excluded. Similarly, sites that were situated in reservoirs or immediately upstream of small dams, dam spillways, beaver ponds, or lake outlets or were subject to experimental manipulation were omitted. We did not enter fluxes derived from chambers attached to collars or inserted into sediments because we could not be certain that such measurements were capturing air-water fluxes. Sources that reported minimum and maximum gas concentrations or fluxes only ~~as ranges (i.e., minimum and maximum values only)~~ were not included. Finally, rates expressed on an annual basis were also excluded to avoid introducing uncertainty associated with different upscaling assumptions and methods.

2.1 Sources Table

The Sources Table contains the list of all sources used to build GRiMeDB, a unique identification number (*Source_ID*) for each CH_4 data source, and basic bibliographic information for the data source (*Title*, *Author*, *Source*, publication year [*Pub_year*], and digital object identifiers [*Paper_DOI* or *Data_DOI_primarty*] or other persistent identifier; all column titles for this table are defined in Table A1). In several cases, data sources were supplemented with additional supporting information (e.g., associated physicochemical data) from separate sources (described further in Sect. 2.3) or additional or corrected information from authors (Fig. 2~~1~~). In the latter case, we contacted authors if questions arose regarding their data (e.g., clarification regarding units) and/or to request supporting information or site- or date-specific concentrations or fluxes if published values were aggregated. Inclusion of additional unpublished data from authors is noted in the Sources Table along with a description of the addition or correction. If supporting data from separate published sources were used, the DOI or other persistent identifier for the secondary source was listed in a separate column [*Data_DOI_supporting*].~~.~~

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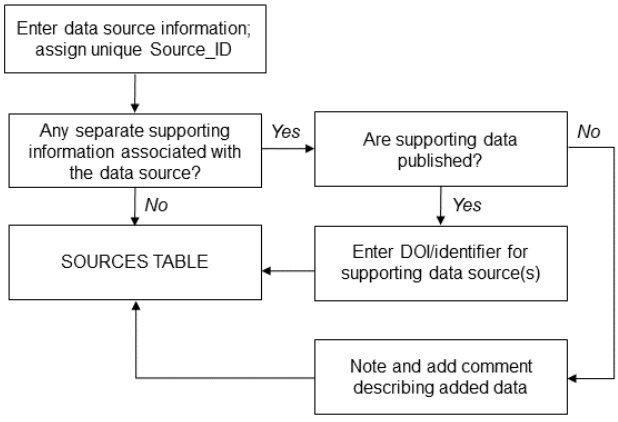

Figure 2~~1~~: Workflow for entering data into the Sources Table of GRiMeDB.

2.2 SitesSite Table

The Sites Table reports basic information on attributes for all sites where CH_4 was sampled. Each site has a unique identification code (*Site_ID*) and name (usually taken directly from the data source) and is linked to the Sources Table via the *Source_ID* (see Table A2 for detailed descriptions of all columns in the Sites Table). What composes a 'site' (i.e., the spatial extent of data collection) variedsite varies among data sources and includes (1) discrete sampling points, (2) geomorphically distinct discrete study reaches, and aggregations of points and/or (3) reaches across larger channel sections, areas such as a drainage networks, or other geographic units. The second case typically corresponded to reaches such as riffles or pools basin. Because gas data for sites in small streams. In thethis third case, multiple points were often sampled within the 'site' and data were then presented ascategory are averages. The distance between sampling points from locations that had been averaged varied widely, but were typically >1 km, and in some cases exceeded 100 km. Because may vary with respect to land use, channel order, slope, etc., can vary substantially across such distances, we included fields to indicate if a site was an aggregation of widespread points ('aggregated'),aggregated and if so, the number of locations in the aggregation (if available). We also limited the resolution of latitude and longitude for these sites to < 3 decimal places. At the opposite extreme, gas sampling at points very close to one another (a 'high density site' *sensu* Fig. 32) has the potential to create ambiguities for site delineation and data analysis. To avoid these pitfalls, we combined points with slightly different latitude-longitude values to represent a single site for three specific cases. First, multiple samples collected at different points and/or depths within a channel cross-section were averaged to form a single site. Second, some drainages or regions were surveyed repeatedly (particularly the Congo River basin and streams in Pennsylvania, USA) and it was not always clear if closely-situated (ca. 10-50 m) points from different surveys were intended to be a repeated sampling of the same location or sampling of discrete sites. Some judgment was involved in choosing between these two possibilities, and in a subset of cases, points in close proximity to one another that were sampled on separate dates were treated as a single site. What constituted 'close proximity' varied between small streams and large rivers but was always <100 m, and typically <50 m. Finally, three data sources had extremely high sampling densities within discrete reaches (50 - >20,000 samples per reach; Crawford et al., 2016; Call et al., 2018; Loken et al., 2018). Because closely adjacent gas samples can be spatially autocorrelated (Crawford et al., 2017) and including all individual values from these studies would have resulted in their over-representation in the database, individual point measurements were treated as within-reach replicates.

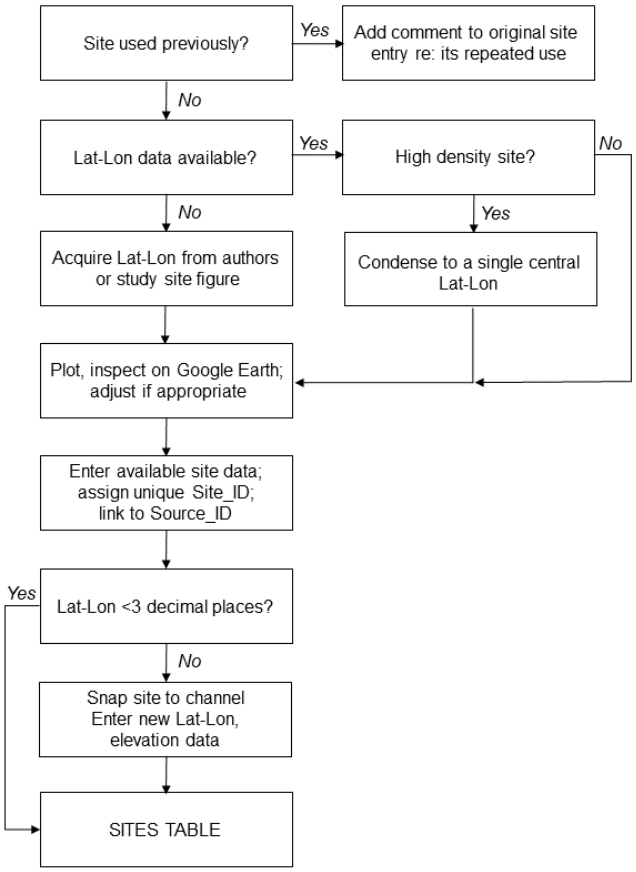

Figure 32: Workflow for entering and checking data for the GRiMeDB Sites Table. 'Lat-Lon' is an abbreviation for latitude and longitude.

For a site used in multiple studies, the *Site_ID* was assigned to the earliest paper and a comment was added to the site entry noting its use in other data sources (Fig. 32). Latitude and longitude coordinates were available for most sites; however, in several cases, location information was acquired from authors or estimated from study site figures using Google Earth (© Google Earth 2020). All sites~~Sites~~ were plotted on Google Earth and inspected (Fig. 32) to identify and correct data errors. If a site's coordinates were immediately adjacent to, but not on a channel, the coordinates were adjusted to fall on the channel

and this modification was noted in the *Comments* field. If available, additional variables drawn from the data sources were entered to characterize the site, including stream name, basin or region name, elevation, channel slope, Strahler order, basin area, and codes denoting distinct channel or site types (described below). To supplement the available elevation data, we~~We~~ also estimated elevation for all sites except aggregated sites or sites with poorly-resolved coordinates (≤~~(less than~~ 3 decimal places for both latitude and longitude) after snapping coordinates to the nearest stream. To determine the adjusted within-channel coordinates, we first~~using the following procedure. First, we~~ downloaded a digital elevation model (DEM) for each site using the function *get_elev_raster()* from the package "elevatr" (version 0.4.2; Hollister et al., 2021) for R statistical software (version 4.2, R Core Team 2021) at a resolution of 6-9 m depending on the location in the globe. Second, the DEM was processed for hydrological correctness using the package "whitebox" (version 1.2.0, Wu, 2020) by filling single cell pits (*fill_single_cell_pits()* function) and breaching depressions (*breach_depressions()* function) to obtain a flow direction model (*d8_pointer()* function). Finally, we calculated a flow accumulation model (*d8_flow accumulation()* function). If the coordinates reported in the data source had a flow accumulation ≤~~less than~~ 10 cells (indicating that they were not located in a preferential flow path), ~~the~~ new coordinates were assigned to the cell with the highest flow accumulation within a 50 m radius. If the initial site had a high flow accumulation value (>10 cells), we assumed the site was in a stream channel. Typically, the snapping procedure resulted in very minor changes to a site's location (median <3 m).

Many studies of CH_4 dynamics have been undertaken to determine if and how specific phenomena such as presence of upstream reservoirs, point source discharges, thermokarst features, or oil and gas extraction potentially affect fluvial CH_4 (and other constituents), usually with an expectation of a net enhancement of concentrations and fluxes. Similarly, other studies have examined sites that may be expected to be enriched in CH_4, but whose fluvial identity might be considered marginal or ambiguous (e.g., springs, floodplain backwaters, ditches, canals). Inclusion of such 'methane hunting' studies has the potential to bias the dataset toward higher values (Stanley et al., 2016). Nonetheless we included these studies in GRiMeDB because they provide an opportunity to investigate the consequences of human activity and gain a more comprehensive understanding of fluvial CH_4 dynamics. ~~(e.g., see Alshboul et al., 2016; Peacock et al., 2021).~~ However, to accommodate future analyses in which use of such data might be unsuitable, or alternatively, when these sites might be the sole focus of a study, we generated a set of channel codes to identify targeted site types (Table 1). Information about four of the codes was not consistently available among data sources and thus their assignment often involved judgment calls. The first case involved determining if the presence of an upstream dam (code DD) was ~~or was not~~ relevant for sites of varying distances downstream ~~distances~~. We used a ~~value of~~ distance of 7 km as a cut-off for this category, although the zone of influence ~~of small or large dams~~ may be far shorter or extend far beyond this distance depending on dam size and operation (Kemenes et al., 2007), respectively. To provide some context for this code, a site's distance from a dam was acquired from the data source or estimated in Google Earth using the Path tool and reported in the *Comments* field whenever possible. The second case involved straight, symmetrical channels that are common in many agricultural and urban areas. Frequently, it was not known if this unnatural geometry was due to channelization (straightening) of a stream (code CH) or creation of a new channel (ditches and canals; codes DIT and CAN). In the absence of specific information, straight channels were classified

as CH. Third, channels draining or passing through wetlands (WS) were often difficult to identify, particularly given seasonal variation in wetland appearance~~. in tropical systems with wet-dry climates.~~ Finally, floodplain channels presented a distinct challenge because of the complex nature of these environments and their potential to be classified as either riverine or wetland systems. We used the FP code to indicate habitats that were described as, or appeared to be lentic (i.e., backwaters or connected floodplain lakes) but were persistently connected to the main river channel and thus were part of the fluvial system. Given these ambiguities, we recommend that these four codes be viewed and used with care.

2.3 Concentration Table~~Concentrations~~ and Flux Table ~~Fluxes Tables~~

The Concentration Table~~Concentrations~~ and the Flux Table ~~Fluxes Tables~~ contain the primary gas data central to GRiMeDB, and the Concentration~~Concentrations~~ Table also hosts physical and chemical variables associated with concentration and/or flux observations (see Tables A3 and A4 for the full list of columns ~~Concentrations Table~~ and ~~Fluxes Table columns and~~ their descriptions). The vast majority of concentration and flux data were extracted from tables within data sources, data repositories, or provided by authors. However, in some cases, values were acquired from figures using graphical digitizing software (WebPlotDigitizer (https://automeris.io/WebPlotDigitizer/), GetData (http://getdata-graph-digitizer.com/), or DigitizeIt (https://www.digitizeit.xyz/)). Plots with log scales or that were difficult to interpret were not digitized. The accuracy and consistency of this method were evaluated by comparing data generated by different individuals digitizing a set of common figures and by comparing digitized results to known results. Agreement ~~both~~ between both comparisons was strong (average slope = 0.994, average R^2 = 0.9996 for 5 comparisons between individuals digitizing the same dataset, and average slope = 0.998, average R^2 =0.997 for digitized versus actual data for 7 datasets; see Table S1 for further details), demonstrating the reliability of this method of data gathering.

Whenever possible, concentrations and fluxes were entered as values for individual sites on individual days (i.e., not averaged across sites or days) (Fig. 4~~3~~). Because 1 day represented the lowest level of temporal resolution in GRiMeDB, repeated measurements made on a sub-daily scale were averaged and expressed as a daily value and were not considered to be aggregated over time. If multiple replicates were collected at different times on the same day (e.g., a study of diurnal gas dynamics), this was noted in the *Comments* fields and measurements prior to and after 12:00 a.m. (local time) were entered as separate, consecutive days. Observations resolved to the daily scale can be identified using either a "No" in the *Aggregated_Time* field or by having the same reported starting (*Date_start*) and ending (*Date_end*) dates. If the specific start and end dates were not specified in the data source, we entered the day as the 15th of the month and noted this approximation in the *Comments* field. If available, we also reported minimum and maximum values and standard deviations (SD) for entries that were aggregated over space and/or time. SDs, but not minima and maxima were reported for replicates from non-aggregated sampling when available, except for reach-averaged entries with multiple within-reach measurements and diel studies with multiple within-day values. In these cases, minima and maxima were also included.

Table 1. Codes denoting distinct site or channel attributes or presence of conditions that potentially affect methane (CH_4) concentrations or fluxes. Assignment of codes to a site is based on information provided in the data source and/or physical appearance of a site and a site may have more than one code. Codes are reported in the *Channel_type* field of the Sites Table.

