



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

- 20 effort has been put into identifying patterns and drivers of CH₄ 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 CH₄ concentrations and fluxes and by limited data availability across diverse habitats and physicochemical conditions. To address these challenges, we present the first comprehensive database of CH₄ concentrations and fluxes for fluvial ecosystems along with broadly relevant and concurrent physical and chemical data. The Global River
- 25 Methane database (GriMeDB; <u>https://doi.org/10.6073/pasta/b7d1fba4f9a3e365c9861ac3b58b4a90</u>) includes 24,024 records of CH₄ concentration and 8,205 flux measurements from 5,037 unique sites that were extracted from publications, reports, data repositories, and other outlets published between 1973 and 2021. GriMeDB also includes 17,655 and 8,409 concurrent measurements of concentrations and 4,444 and 1,521 of fluxes for CO₂ and nitrous oxide (N₂O) respectively. Most observations are date-specific (i.e., not site averages) and many are supported by data for 12 physicochemical variables and 6
- 30 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 a 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 CH₄ concentration (0.20 µmol L⁻¹) and diffusive flux (0.44 mmol m⁻² d⁻¹) in GRiMeDB are lower than estimates from past, site-averaged compilations, although
- 35 ranges and standard deviations are greater from this larger and more temporally-resolved database. Available flux data are 35 dominated by diffusive measurements despite the recognized importance of ebullitive and plant-mediated CH₄ fluxes. Despite 36 these limitations, GriMeDB provides a comprehensive and cohesive resource for examining relationships between CH₄ and 37 environmental drivers, estimating the contribution of fluvial ecosystems to CH₄ emissions, and to contextualize site-based 38 investigations.

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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

45 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 CH₄ dynamics in aquatic environments in general, and fluvial ecosystems in particular. These studies reveal widespread supersaturation of CH₄ in running waters that underlies their larger than expected contribution to the atmospheric pool (Stanley et al., 2016).



- 50 Efforts to quantify fluvial CH₄ 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 CH₄ are often unevenly distributed over space and time within drainage systems and, as a result, concentrations can vary over 3-4 orders of magnitude over short time periods or relatively small spatial extents (e.g., Anthony et al., 2012; Crawford et al., 2017; Bretz et al., 2021; Robison et al., 2021). Similarly, several drivers or predictors of CH₄ have been
- 55 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 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
- 60 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
- 70 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 CH₄, to test hypotheses related to greenhouse gas dynamics, and 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. Data included in GRiMeDB were gathered from scientific journals,



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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, NERC Environmental Information Data Centre, Arctic Data Center, and PANGAEA) using the keywords: methane and stream* or river* or ditch* or canal*, and searches were repeated numerous times between 2018 and December 2021 for completeness. We also used informal 'word of mouth' approaches to discover additional, often unpublished data sets.

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, dam spillways, 90 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 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 95 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 CH₄ data source, and basic bibliographic information for the data source (*Title, Author, Source*, publication year [*Pub year*], and digital object identifiers [DOI] 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. 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



105 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.

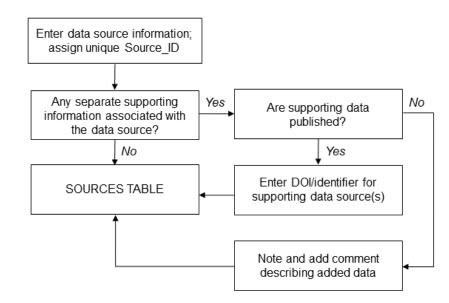


Figure 1: Workflow for entering data into the Sources Table of GRiMeDB.