Code	Definition
CAN	Canal or other artificial channel with hardened channel boundaries
CH	Channelized; a channel that has long straight-line sections of uniform width and changes in channel direction are typically distinct angular features rather than curves
DC	Channel in a river delta
DD	Downstream (within 7 km) of a dam. Samples from spillways were excluded~~not included~~.
DIT	Ditch, typically for agricultural drainage, without channel hardening
FP	Site in a floodplain water body connected to the main channel that appears lentic or is described as a floodplain lake or backwater. This category does not include ~~braided river~~ side-channels within floodplains or tributary channels transecting a floodplain
GT	Site below the toe or terminus of a glacier
IMP	Presence of multiple and typically small impoundments in a site's vicinity (e.g., various European rivers, Mississippi River)
PI	Permafrost influenced; this refers specifically to sites at or immediately below thermokarst outflows and not to sites in areas underlain with permafrost
PS	Immediately (<1 km) downstream of a point source discharge
SP	Spring channel; this does not include sites characterized as seeps (features with low flow volume adjacent to channels)
TH	Site receiving inputs of thermogenic CH_4, either naturally or as a result of mining, fracking, oil extraction, and other related activities.
WS	Wetland stream; site is in a wetland or immediately downstream from the outlet of a wetland
NORM	Non-targeted site

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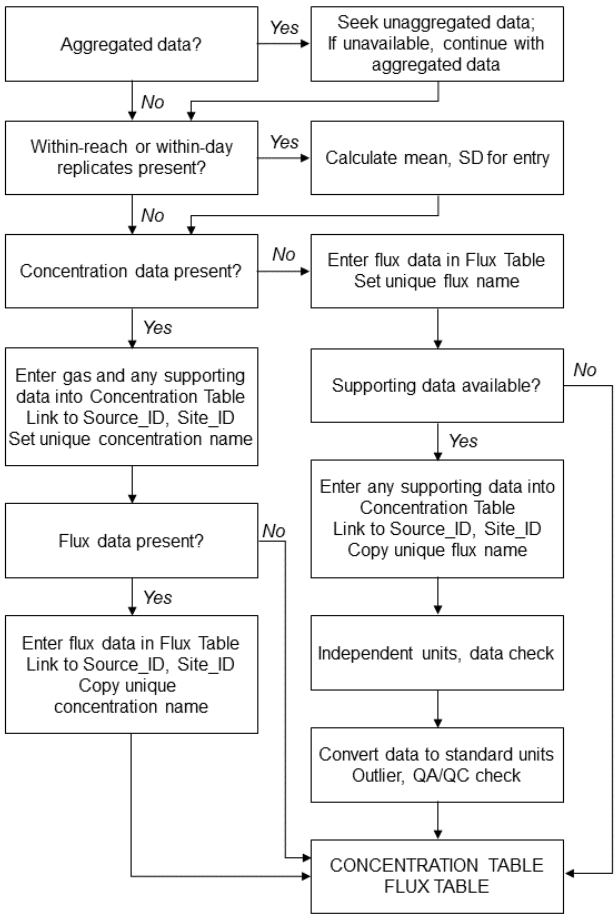
Dealing with concentration data reported as a negative value, zero, or below a detection limit (BDL) is problematic because of inconsistencies in detection limits and reporting practices, and any decision about handling these records introduces some bias (Stow et al., 2018). For example, using a non-numerical format such as BDL or <0.01 is likely to lead to the elimination of these entries during data analysis and thus would introduce a bias against low-value observations. Alternatively, converting any such

value to zero would introduce a bias in the opposite direction. As a compromise solution, concentrations recorded as zero in the original data source were entered as zero in GRiMeDB and other below-detection values were entered as -999999. In this latter case, the original data entry format was noted in the *Comments* column. For fluxes, negative and zero values were entered without modification or comment.

The FluxFluxes Table reports diffusive, ebullitive, and total CH_4 fluxes along with CO_2 and N_2O diffusive fluxes. Given the diverse strategies for measuring each of the three CH_4 flux pathways and potential biases associated with different approaches (Lorke et al., 2015; Chen et al., 2021), values are accompanied by brief categorical descriptions of methods used for each CH_4 flux type as well as for CO_2 fluxes and the gas exchange coefficient *k*. For a small number of entries, CH_4 fluxes were not directly reported in the data source but information was available (dissolved gas concentration, temperature, and a corresponding gas exchange coefficient (*k* value))) that allowed us to calculate these fluxes. We also entered BDL values for flux for one data source in which fluxes had been calculated from concentration, but fluxes associated with BDL concentrations had been omitted from the results. Finally, a small number of observations listed diffusive and ebullitive but not total fluxes, so diffusion and ebullition were summed and entered as total flux. In all cases, the added calculations are noted in the *Comments* field.

The GRiMeDB ConcentrationConcentrations Table includes physicochemical measurements in support of concentration and flux observations (Figs. 1, 4Fig. 3, Table A3). Availability of this supplemental information varied widely among data sources, and was limited to data collected concurrently with gas samples. For data sources with gas fluxes and physicochemical data but not gas concentrations, we created rows in the ConcentrationConcentrations Table to capture the supporting data. These records are identified by a "Yes" in the *FluxYesNo* column, *SampleCount* = 0, and NA in the *CH4mean* column. Finally, water temperature was estimated for entries if it was needed to convert gas units and entered in the *WaterTemp_degC_estimated* column. Estimates were typically based on values from the same or adjacent sites or the same site at a a similar timetimes (e.g., averages of temperature from the prior and subsequent dates, or from the same month in an adjacenta prior year). Error introduced from these estimates should be small; e.g., ca. <10% of the actual value if the estimated temperature is off by 3°C.

Following completion of all data entry, gas and physicochemical variables were converted to 'new' standard units (Tables A3, A4). The identities of the new and original units are included in both the Concentration TableConcentrations and Flux TableFluxes Tables for clarity. Elevation was used to estimate atmospheric pressure if needed for unit conversions. We used Henry's Law, water temperature, and atmospheric pressure to convert dissolved gas values reported in ppm, ppb, µatm, and % saturation (~13% of observations). For observations that reported gas values as percent saturation (<1% of all observations),%), we also used the global average CH_4, CO_2, and the N_2O atmospheric concentrations from the NOAA Global Monitoring Laboratory (https://gml.noaa.gov/ccgg/) for the year 2013, which corresponds to median observation year in the database.


Aggregated data?
Yes
Seek unaggregated data; If unavailable, continue with aggregated data
No
Within-reach or within-day replicates present?
Yes
Calculate mean, SD for entry
No
Concentration data present?
No
Enter flux data in Flux Table Set unique flux name
Yes
Enter gas and any supporting data into Concentration Table Link to Source_ID, Site_ID Set unique concentration name
Supporting data available?
No
Yes
Enter any supporting data into Concentration Table Link to Source_ID, Site_ID Copy unique flux name
Flux data present?
No
Yes
Enter flux data in Flux Table Link to Source_ID, Site_ID Copy unique concentration name
Independent units, data check
Convert data to standard units Outlier, QA/QC check
CONCENTRATION TABLE FLUX TABLE

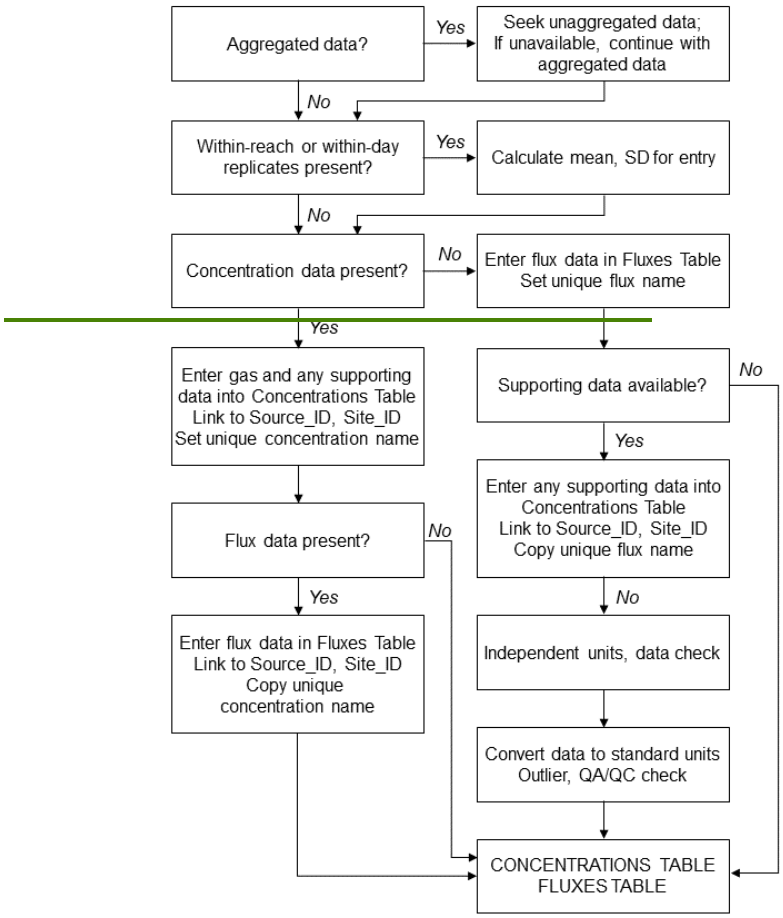


Figure 43: Workflow for entering and checking data for the GRiMeDB Concentration Table~~Concentrations~~ and Flux Table~~Fluxes Tables~~.

2.4 Assessment of representativeness

We assessed the representativeness of sites in GRiMeDB relative to the global distribution of biological, physical, and climatic properties following van den Hoogen et al. (2021). Briefly, we first assigned each site to a corresponding river reach in

HydroSHEDS (Linke et al., 2019), which is a global hydrological network database that contains spatial data for a wide array of hydrological, physiographical, climatic, land cover, geological, edaphic and anthropogenic variables for each river reach. HydroSHEDS thus provides a multidimensional characterization of global rivers that is well suited for assessing how representative GRiMeDB sites are in terms of key biophysical and anthropogenic features. After excluding non-numerical variables (e.g., biome) and variables with monthly values (e.g., monthly precipitation), we performed a principal component analysis (PCA) on all HydroSHEDS subcatchments using all possible combinations of the 54 remaining HydroSHEDS variables. From this, we selected all principal components (PCs) needed to explain 90% of the variance in the PCA, which corresponded to 28 PCs and 378 possible bivariate combinations of these PCs. For each unique PC pair, we computed the convex hull of all sampled sites to determine the distribution of these sites relative to all global river subcatchments for the specified PCs (Fig. 54). Each HydroSHEDS subcatchment was then assigned a value of 1 or 0 if it fell within or outside the convex hull, respectively. This process was repeated for each of the 378 possible PC combinations. To collapse this information, we calculated the fraction of cases that a given subcatchment fell within the convex hull for all PC combinations to obtain a dimensionless summary value ranging from 0 to 1. A subcatchment with a value of 1 for this index of "representativeness" means that it fell within the convex hull for 100% of the PC combinations, indicating that its overall characteristics are well captured in the database. It is important to note that this analysis only captures average catchment properties of relatively large river reaches (average subcatchment area: 130 km^2). Given the strong local controls on CH_4 concentrations and fluxes, interpretations from this analysis should be made with some caution.

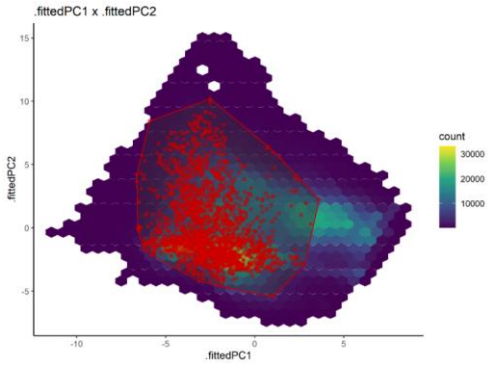


Figure 54: Example of a representative principal components analysis (PCA) hexagon plot based on variability in HydroSHEDS river subcatchment attributes. Hexagon colour indicates the number of subcatchments per hexagon. Subcatchments hosting GRiMeDB sites are plotted in red and contained within the convex hull delineated by red lines. Subcatchments that fall within this polygon are assigned a value of 1 and those outside the perimeter are given a value of 0 to indicate the representativeness of sampled reaches for this pair of PC axes. See Sect. 2.4 for further explanation.

2.5 Data checking and data analysis

Several approaches were taken to check the accuracy of data in GRiMeDB. This included evaluation of the reliability of digitized data (Sect. 2.3) along with several additional inspection steps. Entries were error checked by a co-author other than the individual who entered the data, including confirmation of site location information, validating units for all variables, and spot- or complete checking of entered gas data (independent units and data check in Fig. 4~~3~~), depending on dataset length and if data were manually entered or imported directly from a file. Once values had been converted to standard units, all variables were plotted to identify outliers (outlier check; Fig. 4~~3~~), and extreme values were checked against the original data source. In cases in which errors were present in the original data, if possible, authors were contacted for clarification. In the few rare cases in which issues could not be resolved, the data were excluded. These and all other calculations and analyses were performed in R (version 4.2, R Core Team 2021), using the "dplyr" package (version 1.0.7, Wickham et al., 2021) and "data.table (Dowle and Srinivasin, 2021) for data analysis, "sf" package (version, 1.0, Pebesma, 2018) for spatial data processing, and "ggplot2" (version 3.3.5, Wickham, 2016) and patchwork (Pedersen, 2020) packages for visualization.

3. RESULTS

3.1 Overview of GRiMeDB data

GRiMeDB includes 24,024 records of CH_4 concentration and 8,205 CH_4 flux values from 5,037 unique sites, along with 17,655 and 8,409 concurrent measurements of concentration and 4,444 and 1,521 of flux for~~of~~ CO_2 and N_2O, respectively (Table S2). Although the first concentration and flux values in GRiMeDB were published in 1973 (Lamontagne et al., 1973) and 1987 (de Angelis and Lilley, 1987), respectively, over 70% of all CH_4 concentrations and 80% of flux observations became available after 2015 (the year of publication of MethDB; Fig. 6, Fig. S1~~5~~). This growth in data availability has occurred predominantly along the spatial axis, as almost two thirds of all sites were added in or after 2015 and over half of all sites in the database have a single concentration and/or flux observation. Conversely, long timeseries are rare, with only 8% of the 5,037 sites having > 10 concentration observations and 4% having >10 diffusive flux records (Fig. 6, 5~~,~~Fig. S1). The longest concentration record includes 590 observations distributed over 28 years (Toolik Inlet, *Site_ID* 9025; Kling, 2019a~~;~~ 2022) while the longest flux record has 82 observations of diffusive flux over 4 years (*Site_ID* 3644; Aho et al. 2021). Further, among the 15 sites with time series >~~-~~5 years, 12 are situated in either the Toolik Lake region of Alaska, USA (Kling, 2019a~~;~~ 2019b~~;~~ 2022) or within the Krycklan watershed in Sweden (Wallin et al., 2018~~;~~ Wallin, 2021~~unpublished~~).