110 2.2 Site Table

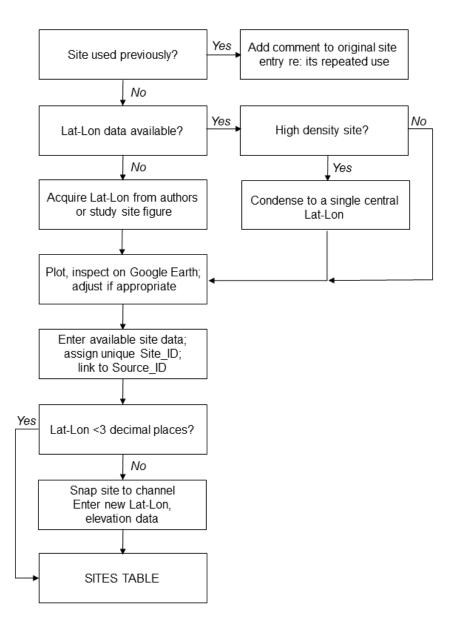
The Sites Table reports basic information on attributes for all sites where CH_4 was sampled. Each site has a unique identification code (*Site_ID*) and name (usually taken directly from the data source) and is linked to the Sources Table via the *Source_ID* (see Table A2 for detailed descriptions of all columns in the Sites Table). What composes a site varies among data sources and includes discrete sampling points, discrete study reaches, and aggregations of points and/or reaches across larger areas such as a drainage

- 115 basin. Because gas data for sites in this third category are averages from locations that may vary with respect to land use, channel order, slope, etc., we included fields to indicate if a site was 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. 2) 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
- 120 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 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
- 125 dates were treated as a single site. 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.



130 Figure 2: 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. 2). Latitude and longitude coordinates were available for most sites; however, in

cells), we assumed the site was in a stream channel.





several cases, location information was acquired from authors or estimated from study site figures using Google Earth (© Google Earth 2020). Sites were plotted on Google Earth and inspected (Fig. 2) to identify and correct data errors. If a site's 135 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). 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 140 coordinates to the nearest stream channel 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) 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

145 accumulation less than 10 cells (indicating that 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

- 150 Many studies of CH₄ 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 CH₄ (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 CH₄, 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
- 155 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 CH₄ 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
- 160 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 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 (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
- 165 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



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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 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.

175 **2.3 Concentrations and Fluxes Tables**

The Concentrations and Fluxes Tables contain the primary gas data central to GRiMeDB, and the 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 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

- 180 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
- 185 dataset, and average slope = 0.998, average R²=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. 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

195 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 CH₄ 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
СН	Channelized; a channel that has long straight-line sections 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 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

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 -9999999. 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.

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The Fluxes 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), 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*)) 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.

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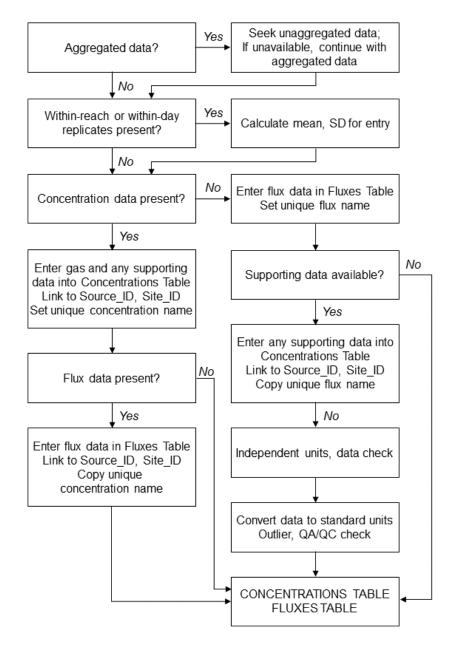
The GRiMeDB Concentrations Table includes physicochemical measurements in support of concentration and flux observations (Fig. 3, Table A3). Availability of this supplemental information varied widely among data sources, and was limited to data collected concurrently with gas samples. For data sources with gas fluxes and physicochemical data but not gas concentrations, we created rows in the Concentrations 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 similar times (e.g., averages of temperature from the prior and subsequent dates, or from the same month in a prior year).

- 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 Concentrations and Fluxes 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%), we also used the global average CH₄, CO₂, and the N₂O
- 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.







240 Figure 3: Workflow for entering and checking data for the GRiMeDB Concentrations and 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

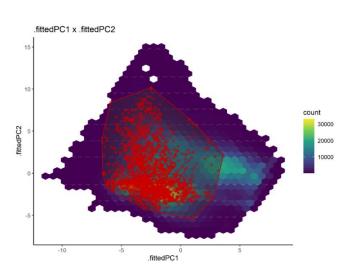
245 HydroSHEDS (Linke et al., 2019), which is a global hydrological network database that contains spatial data for a wide array





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 250 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. 4). Each HydroSHEDS subcatchment was then assigned a value of 1 or 0 if it fell within or outside the 255 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 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

260 reaches (average subcatchment area: 130 km²). Given the strong local controls on CH_4 concentrations and fluxes, interpretations from this analysis should be made with some caution.