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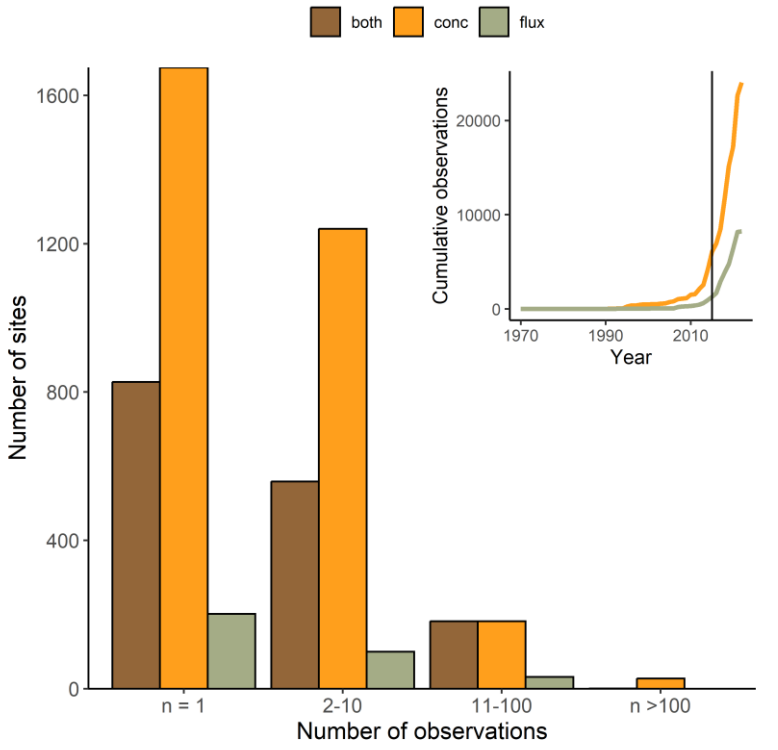
both
conc
flux
Number of sites
1600
1200
800
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1970
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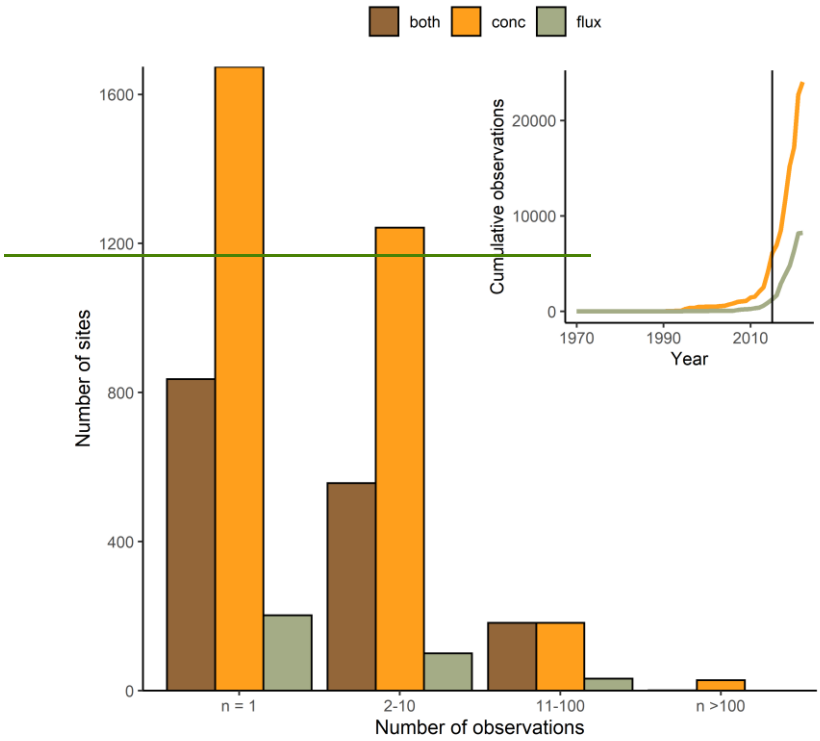

Figure 6~~5~~: Distribution of the number of methane (CH_4) observations per site. Brown bars indicate sites with both concentration (conc) and flux observations. Orange and green bars show sites with only concentration and only flux observations, respectively. Inset: Cumulative observations of CH_4 concentration and flux data based on the year of publication of the data source. The vertical line (2015) indicates the year of MethDB (Stanley et al., 2015) publication. See Fig. S1 for data accumulation and length resolved by CH_4 flux type.

3.2 Spatial and temporal distribution of data

Spatially, 40% of all sites and 52% of all CH_4 concentration observations are in North America, followed by Europe (25% of all sites and 26% of all CH_4 concentration values~~observations~~; Table S2). Conversely, there are vast geographic areas with moderate to high channel densities with few or no observations, such as central Canada, Central America, South America beyond the Amazon mainstem area, most of Russia, central and western Asia, New Zealand, and the Malay Archipelago (Fig. 7a). Geographic limitations in availability of flux data, particularly of ebullition, are pronounced given smaller number of observations and domination of diffusion measurements. Observations of ebullition are absent or limited to 1-2 studies for

Africa, Oceania, Central America, South America, and Russia (~~6a~~Fig. S2). Despite these gaps, there is surprisingly good representation in terms of the range of hydrological, physiographical, climatic, land cover, geological, edaphic, and anthropogenic conditions that exist globally (Fig. 7b~~6b~~). Areas that are poorly represented are characterized by very low channel density associated with arid or polar climates as well as high altitude regions (Greenland, northern Africa, central Australia, Middle Eastern nations, western China, Mongolia, Chile, southern Argentina). Evaluating the distribution or representativeness of sites in terms of system size is difficult given the limited availability of relevant information such as Strahler stream order or basin area, which were reported for only 26% and ~~-~~28%, respectively,~~%~~ of all sites (Table S2). For sites with these data, counts of observations decline with increasing stream order (Fig. 8~~7~~) in a log-linear fashion (R^2 = 0.92 for concentration and 0.90 for flux; P <0.0005 for both regressions after excluding zero-order counts), consistent with Horton's Law of Stream Numbers (Horton, 1945). Thus, other than the extreme under-representation of zero-order channels, this predictable decline suggests reasonable representation by order. Nonetheless, ~~, although~~ this result should be interpreted with caution given the scarcity of relevant data. The distribution of counts by basin size follows a similar pattern of under-representation of sites draining very small basins and also indicates a potential over-representation of some large basin sizes (~~Fig. 7; e.g.,~~ basins of ca. 10,000 km^2; Fig. 8).

The distribution of observations among months illustrates seasonal sampling regimes dominated by summer sampling in northern (>~~-~~40˚) and southern (< -20˚) latitudes contrasted by even or erratic sampling at mid-latitudes (Fig. 9~~8~~). Consistent with the lower representation of southern hemisphere rivers and streams, several months lack concentration and/or flux measurements south of -10˚ latitude, particularly during winter months. Beyond these gaps, the only months missing data in the northern hemisphere are fluxes in January and February at sites north of 60˚ latitude and several missing months north of 70˚, presumably due to pervasive ice and snow cover.

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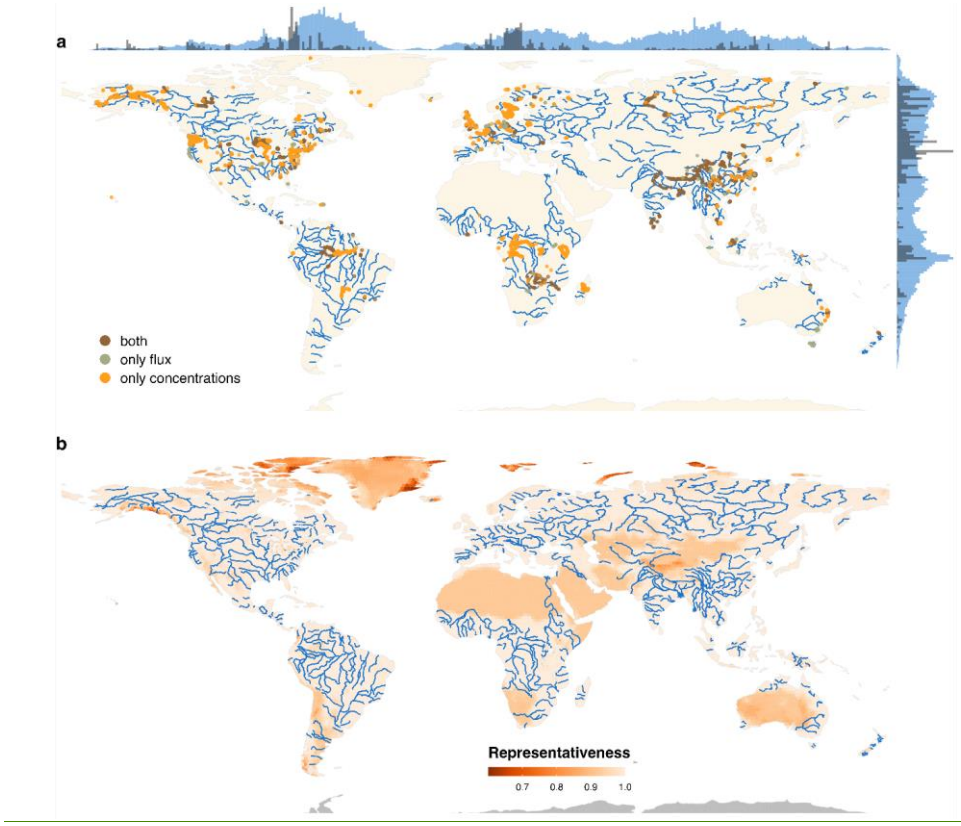
a
both
only flux
only concentrations
b
Representativeness
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1.0

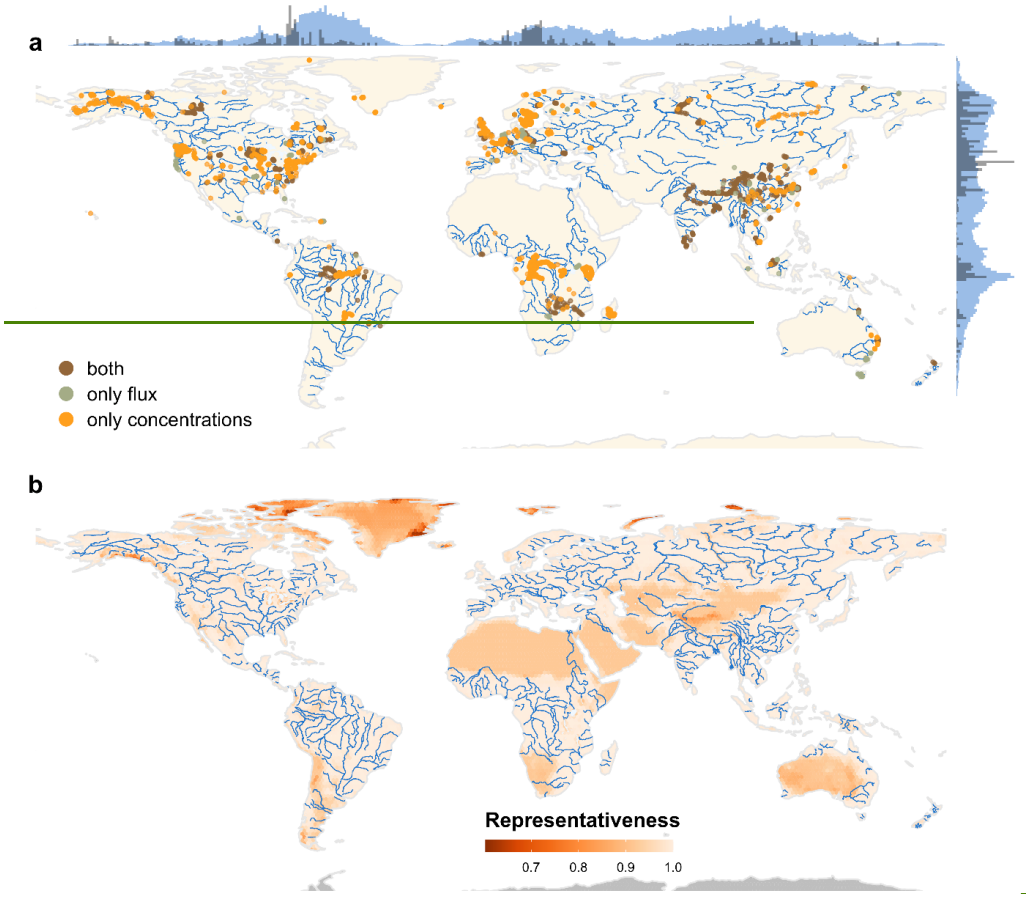


Figure 7: (a) Global distribution of methane (CH_4) observations in the database, colour coded for sites with concentration data only, flux data only, or both concentration and flux data. Top and right panels show, respectively, longitudinal and latitudinal patterns of the density of CH_4 observations (grey bars) and the density of river area (blue bars). These bars have been aggregated at a 1 latitudinal or longitudinal degree and rescaled from 0 to 1 for this visualization. River area was obtained from BasinAtlas (Linke et al. 2019). (b) Representativeness (dimensionless) of the database based on a wide array of biological, physical, hydrological and land cover variables (see Sect. 2.4 for details). Values close to 1 indicate a high representativeness, with only 4% of the global river surface below a threshold of 0.9. See Fig. S2 for data distribution resolved by CH_4 flux type.

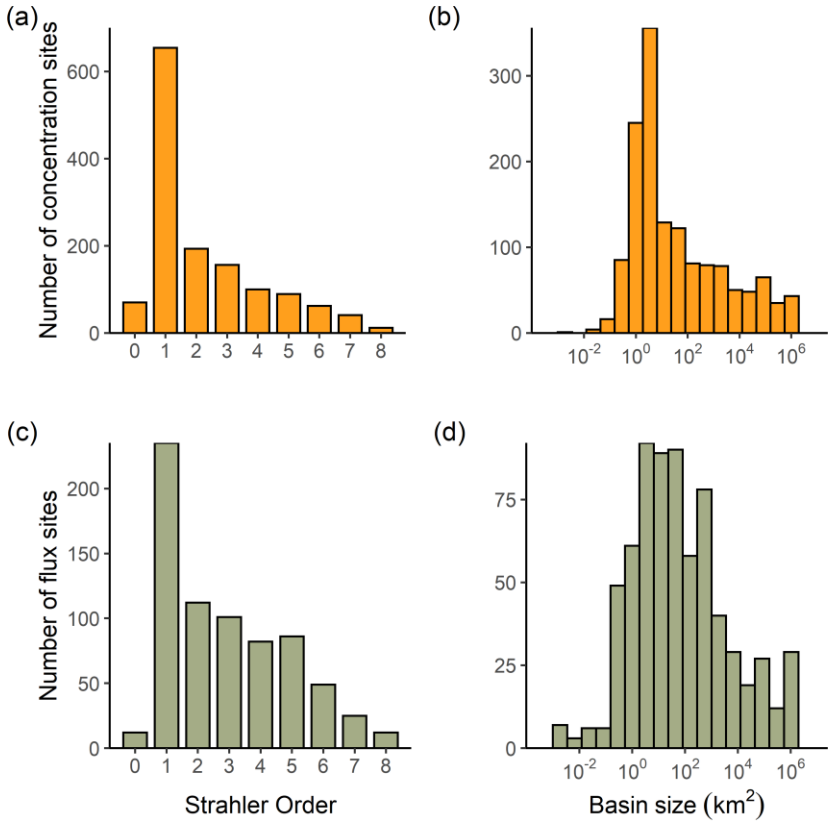


Figure 87: Number of sites with concentration (a, btop) or diffusive flux (c, dbottom) observations as a function of stream order (a, cleft) and basin size (b, dright) for the subset of sites with channel order and/or basin size information.