265 Figure 4: Example of a representative 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.



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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. 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. 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 were performed in R (version 4.2, R Core Team 2021), using the "dplyr" package (version 1.0.7, Wickham et al., 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 CH₄ concentration and 8,205 CH₄ 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 of CO₂ and N₂O, 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 CH₄ concentrations and 80% of flux observations became available after 2015 (the year of publication of MethDB; Fig. 5). This growth in data availability has occurred predominantly along the
- 290 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. 5). 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
- 295 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, unpublished).





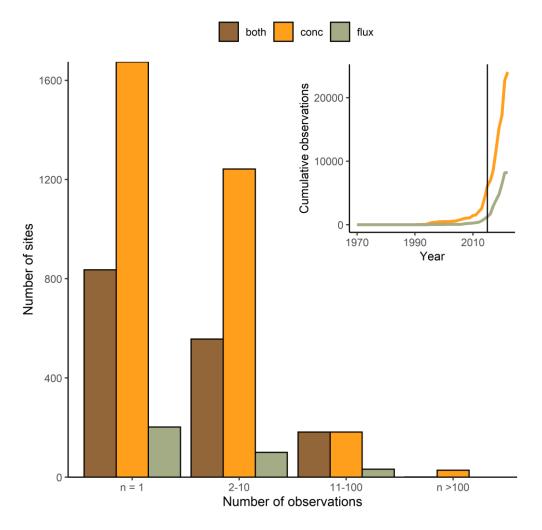


Figure 5: Distribution of the number of observations per site. Brown bars indicate sites with both concentration and flux observations. Orange and green bars show sites with only concentration and only flux observations, respectively. Inset: Cumulative observations of CH₄ concentration and flux data based on the year of publication of the data source. The vertical line (2015) indicates the year of MethDB (Stanley et al., 2015) publication.

3.2 Spatial and temporal distribution of data

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Spatially, 40% of all sites and 52% of all CH₄ concentration observations are in North America, followed by Europe (25% of all sites and 26% of all CH₄ 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. 6a). 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. 6b). Areas that are poorly represented are characterized by very



- 310 low channel density associated with arid or polar climates as well as high altitude regions (Greenland, northern Canada, 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 26-28% of all sites (Table S2). For sites with these data, counts of observations decline with increasing stream order (Fig. 7) in a log-linear fashion ($R^2 = 0.92$ for 315 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, 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
- 320 ca. 10,000 km²).

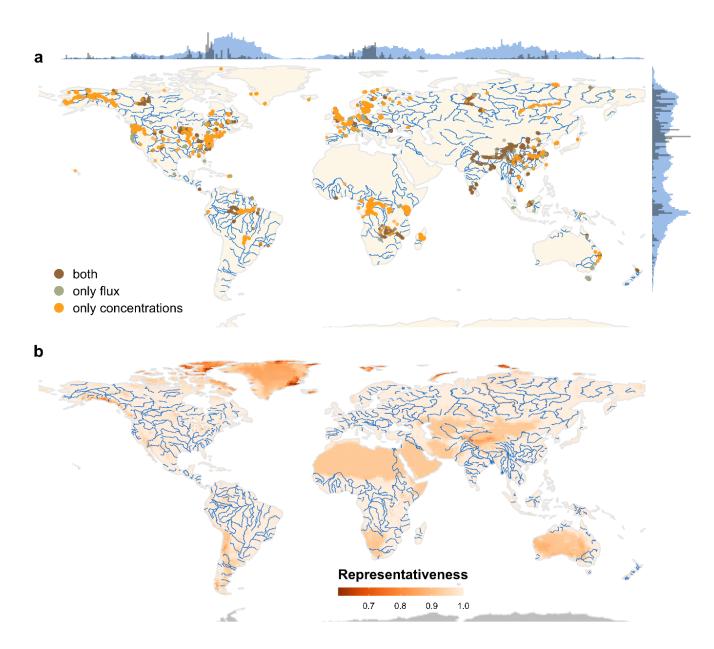
The distribution of observations among months illustrates seasonal sampling regimes dominated by summer sampling in northern (> 40°) and southern (< -20°) latitudes contrasted by even or erratic sampling at mid-latitudes (Fig. 8). Consistent with the lower representation of southern hemisphere rivers 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°

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70°, presumably due to pervasive ice and snow cover.