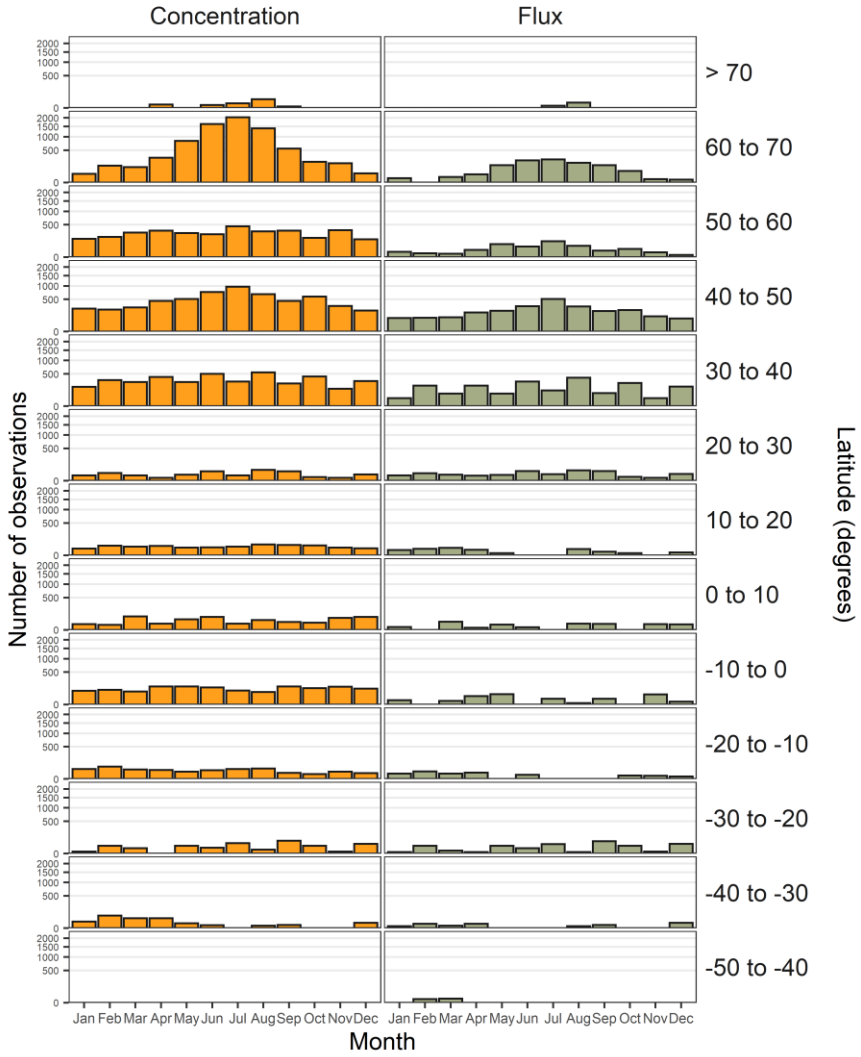
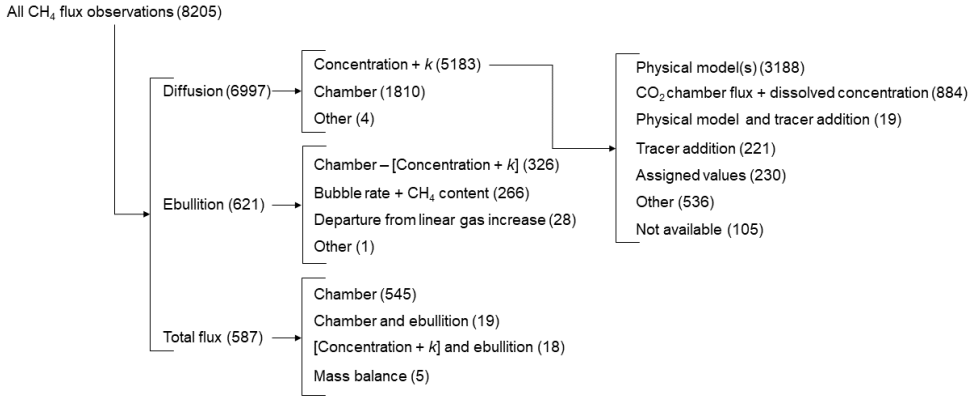


Figure 98: Number of observations of concentration (left) and flux (right) by month for 10˚ latitude bands.

3.3 CH_4 flux methodology

Records of CH_4 flux are dominated by diffusive flux measurements, which represent 85% of all flux values in the database, with ebullition (8%) and total flux (7%) accounting for the remaining entries (Fig. 109). Not surprisingly, a variety of methods have been used to quantify each flux type, although diffusive flux methods are dominated by calculations based on dissolved gas concentration and a gas exchange coefficient (*k*) (74% of all observations), while chamber-based methods are most common for quantifying total flux (93% of all observations). Similarly, ~~the gas exchange coefficient~~ *k* is most commonly estimated via physical models (*n* = 31883, 188). Several models have been employed for this calculation, as indicated by >25 different references for *k* model sources listed in GRiMeDB.



~~Fig.~~ **~~Figure~~ 109: Counts of methane (CH_4) flux observations by type (left), by major methodological categories for each pathway (middle), and for method type used to estimate the gas exchange coefficient *k* (right). For clarity, the chamber category includes all chamber types and patterns of gas increase in the chamber unless specified; more resolved methodological data are presented in the GRiMeDB Flux~~Fluxes~~ Table. See Table A4 for further details about category definitions.**

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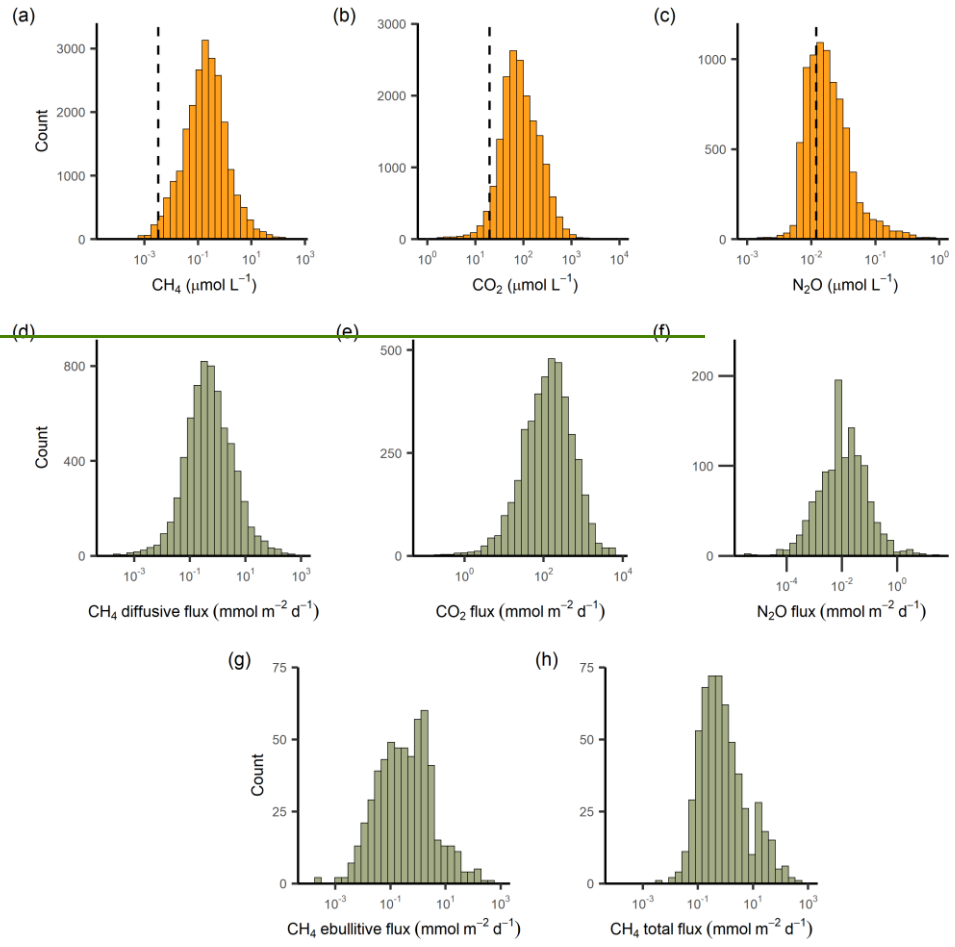
3.4 Overview of concentration and flux data

Concentrations and fluxes of all three gases are characterized by log-normal distributions that vary over~~range across~~ several orders of magnitude (Fig. 1110) and large coefficients of variation (CVs) for CH_4 and especially N_2O (Table 2). The vast majority (~95%) of CH_4 and CO_2 concentrations appear to be supersaturated, in contrast to N_2O concentrations with~~in which~~ 67% of observations ~~were~~ above this threshold. Reports of concentrations below detection are scarce for all gases, including N_2O (Table 2). ~~For fluxes,~~



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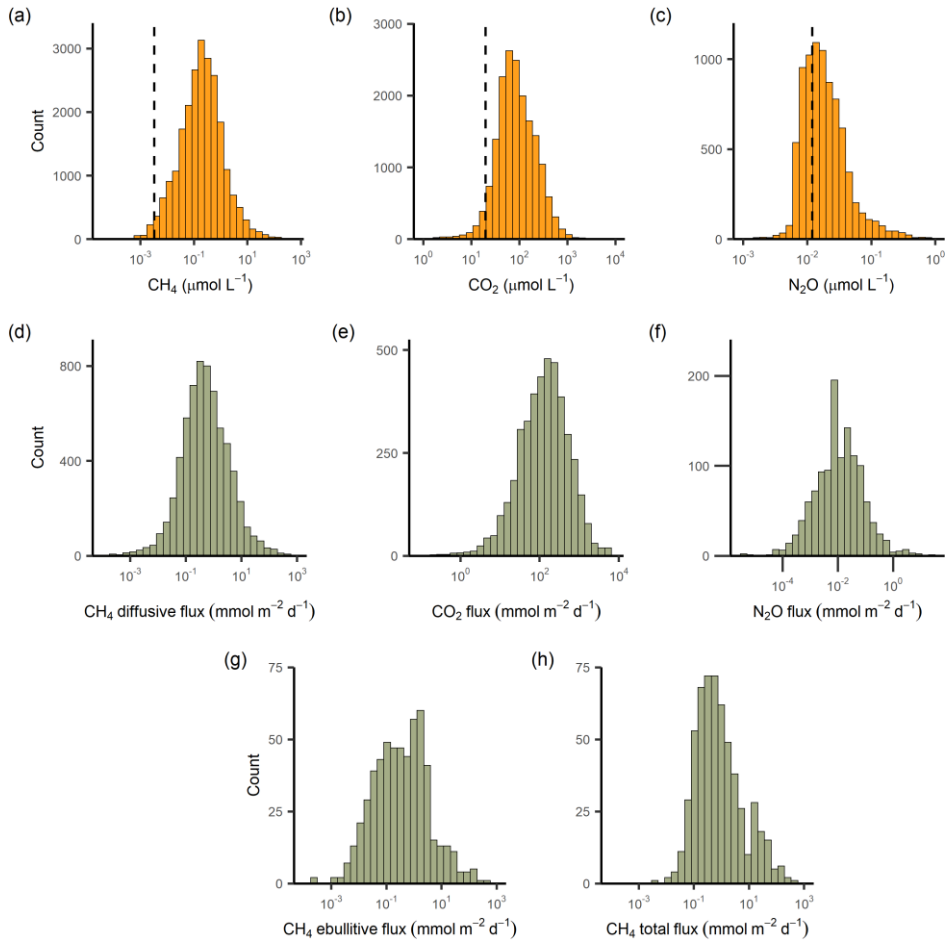

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Figure 11~~10~~: Histograms of gas concentrations and fluxes in GRiMeDB, excluding values reported as below detection or zero; counts of these values are reported in Table 3. Dashed vertical lines in the concentration histograms indicate the 100% saturation concentration based on the median estimated elevation (250 m) and water temperature (12.5) for all sites and atmospheric concentrations of 1.83, 400, and 0.325 ppm for methane (CH_4), carbon dioxide (~~,~~CO_2),~~,~~ and nitrous oxide (N_2O),~~,~~ respectively.

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The~~the~~ fraction of observations with zero, below detection, or negative fluxes (5%,~~,~~ 5%,~~,~~ and 19% for diffusive CH_4, CO_2, and N_2O fluxes, respectively~~), were similar to~~~~). corresponded reasonably well with~~ the frequency of subsaturated concentrations. At the other extreme, the highest CH_4 concentrations (>~~-~~200 µmol L^{-1}) paradoxically occur in either anthropogenically~~-~~influenced large rivers of the warm tropics (e.g., Amazon basin: Kemenes et al., 2007; Ganges, Mekong: Begum et al., 2021) or in small boreal headwater streams (e.g., Campeau et al., 2018; Wallin et al, 2018).

There were no meaningful univariate relationships between variables that may be used for upscaling (latitude, basin area, and stream order) and mean site concentration or flux (Fig. 12~~11~~, Table S3). Linear~~Although~~ regressions indicated that ~~were significant for~~ latitude and flux~~, latitude~~ accounted for a very small percent of the variation in both concentration (R^2 = 0.006 and 0.002, respectively~~0025~~) and flux (R^2 = 0.036 and 0.055~~004~~) among sites. Similarly, concentration and flux among stream orders suggested possible differences ~~were significantly different~~ for concentration (Kruskal-Wallis ~~tests: Kruskal-Wallis~~ χ^2 = 47.165, *df* = 8~~46.072~~, *P* <0.001) and ~~marginally different for~~ flux (χ^2 = 14.777, *df* =8, ~~796,~~ *P* = 0.070~~06~~). However, results of corrected pairwise comparisons (using the method of Benjamini and Hochberg, 1995) ~~revealed no significant differences~~ among orders were ambiguous, suggesting no ~~for flux, and~~ differences among orders for flux. For concentration, these comparisons indicated possible differences in distributions~~(*P* <0.05)~~ only between 7th order channels and all other orders,~~-~~and between 6th vs 1st order sites for concentration. Collectively, these results indicate a lack~~, indicating an absence~~ of a consistent change in CH_4 magnitude across channel orders for flux.~~. In contrast, variability decreased with increasing order and basin size, although this pattern is likely influenced by the accompanying decrease in sample size across this gradient.~~

Table 2. Summary statistics for methane (CH_4), carbon dioxide (~~,~~ CO_2)~~,~~ and nitrous oxide (N_2O) concentrations and fluxes. The %BDL (below detection level) column reports the percent of all observations that are below detection limits (including values reported as zero) for concentration. See Table S2 for counts and Table S3 for statistical summaries for all other variables. Standard deviation (SD) and coefficient of variation (CV).

Metric ~~Gas~~	Gas (and type)~~Variable~~	Mean	Median	Max	Min	SD	CV	%BDL
Concentration (µmol L^{-1})								
	CH_4	1.49	0.20	456	0	10.69	718	3.2
	CO_2	135	81.7	5,479	0	174.8	130	0.05
	N_2O	0.058	0.017	32.9	0	0.602	1,042	0.59
Flux (mmol m^{-2} d^{-1})								
	CH_4-diffusive	7.31	0.44	4,057	-136	86.4	1,182	
	CH_4-ebullitive	5.42~~4.65~~	0.28~~26~~	366	0	24.02~~22.75~~	443~~490~~	
	CH_4- total	8.71~~7.62~~	0.63~~62~~	366	-0.05	31.90~~28.5~~	366~~375~~	

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CO_2	319	128	23,749	-1625	770	242
N_2O	0.082	0.008	31.3	-11.3	0.981	1,199

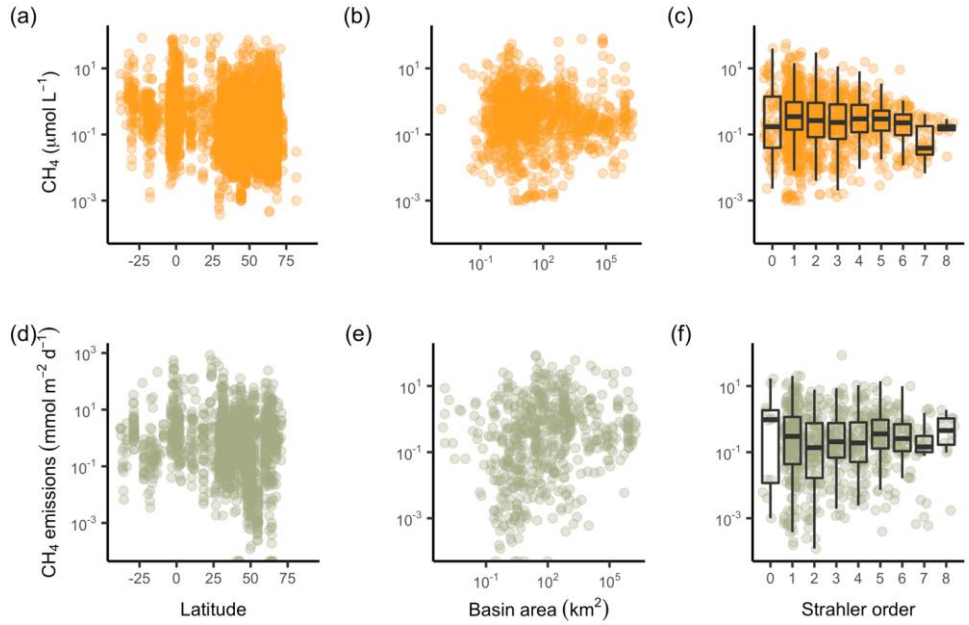
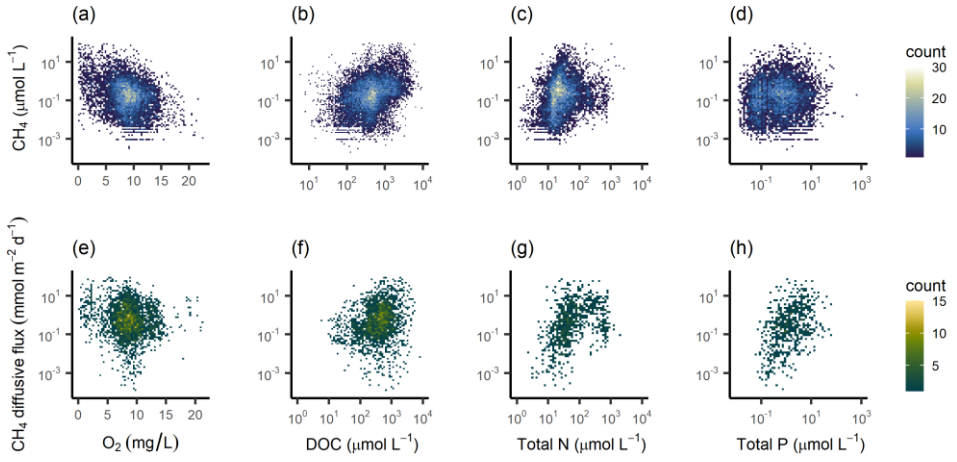


Figure 1211: Site-average methane (CH_4) concentrations (a-c) and flux (d-f) as function of latitude, basin area, and Strahler stream order. For boxplots, the upper and lower edges of each box are the 25th and 75th percentiles and whiskers are drawn up to 1.5 times the interquartile range.

As with relationships between CH_4 and physical site attributes, relationships between CH_4 concentration or flux and water chemistry parameters are also characterized by substantial variability. Representative examples indicate increasing, decreasing, and ambiguous relationships between CH_4 concentrations and fluxes and selected chemical constituents (Fig. 1312). One source of the variation in the relationship shown in Fig. 13 may12 can be attributed to differences among sites, as is illustrated for the case of CH_4 concentration versus discharge. (Fig. 13). The cluster of all points in this plot (Fig. 14a)by itself does not suggest an obvious linear relationship between concentration and discharge; however, resolving the data to the site level for

sites with multiple observations reveals several significant trends (Fig. 14b).~~.~~ Among 57 sites with >30 observations, 42 had significant relationships ($P < 0.05$) between concentration and discharge and 30 of these 42 trends were negative.

Median site concentrations ~~and fluxes~~ for most categories of targeted channels (Fig. 15) differed from ~~14) were significantly different than~~ "normal" (NORM) sites (Kruskal-Wallis test $\chi^2 = 460.1$, $df = 12$, $P < 0.0001$ after dropping channel types with <10 observations to improve test reliabilty).~~)~~. Pairwise Wilcoxon comparisons adjusted to account for multiple comparisons (Benjamini and Hochberg, 1995) indicated that springs (SP) and delta channels (DC) were similar to ~~did not differ from~~ NORM sites ($P > 0.4$) and impoundment-influence (IMP) sites were marginally different ($P = 0.053$). Concentrations in channels at glacial termini (GT) and floodplain backwaters (FP) were lower ($P < 0.0001$), whereas all other site types had ~~significantly~~ higher site average CH_4 concentrations than NORM sites. Fluxes also varied among channel type ~~were also significantly different~~ (Kruskal-Wallis test $\chi^2 = 126.4$~~143.8~~, $df = 8$~~12~~, $P < 0.0001$ after dropping channel types with <10 observations), and similar to concentration, fluxes in delta channels (DC) and channelized sites (CH)~~, permafrost-influenced channels (PI), and springs~~ were similar to NORM channels while all other channel types considered had~~. Pairwise comparisons indicated that all other site types differed from NORM sites. Further, fluxes at floodplain sites were significantly~~ higher median fluxes~~($P < 0.02$) than NORM sites, in contrast to the significantly lower concentrations for this site type. However, sample sizes were very small for FP, PI, as well as GT sites (in addition to an absence of flux data for TH sites), so comparisons for these sites should be viewed very cautiously~~.



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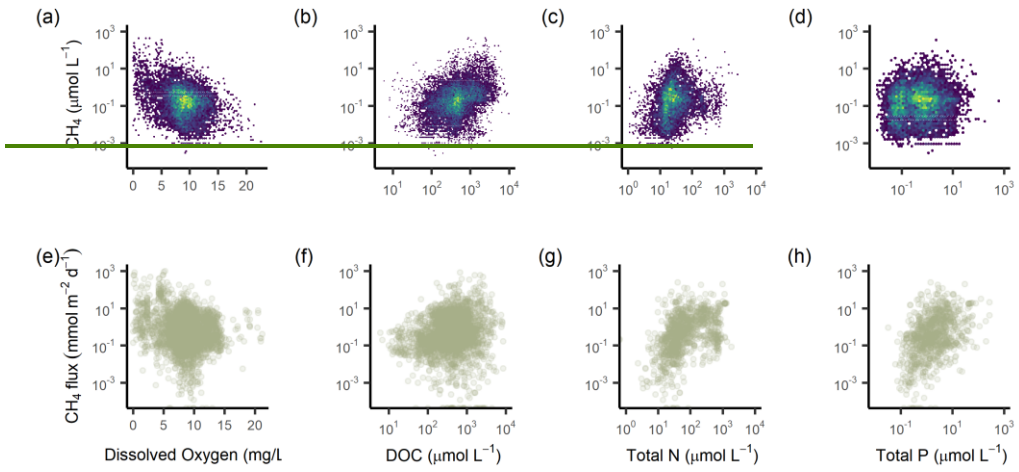

Figure 13: Methane (12: CH_4) concentration (a-dtop row) and diffusive flux (e-hbottom row) versus concurrent measures of dissolved oxygen (O_2; *n* = 8,529 and 2,316 for concentration and flux, respectively), dissolved organic carbon (DOC; *n* = 14,441 and 1,901), total nitrogen (Total N; *n* = 8,378 and 467) and total phosphorus (Total P; *n* = 6,904 and 240). Three outliers were excluded from the DOC plots, and because of the log scale for CH_4, negative and zero values have been omitted. For concentration plots, colours represent number of observations per polygon, varying from 1 (dark blue) to 30 (yellow).

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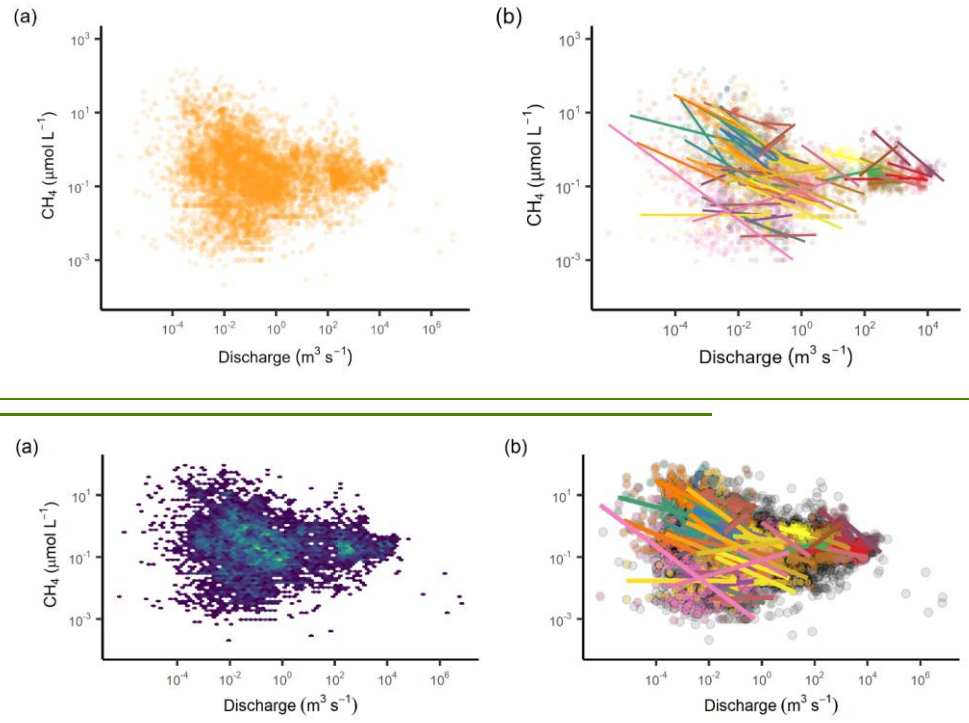

Figure 14: Methane (~~13:~~ CH_4) concentration versus concurrent measures of discharge for (a) all sites with discharge data and (b) sites with >30 observations (57 sites) with trend lines denoting within-site relationships between concentration and discharge. Each site is represented by a separate colour. Because of the log scale for CH_4, negative and zero values are omitted.