330 Figure 6: (a) Global distribution of 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 CH4 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 of the database based on a wide array of biological, physical, hydrological and land cover variables (see Sect. 2.4 335 for details). Values close to 1 indicate a high representativeness, with only 4% of the global surface below a threshold of 0.9.





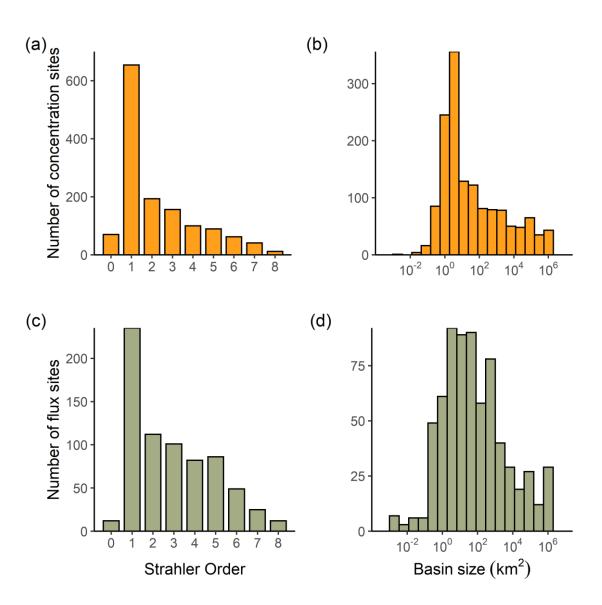
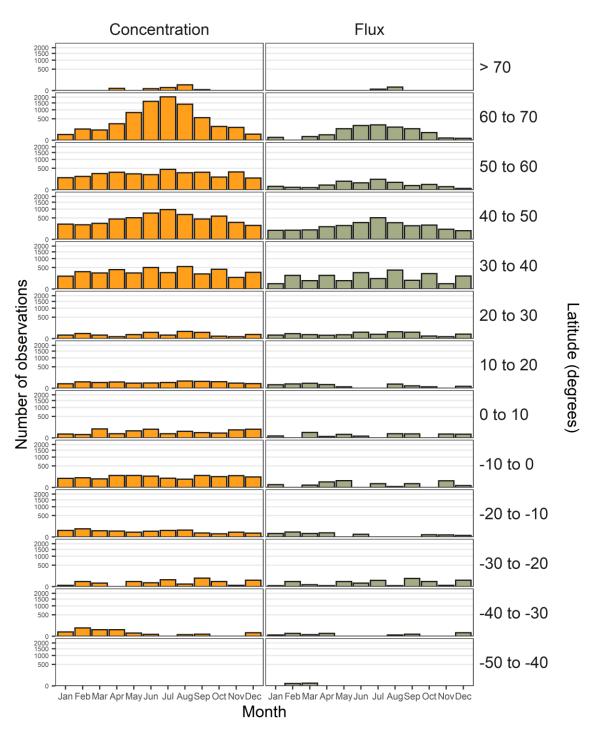


Figure 7: Number of sites with concentration (top) or diffusive flux (bottom) observations as a function of stream order (left) and basin size (right) for the subset of sites with channel order and/or basin size information.







340 Figure 8: Number of observations of concentration (left) and flux (right) by month for 10° latitude bands.



3.3 CH₄ flux methodology

- Records of CH₄ flux are dominated by diffusive flux measurements, which represent 85% of all flux values in the database, 345 with ebullition (8%) and total flux (7%) accounting for the remaining entries (Fig. 9). 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 = 3, 188). Several models have been employed for this calculation, as indicated by >25 350 different references for *k* model sources listed in CPiMoDP.
- 350 different references for k model sources listed in GRiMeDB.