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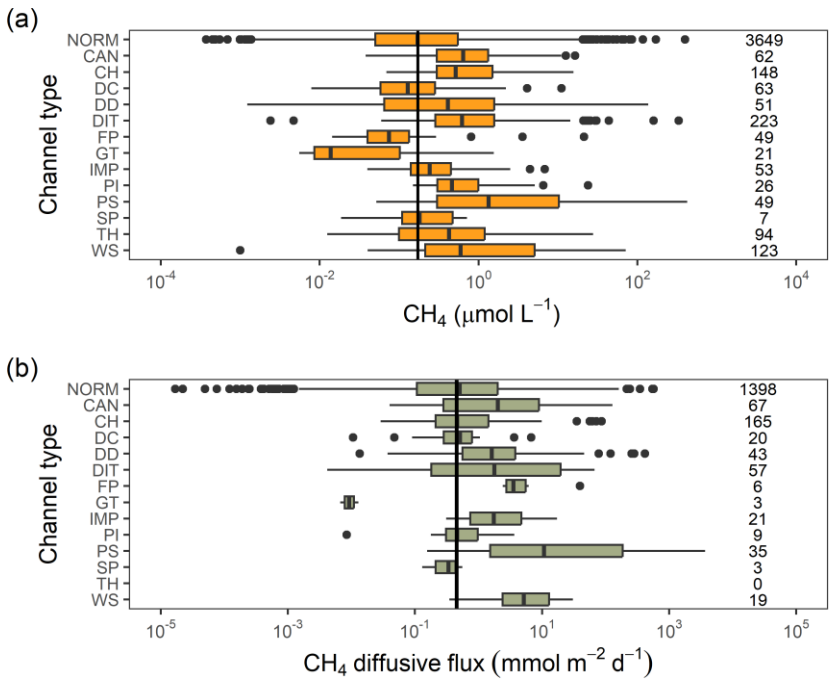
(a)
Channel type
NORM
CAN
CH
DC
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GT
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PI
PS
SP
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WS
3649
62
148
63
51
223
49
21
53
26
49
7
94
123
10^{-4}
10^{-2}
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10^{2}
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CH_4 (μmol L^{-1})
(b)
Channel type
NORM
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DC
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PI
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1398
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165
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43
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CH_4 diffusive flux (mmol m^{-2} d^{-1})

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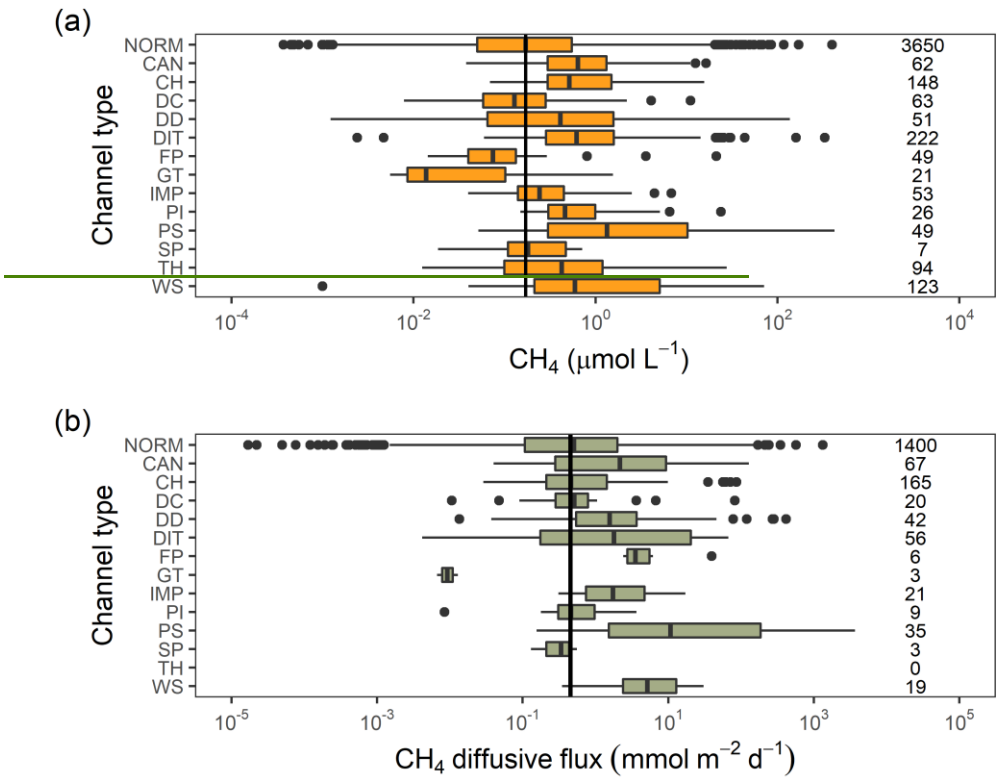

~~Fig.~~ **~~Figure 15~~14: Boxplots of site-averaged Methane (CH_4) concentration (a) and diffusive flux (b) for channel type categories. Channel categories are defined in Table 1, but briefly are as follows: NORM- non-targeted sites; CAN-canals; CH- channelized streams; DC-river delta channels; DD- downstream of dams; DIT- ditches; FP- floodplain backwaters; GT- glacial outflows; IMP- impounded reaches; PI- permafrost (thermokarst) influenced; PS- point source influenced; SP- springs; TH- thermogenic CH_4 inputs; WS- wetland streams. Number of sites per channel type are listed on the right side of each plot. The vertical black line denotes the median concentration and flux for non-targeted (NORM) sites. Because a log-scale is used in these plots, zeros and negative values were excluded. The actual median for non-targeted sites represented by the vertical line is therefore slightly different than the median displayed in the corresponding box plot because of this exclusion. The upper and lower edges of each box are the 25th and 75th percentiles, whiskers are drawn up to 1.5 times the interquartile range, and points are plotted if beyond the whiskers.**

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4. Discussion

The rapid increase in availability of aquatic CH_4 (as well as CO_2 and N_2O) data over the past 5-10 years has been remarkable and creates new opportunities for examining patterns and drivers of these gases in lotic ecosystems across broad spatial scales. ~~in lotic ecosystems.~~ Similarly, constructing GRiMeDB provided us with an unprecedented opportunity to identify tendencies in when, where, and how CH_4 has been sampled in streams and rivers. Examination of such data collection tendencies can reveal important biases and gaps within a field (Stanley et al. 2019, Gomez-Gener et al. 2021b) and thus points to future research needs and opportunities. Below, we discuss the distribution of sampling efforts and methodological issues, preliminary data analyses, and consider questions that GRiMeDB can help to answer.

4.1 When and where: sampling effort considerations

The growth of GHG studies in flowing water systems in the past decade includes a geographic expansion beyond the large body of historic and current work in temperate regions of North America and Europe. In particular, recent research in Africa, Australia, and especially southeast Asia has greatly improved the global coverage of available data. However, studies in arid drainages remain scarce- even beyond what would be expected given their small river surface area. A possible explanation for the limited study of CH_4 in these systems may be the pervasive focus on the contribution of streams and rivers to the global atmospheric CH_4 pool, and the corresponding assumption that aridland ~~arid land~~ systems play a minor role in this context. Yet we suggest that limited study in arid and semi-arid drainages represents a missed opportunity to understand metabolism and carbon cycling in a set of streams and rivers that drain nearly half of the global land surface, are increasingly stressed by growing human water demands (e.g., Sabo et al., 2010; Lian et al., 2021; Stringer et al., 2022), and support ecosystem process rates that are amplified by warm temperatures and highly variable flow regimes (Fisher et al., 1982; Ran et al., 2021). Beyond arid and semi-arid basins, further research emphasis in tropical and high-latitude regions would also be beneficial even given recent improvements in data availability and geographic representation of both areas. Existing data for tropical forests and grasslands are dominated by studies of African rivers (especially the Congo drainage) and the Amazon River system. In fact, observations from tropical areas of the Indomalayan and northern Australasian region represent <3% of all sites, and Central America is represented by a single study. Tropical drainages are frequently characterized by high CH_4 concentrations and fluxes, along with rapid changes in land use and river regulation that are affecting C cycling and GHG dynamics (Park et al., 2018; Flecker et al. 2022). However, understanding or detecting the magnitude and consequences of these anthropogenic changes on fluvial CH_4 is constrained by these current sampling limitations. Finally, while high latitude regions (north of the Arctic Circle) are well represented in GriMeDB with >3,600 concentration observations, more than 80% of these values are derived from studies in the vicinity of the Toolik Field Station in Alaska, USA, and thus do not capture the full biophysical diversity of Arctic biomes (Metcalfe et al. 2018). Given that climate change at high latitudes is progressing faster than elsewhere on the planet (IPCC, 2021 ~~in press~~), and that the global north stores massive quantities of C in soils (Hugelius et al., 2014), more extensive coverage of CH_4 across Arctic drainage systems is warranted.

Although the spatial coverage of CH_4 data has improved markedly over the past decade, expansion across temporal dimensions has lagged. The predominant mode of sample collection has been and continues to be through surveys that yield one or a few observations from individual sites (e.g., Bouillon et al., 2012; Kuhn et al., 2017; Jin et al., 2018; Ho et al., 2022),) and studies characterizing seasonal dynamics or responses to a site-specific environmental change are limited. Indeed, long-term (>5 years) CH_4 datasets in general are extremely rare (Leng et al., 2021); no such data are currently available for fluxes and most long-term concentration records are derived from just a few clustered locations. Determining the consequences of changes in land use or habitat attributes on fluvial CH_4 dynamics have instead relied on space-for-time substitutions (e.g., Smith et al., 2017; Gatti et al., 2018; Woda et al. 2020) rather than on direct observations of change over time. Although this strategy has been successful in revealing variation in GHG dynamics among different site types, current knowledge about how gases vary over time and respond to perturbations is poorly developed because of these data limitations. This deficit may be particularly consequential in the case of climate change, as the broad scope of this phenomenon will inevitably limit the effectiveness of spatial sampling approaches.

The discussion above regarding the 'when' and 'where' of sampling emphasizes large spatial and relatively long temporal scales, consistent with the extent of GRiMeDB. However, another current deficit in our understanding relates to the degree of heterogeneity of this gas at fine spatial and temporal scales, and thus if current sampling strategies are missing meaningful variation. Recent studies of CO_2 provide a cautionary tale in this context, as failure to account for diurnal variation in this gas results in a consistent under-estimation of fluvial emissions that is quantifiable at regional (Attermeyer et al., 2021) and global (Gómez-Gener et al., 2021b) scales. Similar questions may arise for spatial variation; that is, what is the minimum grain size or appropriate spatial scale for sampling ~~of~~ CH_4 in running waters (Crawford et al., 2017; Lupon et al., 2019)? The potential to examine very short-term variation is not possible using GRiMeDB data because of our decision to average ~~of~~ within-day measurements given the current small number (ca. 20) of these temporally ~~-~~detailed studies. Assessment of fine-scale spatial variation is also limited because of limited fine-scale sampling in general, as well as by decisions made both by investigators and during database construction. For example, geomorphologically distinct units (e.g., an individual riffle or pool) are often used as a basic sampling unit and results are presented as averages of replicates collected at different points within the study reach (e.g., Hlaváčová et al., 2006; Smith et al., 2017). In general, information about replication was frequently omitted, or if reported, information about variability among replicates was frequently absent. In addition to this limitation, our decision to combine replicates taken at different points in a channel cross-section and within individual channel units that had hundreds to thousands of datapoints to avoid ambiguities for site delineation and data analysis also constrains the opportunity to examine variation at fine spatial scales. However, we anticipate that this situation will change over the next few ~~several~~ years, as *in situ* sensors or other devices capable of collecting high-frequency/high density gas measurements become more widely available. Recent papers signal this new frontier and have highlighted the presence (e.g., Lamarche-Gagnon et al., 2019; Smith and Bohlke, 2019; Chen et al., 2021; Taillardat et al., 2022) and absence (e.g., Castro-Morales et al., Chen et al., 2021; 2022;

Rovelli et al., 2022; Zhang et al., 2021) of predictable diel variation in CH_4 concentrations and fluxes, and varying degrees of within-reach spatial variability (Crawford et al., 2016; 2017; Call et al., 2018; Bussman et al., 2022).

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4.2 How: methodological considerations

Measuring dissolved GHG concentrations or fluxes involves multiple steps and calculations. Field and laboratory protocols vary widely in the literature, and methodological variety is particularly conspicuous for flux determination. Ironically, even
575 though many studies of lotic CH_4 dynamics are framed in terms of understanding the contribution of these ecosystems to the rapidly increasing atmospheric CH_4 pool, flux measurements lag far behind those of concentration~~,~~ and the vast majority (ca. 85%) of observations are of ~~flux data quantify only~~ the diffusive pathway alone. Further, the most common method for estimating this pathway involve combining dissolved CH_4 concentration with *k*, the gas exchange coefficient. Quantifying *k* is notoriously challenging (Hall and Ulseth, 2019) and the large number of approaches for calculating *k* used among data
580 providers is concerning and undoubtedly introduces substantial uncertainty. A more in-depth consideration of the consequences of different models or strategies for arriving at a *k* value was beyond the scope of this paper, but inclusion of methodological information should be useful for such an analysis in the future.

~~.~~ Ebullition measurements are notably scarce despite the potential of this pathway to account for a large fraction of total emissions in some streams (e.g., from 30-9890% of total CH_4 emissions, as shown in~~;~~ Baulch et al., 2011; Crawford et al.,
585 2014; Chen et al., 2021). The conventional approach to quantifying ebullition involves a combination of capturing bubbles just below the water surface to determine the area and time-specific rate of bubble volume reaching the surface and measuring CH_4 content of recently-erupted bubbles. The episodic nature and extreme spatial heterogeneity of ebullition (Crawford et al., 2014; Spawn et al., 2015; Chen~~Robison~~ et al., 2021; Robison et al., 2021) require multiple ~~) requires good replication of~~ bubble trap replicates~~traps~~ that need to be deployed over several~~multiple~~ days to generate reliable measurements~~measurement~~. Given
590 the logistic challenges and labour-intensive work involved, indirect approaches are becoming more common. These approaches typically use the difference between a chamber-based measurement of flux, which is assumed to represent total flux (diffusion + ebullition) and diffusion calculated from dissolved CH_4 and *k* (i.e., the 'chamber – [concentration + *k*]' method in Fig. 109) to estimate ebullition (e.g., Campeau et al., 2014; Zhang et al., 2020; Ran et al., 2021). We suggest that this approach should be used cautiously, however. This~~For example, this~~ strategy is arguably inappropriate for situations in which the ~~chamber~~ gas
595 content within a chamber increases in a linear fashion during the measurement period, consistent with the occurrence of diffusive flux alone. Second, it is not clear if it is reasonable to assume that chamber-based measurements capture both diffusion and ebullition, even if a chamber-based flux value is greater than that calculated from dissolved CH_4 concentration. Relatively~~Further, relatively~~ short chamber deployments are likely to miss or incompletely capture bubble releases, while long-term deployments are vulnerable to sampling artefacts associated with altered concentration gradients within, and/or turbulence

around the chamber (Sawakuchi et al., 2014; Lorke et al., 2015). Given these challenges, it is not altogether surprising that comparisons between direct and indirect measurements of ebullition can yield substantially different results (e.g., Yang et al., 2012; Bednařík et al., 2017; Chen et al., 2021).

The final and most profound knowledge gap in the collection of flux data is the absence of measurements of plant-mediated emissions. Plant-mediated fluxes can account for a substantial fraction of total emissions from wetlands and shallow lake habitats (Bodmer et al., 2021),) but the contribution of this pathway is unknown in fluvial systems. Indeed, we did not include plant mediated fluxes in GRiMeDBGRiMe DB because we encountered only two papers that had explicitly quantified this pathway in streams (Sanders et al., 2007; Wilcock and Sorrell, 2008). Although aquatic macrophytes are sparse or absent from many streams and rivers, they can be abundant in low-gradient, low-disturbance environments (Riis and Biggs, 2003; Gurnell et al., 2010) where diffusive fluxes would be constrained by low gas exchange rates. Sediment trapping and venting by macrophytes enhances both methanogenesis and methane emission in these systems (Sanders et al., 2007), but the significance of such processes and the contribution of plant-mediated fluxes at larger spatial scales remain to be determined for fluvial systems (Bodmer et al., 2021).

4.3 Concentration and flux patterns

Not surprisingly, the massive increase in data availability hashave led to differences in averages and measures of variability for CH_4 concentrations and fluxes compared to our previous efforts. Median values for all three CH_4 flux pathways in GRiMeDB are 1.2-2.2 times lower than those reported by Stanley et al. (2016), as well as those from Rosentreter et al. (2021). Conversely, measures of variability (SD, CV) in GRiMeDB are almost 3-fold greater than previous estimates, undoubtedly due to the far larger number of observations, the associated expansion of geographic scope and channel types, and the inclusionhigher temporal resolution of temporally resolved data.the data. For any sampling effort, the standard deviation increases with increasing sample size, but eventually reaches a plateau that indicates a sample size sufficient to capture the true population variability. It is not yet clear if the sample sizes are sufficient to capture the true global-scale variability of fluvial concentrations and fluxes, and future database updates couldshould be used to examine this possibilityrelationship.

Despite the slight lowering of median values compared to previous estimates, supersaturated concentrations and positive fluxes are the norm for CH_4 as well as for CO_2 and N_2O. However, it is likely that CH_4 concentrations and fluxes below detection limits (BDLs) are under-reported, as is common with environmental data in general (Stow et al., 2018), so these latest estimatescurrent averages may still be slight overestimations of true population medians. Even given the modest number of zero or undetectable CH_4 concentrations in GriMeDB (<2.5%), decisions about handling BDLs can have a small but detectable effect on the estimation of global averages. For example, if these observations are excluded, median CH_4 concentrations for all other observations increases from 1.49 to 1.51 µmol L^{-1}. If we keep all of these observations and assign them a value of zero (an unlikely scenario, but used here to provide a lower limit for this example), then the overall median declines to 1.46

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µmol L^{-1}. Although these differences are~~this difference is~~ relatively small, it would likely be consequential for upscaling estimates. At a minimum, we urge GRiMeDB users to be aware of how these values are handled and encourage future researchers to determine and report detection limits and include samples that fall below these limits in their results.

A goal of assembling GRiMeDB was to centralize CH_4 data to foster future research efforts. To this end, we ~~also~~ included information about habitat conditions that allows the exploration of relationships between CH_4 and potential explanatory variables and covariates. To demonstrate this opportunity, we provided a limited number of ~~graphic~~ examples of CH_4 versus variables that have been identified as potential predictors or drivers of CH_4 production, concentration, or flux (Figs. 12-14~~11-13~~), and these plots suggest both the presence and absence of relationships. For example, increasing CH_4 concentrations have been associated with low or decreasing dissolved oxygen and/or increasing organic carbon (e.g., Borges et al., 2018; Jin et al. 2018; Begum et al., 2021) and these relationships are recognizable for concentration but ambiguous for flux across the entirety of the GRiMeDB dataset. Similarly, increased CH_4 production and emissions tend to be elevated in nutrient-rich (eutrophication) lakes (DelSontro et al., 2018) and polluted rivers (Rajkumar et al. 2008; Ho et al., 2022), consistent with positive relationships between CH_4 flux and TN and TP. However, nutrient enrichment in rivers often occurs concurrently with fine sediment and organic matter input; thus it remains to be determined if positive relationships in Figs. 13g and 13h are correlative or reflect a causal mechanism. Finally~~data. Similarly~~, increases in discharge have been linked to declines in gas concentration, likely due to source limitation (i.e., dilution) of terrestrial supply (Aho et al., 2021; Gómez- Gómez-Gener et al., 2021a) and/or greater water turbulence, which increases gas exchange and thus reduces~~in turn can reduce~~ supersaturated CH_4 stocks (Billett and Harvey, 2013; Kokic et al. 2018). This relationship is not obvious when all data were considered en masse, but became more apparent when examining within-site dynamics. In contrast to these three confirmatory examples, although latitude and channel size have also been identified as determinants of CH_4 concentrations or used to extrapolate site-specific gas measurements to larger (even global) scales (e.g., Bastviken et al., 2011; Li et al., 2020; Rosentreter et al., 2021), evidence for such relationships is absent~~not apparent~~ from our analysis. Further, even for the former examples that indicated relationships between CH_4 concentration and DO, DOC, or discharge, there is substantial variability present in these relationships, the strength of these predictors is likely to vary across scales, and they explain~~explains~~ little of the variability for diffusive fluxes. In short, substantial opportunities exist to identify multivariate relationships between different predictors and CH_4 concentrations and fluxes across different scales, and pursuit of these opportunities should be improved by the substantial increase in data for both gases and potential predictor variables.

The disproportionate contribution of streams and rivers to atmospheric inputs together with the utility of CH_4 as an indicator of anthropogenic influences on drainage systems have inspired several studies that focus on fluvial habitats that are expected to have high concentrations and fluxes. Many of these 'methane hunting' studies have demonstrated significant increases in CH_4 concentrations and/or fluxes associated with phenomena such as point source inputs (Alshboul et al., 2016)~~,~~ ditch and canal construction (Peacock et al., 2021)~~,~~ oil and gas extraction (Woda et al. 2020)~~,~~ or passage through wetlands (Taillardat

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et al., 2022).~~.~~ Such signals persist at the global scale (Fig. 15~~14~~), highlighting widespread human enhancement of CH_4 emissions from lotic ecosystems. Not all targeted sites are CH_4-rich however. Low concentrations in glacial outflows (GT) likely reflect the effects of cold temperatures and/or low organic carbon availability (Crawford et al., 2015; Burns et al., 2018) while low values at floodplain (FP) sites may be attributable to their more characteristically lentic conditions, which favor~~allows~~ higher rates of CH_4 oxidation in the water column. Indeed, oxidation has been shown to represent a significant CH_4 sink in floodplain lakes associated with the Amazon River (Barbosa et al., 2018) and most of the FP sites in GRiMeDB are part of the Amazon system.

As noted in Sect. 3.4, the availability of supporting information is inconsistent, as, for example, only ~25% of data sources provided data~~information~~ on channel order or basin size. However, ~~the growing availability of~~ open-access regional and global geospatial datasets that provide information about site characteristics (e.g., Linke et al., 2019~~;~~, Yang et al., 2020) have~~has~~ increased rapidly in the past decade, to the benefit of analyses seeking to link landscape attributes to CH_4 distribution aming sites.~~.~~ Recent upscaling efforts analyses (Rosentreter et al., 2021; Liu et al., 2022; Rocher-Ros et al., in review) have, for example, capitalized on~~benefited from~~ improved estimates of the surface area of world streams and rivers (Allen and Pavelsky, 2018; Yang et al., 2020), while the diverse datasets in HydroSHEDS (Linke et al., 2019) allowed us to evaluate the global representativeness of GRiMeDB sites. As new global-scale datasets become available and become more spatially resolved, we anticipate that their pairing with GRiMeDB data will result in significant improvements in the strength and certainty of data assimilation models, regional to ~~continental and~~ global-scale analyses of~~models explaining~~ CH_4 distribution and drivers, and quantification of~~quantifying~~ fluvial emissions to the atmosphere.

5. Data and code availability

GRiMeDB and its associated metadata are available from the the Environmental Data Initiative (Stanley et al., 2023)~~:2022)~~: https://doi.org/10.6073/pasta/f48cdb77282598052349e969920356ef.

~~https://doi.org/10.6073/pasta/b7d1fba4f9a3e365c9861ac3b58b4a90~~

~~Code used for unit conversions, spatial analyses, and general data analysis and visualization will be available from EDI.~~

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6. Conclusion

The data gathered in GRiMeDB highlight~~highlights~~ many new opportunities, both through analysis of CH_4 and supporting data in the database, and by revealing gaps that currently exist across fluvial CH_4 studies. The most conspicuous data limitations include deficits in measurements of non-diffusive flux pathways and underrepresentation of sites from~~in underrepresented~~ arid, tropical, and arctic biomes. Challenges associated with quantifying ebullition discussed earlier~~above~~ also emphasize the

need for more intercomparisons among ~~the various~~ flux methodologies.~~methods.~~ Regardless of pathway, flux is a difficult process to quantify and can be highly sensitive to methods or gas exchange model choices, yet ~~and~~ there are few ~~methodological~~ comparisons (such as ~~available to inform these decisions (~~Raymond et al., 2012; Lorke et al., 2015) available to inform these decisions.~~).~~ Finally, we highlight that the expansion of GHG data ~~world streams and rivers~~ over the past decade has proceeded largely across spatial rather than temporal dimensions. While this expansion has vastly improved the geographic representativeness of the data, long-term datasets are rare despite their power for generating ecological understanding and informing policy/management in the face of environmental change (Hughes et al., 2017). GHG´s~~Unfortunately, GHG´s, particularly CH_4 and N_2O,~~ are rarely included as routine components of water quality monitoring programs. Thus, we emphasize the compelling need to establish such sampling efforts and perpetuate those few that do exist.

Given the rapid growth in both research interest and data in fluvial GHG dynamics, we imagine future updates and expansion of GRiMeDB and we welcome datasets and associated research products (e.g., theses, journal publications, reports, etc.). To facilitate the data acquisition and updating process, a dowloadable spreadsheet template and detailed information about its use and submission are available at https://stanley.limnology.wisc.edu/GRiMe. Regardless of database updates, we recommend that the minimum basic information to collect along with GHG data that would be most valuable for later analyses include: well-resolved site location data (latitude and longitude); information about site size (Strahler order and/or basin size at the sampling site), disturbance or modification relevant to GHGs (e.g., categories listed in Table 1); specific sample dates and times; discharge, dissolved oxygen, and temperature at the time of sample collection; and clear information about units and method(s) used to measure gas flux. Finally, we strongly encourage data package (sensu Gries et al., 2022) publication in a trustworthy public data repository such as the Environmental Data Initiative that requires metadata to meet FAIR data principles and increase data findability, accessibility, and re-use (Wilkinson et al., 2016).

Despite highlighting ~~these~~ areas of data limitation ~~in the field~~, it is important to underscore the opportunities that the growth in GHG data availability- especially of CH_4 data- now provide. Assembly of GRiMeDB was motivated by the goal of having a centralized, standardized resource to facilitate further studies of CH_4 pattern and process in flowing water systems. Our strategy in developing this database was to maximize opportunities for identifying patterns and relationships involving this gas in future analyses. Past difficulties with such efforts may well be a product of the common practice of averaging values over time or among sites and/or of including non-fluvial sites in analyses. Thus, we carefully documented the data and resolved observations to individual sites and dates whenever possible to match the pronounced spatial and temporal variance of this gas. Similarly, while we included a range of habitat types in GRiMeDB, unconventional or targeted sites are easily identifiable. Further, we carefully examined sites to ensure that they ~~were not subject to impounding effects of a dam or~~ were not situated within reservoirs/impoundments or~~in~~ estuaries where distinct processes such as methane oxidation, tidal cycles, or ~~and~~ elevated sulphate reductions~~concentrations~~ may obscure or overtake relationships present in inland flowing water systems. Thus, we are optimistic that analysis of GriMeDB data by itself, or in concert with other complementary datasets, will provide new and

unprecedented opportunities to examine relationships between CH_4 and environmental drivers or correlates, as well as providing broad contextual information for site-based studies of fluvial carbon and GHG dynamics.

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Appendix A. GRiMeDB tables and variables

Table A1. Column titles and description of their content for the GRiMeDB Sources Table.

Column Title	Description
Title	Title of data source.
Author	Lead author last name
Source	Identity of the outlet for the data (e.g., journal, data repository, agency that presented the data). For titles with published papers paired with published datasets, the journal is listed in this column
Pub_year	Year of publication, data release, or acquisition of an unpublished dataset
Source_ID	Unique data source identifier
Additional_data	"Yes" in this column indicates that additional data were acquired directly from the author for any field. Additions are described in the Comments field
Comments	Additional information or clarification about the data source
Paper_DOI	DOI or hyperlink for journal article or other publication based on the CH_4 data
Data_DOI_primary	DOI or hyperlink for CH_4 data posted in a data repository
Data_DOI_supporting	DOI or hyperlink for separate datasets providing supporting data

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Table A2. Column titles and content description for the GRiMeDB Sites Table.

Column Title	Definition
Source_ID	Unique data source identifier from the Sources Table
Site_ID	Unique site identifier
Site_Name	Unique site name
Stream_Name	Stream or river name; taken or modified from the data source or generated *de novo* when a name was not specified in the data source
Aggregated	Yes or No; "Yes" if CH_4 data entered are averages from >1 site
N_sites_aggregated	Number of sites that were averaged for aggregated sites
Basin_Region	Name of the larger drainage basin or region that contains the site. This information is included to facilitate site grouping during data analysis
Latitude	Decimal degrees, WGS84 ensemble: EPSG:4326 coordinate system
Longitude	Decimal degrees, WGS84 ensemble: EPSG:4326 coordinate system
Elevation_m	Reported meters above sea level
Slope_m_per_m	Reported channel slope expressed as m m^{-1}
Strahler_order	Reported Strahler stream order
Basin_size_km^2	Reported basin size in square kilometers
Channel_type	Codes denoting distinct site or channel attributes or presence of specified conditions. See Table 1 for categories and their definitions
Latitude_snapped	Latitude in decimal degrees for site location after snapping to the closest channel for elevation determination
Longitude_snapped	Longitude in decimal degrees for site location after snapping to the closest channel for elevation determination
Elevation_estimated_m	Elevation (meters above sea level) calculated from the DEM. See Sect. 2.2 for details
Comments	Additional information or clarification about the site source

Table A3. Column titles and definitions for the GRiMeDB Concentration Table

Column Title	Definition
Source_ID	Unique paper identifier from the Sources Table
Site_ID	Unique site~~paper~~ identifier from the Sites Table
Site_Name	Unique site name from the Sites Table
Conc_Name	Unique name for the sampling event at the site; same as Flux_Name in the Flux~~Fluxes~~ Table if both concentration and flux data for the same site-date combination are available
Date_start	First sampling date
Date_end	Last sampling date; this is the same date as the Date_start if data are not aggregated over time
Aggregated_Space	Yes or No; "Yes" if CH_4 data entered are averages from >1 site
Aggregated_Time	Yes or No; "Yes: if CH_4 data entered are averages from >1 date
FluxYesNo	Yes or No; "Yes" if there is a corresponding flux measurement associated with this site-date combination
SampleCount	Number of samples or observations corresponding to the mean or median concentration
CH4min	Minimum measured CH_4 concentration in $\mu mol\ L^{-1}$ if data are aggregated spatially or temporally, has multiple within-day measurements (e.g., a diel study), or are from a data-dense spatial study
CH4max	Maximum measured CH_4 concentration in $\mu mol\ L^{-1}$ if data are aggregated spatially or temporally, has multiple within-day measurements (e.g., a diel study), or are from a data-dense spatial study
CH4mean	Mean or sole reported CH_4 concentration in $\mu mol\ L^{-1}$ for the sampling event
CH4_SD	Standard deviation of the mean CH_4 concentration
CH4median	Median CH_4 concentration in $\mu mol\ L^{-1}$
CO2min	Minimum measured CO_2 concentration in $\mu mol\ L^{-1}$ if data are aggregated spatially or temporally, has multiple within-day measurements (e.g., a diel study), or are from a data-dense spatial study
CO2max	Maximum measured CO_2 concentration in $\mu mol\ L^{-1}$ if data are aggregated spatially or temporally, has multiple within-day measurements (e.g., a diel study), or are from a data-dense spatial study
CO2mean	Mean or sole reported CO_2 concentration in $\mu mol\ L^{-1}$ for the sampling event
CO2_SD	Standard deviation of the mean concentration