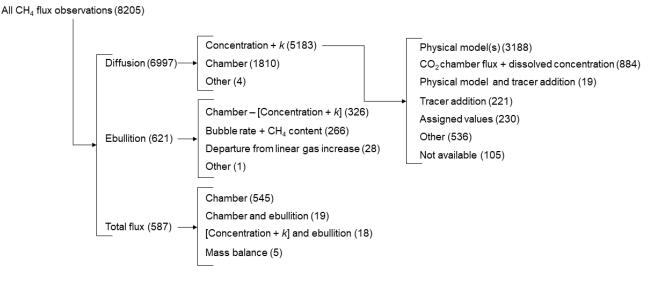


Fig. 9: Counts of CH4 flux observations by type (left), by major methodological categories for each pathway (middle), and for method
 type used to estimate the gas exchange coefficient k (right). For clarity, the chamber category includes all chamber types and patterns of gas increase in the chamber unless specified; more resolved methodological data are presented in the GRiMeDB Fluxes Table. See Table A4 for further details about category definitions.

3.4 Overview of concentration and flux data

360 Concentrations and fluxes of all three gases are characterized by log-normal distributions that range across several orders of magnitude (Fig. 10) and large coefficients of variation (CVs) for CH₄ and especially N₂O (Table 2). The vast majority (~95%) of CH₄ and CO₂ concentrations appear to be supersaturated, in contrast to N₂O concentrations in which 67% of observations were above this threshold. Reports of concentrations below detection are scarce for all gases, including N₂O (Table 2). For fluxes,





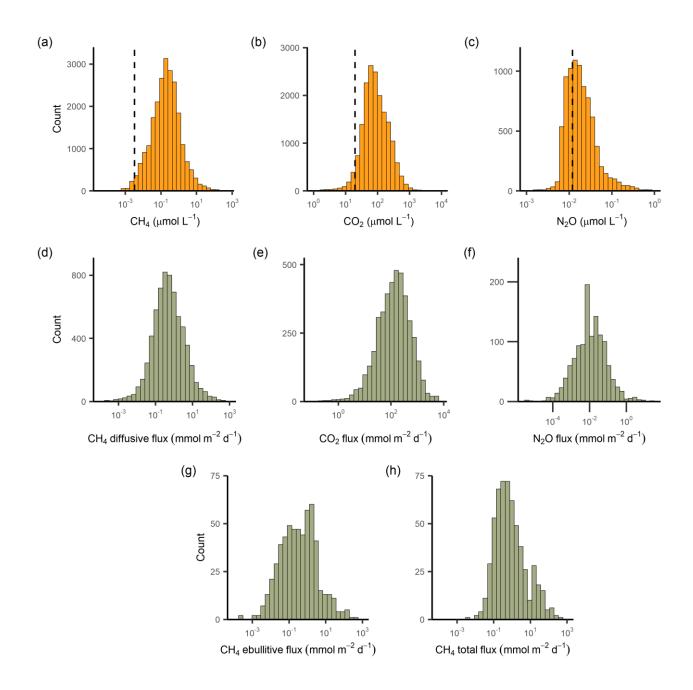


Figure 10: Histograms of gas concentrations and fluxes in GRiMeDB, excluding values reported as below detection or zero; counts of these values are reported in Table 3. Dashed vertical lines in the concentration histograms indicate the 100% saturation concentration based on the median estimated elevation (250 m) and water temperature for all sites and atmospheric concentrations of 1.83, 400, and 0.325 ppm for CH₄, CO₂, and N₂O, respectively.



the fraction of observations with zero, below detection, or negative fluxes (5, 5, and 19% for diffusive CH₄, CO₂, and N₂O fluxes, respectively). corresponded reasonably well with the frequency of subsaturated concentrations. At the other extreme, the highest CH₄ concentrations (> 200 μ mol 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).

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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. 11, Table S3). Although regressions were significant for latitude, latitude accounted for a very small percent of the variation in both concentration ($R^2 = 0.0025$) and flux ($R^2 = 0.004$) among sites. Similarly, concentration and flux among stream orders were significantly different for concentration (Kruskal-Wallis tests: Kruskal-Wallis $\chi^2 = 46.072$, P < 0.001) and marginally different for flux ($\chi^2 = 14.796$, P = 0.06). However, corrected pairwise comparisons (using the method of Benjamini and Hochberg, 1995) revealed no significant differences among orders for flux, and differences (P < 0.05) only between 7th order channels and all other orders, and between 6th vs 1st order sites for

concentration, indicating an absence of a consistent change in CH₄ magnitude across channel orders. 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.