~~CO2median~~**~~Table A3. Continued~~**	~~Median CO_2 concentration~~
~~N2Omin~~CO2median	~~Minimum measured N_2O concentration if data are aggregated or temporally, has multiple within day measurements (e.g., a diel study), or are from a data-dense spatial study~~ Median CO_2 concentration in µmol L^{-1}
~~**Table A3. Continued**~~N2Omin	Minimum measured N_2O concentration in µmol L^{-1} if data are aggregated spatially or temporally, has multiple within-day measurements (e.g., a diel study), or are from a data-dense spatial study
N2Omax	Maximum measured N_2O concentration in µmol L^{-1} if data are aggregated spatially or temporally, has multiple within-day measurements (e.g., a diel study), or are from a data-dense spatial study
N2Omean	Mean or sole reported N_2O concentration in µmol L^{-1} for the concentration for the sampling event
N2O_SD	Standard deviation of the mean N_2O concentration
N2Omedian	Median N_2O concentration in µmol L^{-1}
WaterTemp_degC	Water temperature in degrees C measured concurrently with CH_4
WaterTemp_degC _estimated	Estimated water temperature in degrees C. This field was populated only for cases in which temperature was needed for gas unit conversion. Most estimates were based on temperatures from adjacent sites, averaging temperatures from prior and proceeding sample dates, or from an adjacent day of the year but from another year.
Cond_uScm	Specific conductance in µS cm^{-1}
pH	pH
DO_mgL	Dissolved oxygen in mg L^{-1}
DO_percentsat	Percent saturation of dissolved oxygen
Q	Discharge in m^3 s^{-1} measured at the time of sample collection
NO3	NO_3 or NO_2+NO_3 concentration in µmol L^{-1} measured concurrently with CH_4
NH4	NH_4 concentration in µmol L^{-1} measured concurrently with CH_4
TN	TN or TDN concentration in µmol L^{-1} measured concurrently with CH_4
SRP	SRP or PO_4 concentration in µmol L^{-1} measured concurrently with CH_4
TP	TP or TDP concentration in µmol L^{-1} measured concurrently with CH_4
DOC	DOC or TOC concentration in µmol L^{-1} measured concurrently with CH_4
Comments	Any additional relevant information regarding data

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new_CH4_unit	Current common units for all CH_4 concentrations

Table A3. Continued

new_CO2_unit	Current common units for all CO_2 concentrations
new_N2O_unit	Current common units for all N_2O concentrations
new_NO3_unit	Current common units for all NO_3 or NO_2+NO_3 concentrations
new_NH4_unit	Current common units for all NH_4 concentrations
new_TN_unit	Current common units for all TN or TDN concentrations

~~**Table A3. Continued**~~

new_SRP_unit	Current common units for all SRP or PO_4 concentrations
new_TP_unit	Current common units for all TP or TDP concentrations
new_DOC_unit	Current common units for all DOC or TOC concentrations
new_Q_unit	Current common units for all discharge measurements
orig_CH4_unit	Original units for CH_4 concentration
orig_CO2_unit	Original units for CO_2 concentration
orig_N2O_unit	Original units for N_2O concentration
orig_NO3_unit	Original units for NO_3 or NO_2+NO_3 concentration
orig_NH4_unit	Original units for NH_4 concentration
orig_TN_unit	Original units for TN concentration
orig_SRP_unit	Original units for SRP or PO_4 concentration
orig_TP_unit	Original units for TP concentration
orig_DOC_unit	Original units for DOC concentration
orig_Q_unit	Original units of discharge

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Table A4. Column titles and definitions for the GRiMeDB Flux Table

Column Title	Definition
Source_ID	Unique paper identifier from the Sources Table
Site_ID	Unique site~~paper~~ identifier from the Sites Table
Site_Name	Unique site name from the Sites Table
Flux_Name	Unique name for the sampling event at the site; same as Conc_Name in the Concentration~~Concentrations~~ Table if both concentration and flux data for the same site-date combination are available
Date_start	First sampling date
Date_end	Last sampling date; this is the same date as the Date_start if data are not aggregated over time
Aggregated_Space	Yes or No; "Yes" if CH_4 data entered are averages from >1 site
Aggregated_Time	Yes or No; "Yes: if CH_4 data entered are averages from >1 date
Diffusive_CH4_Flux_Min	Minimum measured CH_4 diffusive flux in mm m^{-2} d^{-1} if data are aggregated or are from diel or data-dense spatial studies
Diffusive_CH4_Flux_Max	Maximum measured CH_4 diffusive flux in mm m^{-2} d^{-1} if data are aggregated or are from diel or data-dense spatial studies
Diffusive_CH4_Flux_Mean	Mean or sole reported CH_4 diffusive flux in mm m^{-2} d^{-1} for the sampling event
Diffusive_CH4_Flux_SD	Standard deviation of the mean CH_4 diffusive flux
Diffusive_CH4_Flux_Median	Median CH_4 diffusive flux in mm m^{-2} d^{-1}
SampleCount_Diffusive	Number of samples or observations corresponding to the mean or median diffusive CH_4 flux
Diff_Method	Methodological category used to measure diffusive gas flux. Categories (with brief explanations in italics) are: chamber (unspecified)- unspecified response *use of an unspecified type of chamber (suspended, tethered, or free-floating) and pattern of change gas concentration over time during flux measurements is also not specified* chamber (unspecified)- linear response *unspecified type of chamber with a linear increase in chamber gas concentration over time or use of a linear model to calculate flux* suspended/tethered chamber-unspecified response

~~Table A4. Continued~~

	chamber is restrained to maintain its position and not float downstream during flux measurement suspended/tethered chamber- linear response floating chamber- unspecified response *chamber is unrestrained and is able to float downstream during flux measurement* floating chamber- linear response conc+k *diffusive flux calculated using the equation:* *flux = $k(C_w-C_{eq})$, where*

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k = gas exchange coefficient
C_w = CH_4 concentration measured in water
~~*Ceq = CH_4 concentration in water in equilibrium with the atmosphere*~~

~~other~~
~~*methods other than those described above*~~

Table A4. Continued.

	Ceq = CH_4 concentration in water in equilibrium with the atmosphere
	other
	methods other than those described above
Eb_CH4_Flux_Min	Minimum measured CH_4 ebullitive flux in mm m^{-2} d^{-1} if data are aggregated or are from diel or data-dense spatial studies
Eb_CH4_Flux_Max	Maximum measured CH_4 ebullitive flux in mm m^{-2} d^{-1} if data are aggregated or are from diel or data-dense spatial studies
Eb_CH4_Flux_Mean	Mean or sole reported CH_4 ebullitive flux in mm m^{-2} d^{-1} for the sampling event
Eb_CH4_Flux_SD	Standard deviation of the mean CH_4 ebullitive flux
Eb_CH4_Flux_Median	Median CH_4 ebullition flux in mm m^{-2} d^{-1}
SampleCount_Eb	Number of samples or observations corresponding to the mean or median ebullitive CH_4 flux
~~Eb_Method~~	~~Methodological category used to measure ebullitive gas flux. Categories (with brief explanations in italics) are:~~
	~~chamber minus conc+k~~
	~~*ebullition calculated as chamber-measured flux (assumed to be total CH_4 flux) minus diffusive flux calculated from the 'conc+k' method*~~
	~~bubble trap + bubble analysis~~
	~~*gas released by ebullition captured in traps to quantify total gas volume; volume data combined with measurement of CH_4 content of recently collected bubbles*~~

~~**Table A4. Continued**~~

	echosounder + bubble analysis
	gas bubble volume determined using echosounder and combined with CH_4 content of recently collected bubbles
	departure from linear increase during measurement
	non-linear change in gas concentrations during chamber-based flux measurements taken as evidence of ebullition; various approaches used to quantify ebullition from these departures
	other
	methods other than those described above
Total_CH4_Flux_Min	Minimum measured total CH_4 flux in mm m^{-2} d^{-1}
Total_CH4_Flux_Max	Maximum measured total CH_4 flux in mm m^{-2} d^{-1}
Total_CH4_Flux_Mean	Mean or sole reported total CH_4 flux for the sampling event in mm m^{-2} d^{-1}
Total_CH4_Flux_SD	Standard deviation of the mean total CH_4 flux
Total_CH4_Flux_Median	Median measured total CH_4 flux in mm m^{-2} d^{-1}

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Total_Method	Methodological category used to measure total CH_4 flux. Categories (with brief explanations in italics) are: conc+k and ebullition *total flux calculated as the sum of separate measurements of diffusion determined by the conc+k method plus ebullition determined from the bubble trap or echosounder approach combined with bubble CH_4 analysis* floating chamber *free-floating chamber is assumed to capture diffusive flux and ebullitive flux (if present)* suspended/tethered chamber *suspended or tethered chamber is assumed to capture diffusive flux and ebullitive flux (if present)* chamber and ebullition *total flux calculated as the sum of separate measurements of diffusion determined using a floating or suspended/tethered chamber plus ebullition determined from the bubble trap or echosounder approach combined with bubble CH_4 analysis* ~~mass balance~~ ~~*total flux represents the difference between all measured inputs to a reach (e.g., dissolved CH_4 from upstream flow, groundwater discharge, and methanogenesis) minus all outputs other than efflux to the atmosphere (e.g., downstream export, methane oxidation)*~~

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Table A4. Continued

	mass balance *total flux represents the difference between all measured inputs to a reach (e.g., dissolved CH_4 from upstream flow, groundwater discharge, and methanogenesis) minus all outputs other than efflux to the atmosphere (e.g., downstream export, methane oxidation)* other *methods other than those described above*
CO2_Flux_Min	Minimum measured CO_2 flux in mm m^{-2} d^{-1} if data are aggregated or are from diel or data-dense spatial studies
CO2_Flux_Max	Maximum measured CO_2 flux in mm m^{-2} d^{-1} if data are aggregated or are from diel or data-dense spatial studies
CO2_Flux_Mean	Mean or sole reported CO_2 diffusive flux in mm m^{-2} d^{-1} for the sampling event
CO2_Flux_SD	Standard deviation of the mean CO_2 flux
CO2_Flux_Median	Median CO_2 flux in mm m^{-2} d^{-1}
N2O_Flux_Min	Minimum measured N_2O flux in mm m^{-2} d^{-1} if data are aggregated or are from diel or data-dense spatial studies
N2O_Flux_Max	Maximum measured N_2O flux in mm m^{-2} d^{-1} if data are aggregated or are from diel or data-dense spatial studies
N2O_Flux_Mean	Mean or sole reported N_2O diffusive flux in mm m^{-2} d^{-1} for the sampling event
N2O_Flux_Stddev	Standard deviation of the mean N_2O flux
N2O_Flux_Median	Median N_2O flux in mm m^{-2} d^{-1}

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k_Method	Methodological category used for estimating the gas exchange coefficient, *k*, Categories (with brief explanations in italics) are: physical model *k calculated using equations based on physical variables such as channel slope, water velocity, etc.* chamber + conc *k determined by chamber-based measurements of flux, dissolved gas concentration, and re-arrangement of the flux equation* $flux = k(C_w - C_{eq})$ *to solve for k. Typically, these measurements are made for* CO_2*, and then* k_{CO2} *is converted to* k_{CH4} tracer addition *paired conservative and gas tracer additions used to calculate k from concentration declines along a stream reach* assigned k value *use of k values from other dates or sites in the same study or k values considered to be characteristic of the site* other *methods other than those described above*

~~**Table A4. Continued**~~

	unknown *method to determine k is not described*
k_ref	k method citation reported in the data source
Comments	Any additional relevant information regarding data entered in this row
new_Diffusive_Flux_unit	Current common units for all diffusive CH_4 flux data
new_Eb_CH4_Flux_unit	Current common units for all ebullitive CH_4 flux data

Table A4. Continued.

new_Total_Flux_unit	Current common units for all total CH_4 flux data
new_CO2_Flux_unit	Current common units for all CO_2 flux data
new_N2O_Flux_unit	Current common units for all N_2O flux data
orig_Diffusive_Flux_unit	Original units for diffusive CH_4 flux
orig_Eb_CH4_Flux_unit	Original units for ebullitive CH_4 flux used
orig_Total_Flux_unit	Original units for total CH_4 flux
orig_CO2_Flux_unit	Original units for CO_2 flux
orig_N2O_Flux_unit	Original units for N_2O flux

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Appendix B. Citations for data sources in GRiMeDB, including citations . ~~Citations are not provided~~ for unpublished datasets. Dates for unpublished dataset correspond to the year the data were provided by data authors.

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Supplement.

The supplement related to this article is available at https://doi.org/10.5194/essd-2022-346-supplement

Author contributions.

EHS conceived of the project idea, and led data entry, manuscript preparation, and data curation. LCL developed the code used for unit conversions, was responsible for data conversion and QA/QC, and contributed to data visualization, data analysis, and code curation. GRR was responsible for spatial analyses, and contributed to data visualization, code curation, and manuscript preparation. The structure and composition of the manuscript were resulted~~the result~~ from collaborative discussions among EHS, GRR, LCL, NJC, SKO, and RAS. All authors contributed to data acquisition, data entry, data checking, and substantial manuscript revising and editing. Any use of trade, firm, or product names is for descriptive purposes only and does not imply endorsement by the U.S. Government.

Competing Interests.

The authors declare that they have no conflict of interest.

Acknowledgements.

We are grateful to all the authors who generously shared data or made their data public; our science is better or this transparency. We are also grateful to several of these investigators for patiently and thoughtfully responding to emails from E. Stanley. Corinna Gries provided advice and assistance with data publication. Stanley thanks K. Forest and K. McMahon for guidance~~support~~ during manuscript preparation. Great thanks to M. W. Diebel, S. N.Topp, Y. Yao, K. Castro-Morales, and B. Deemer for their thoughtful and detailed comments that improved this paper.

Financial Support.

Emily H. Stanley acknowledges support from the National Science Foundation #DEB-2025982, NTL LTER. Nora J. Casson was supported in part by the Canada Research Chair program. Gerard Rocher-Ros was supported by the Swedish Research Council (2021-06667) and the Department of Ecology and Environmental Science, Umeå University. Marcus B. Wallin was supported by the Swedish Research Council's VR (2021-04058) and Formas (2019-01105). Any use of trade, firm, or product names is for descriptive purposes only and does not imply endorsement by the U.S. Government.

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