Gas	Variable	Mean	Median	Max	Min	SD	CV	%BDL
Conce	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 ₂ O	0.058	0.017	32.9	0	0.602	1,042	0.59
Flux (Flux (mmol $m^{-2} d^{-1}$)							
	CH ₄ -diffusive	7.31	0.44	4,057	-136	86.4	1,182	
	CH ₄ -ebullitive	4.65	0.26	366	0	22.75	490	
	CH ₄ - total	7.62	0.62	366	-0.05	28.5	375	
	CO ₂	319	128	23,749	-1625	770	242	
	N ₂ O	0.082	0.008	31.3	-11.3	0.981	1,199	

Table 2. Summary statistics for CH4, CO₂, and N₂O concentrations and fluxes. The %BDL 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.





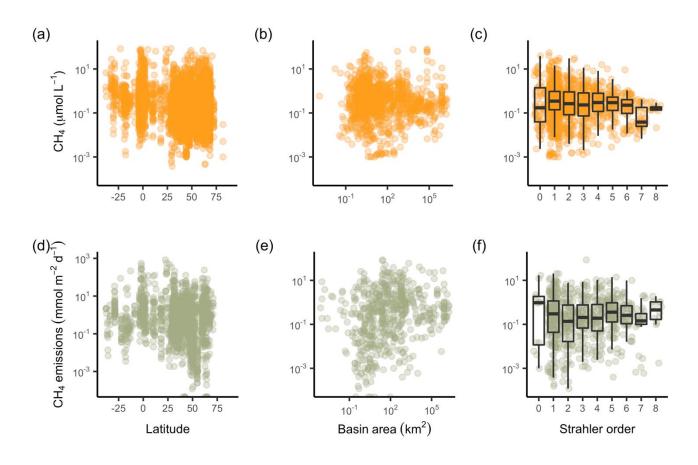


Figure 11: Site-average CH₄ concentrations (a-c) and flux (d-f) as function of latitude, basin area, and Strahler stream order. For boxplots, the upper and lower edges of each box are the 25th and 75th percentiles and whiskers are drawn up to 1.5 times the interquartile range.

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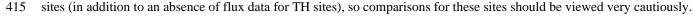
As with relationships between CH_4 physical site attributes, relationships between CH_4 concentration or flux and water chemistry parameters are also characterized by substantial variability. Representative examples indicate increasing, decreasing, and ambiguous relationships between CH_4 concentrations and fluxes and selected chemical constituents (Fig. 12). One source of variation shown in Fig. 12 can be attributed to differences among sites, as is illustrated for the case of CH_4 concentration

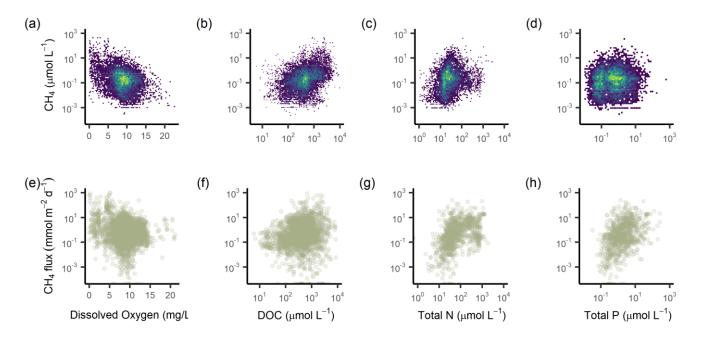
- 400 versus discharge (Fig. 13). The cluster of all points in this plot by itself does not suggest an obvious linear relationship between concentration and discharge; however, resolving the data to the site level for sites with multiple observations reveals several significant trends. Among 57 sites with >30 observations, 42 had significant relationships (P < 0.05) between concentration and discharge and 30 of these 42 trends were negative.
- 405 Median site concentrations and fluxes for most categories of targeted channels (Fig. 14) were significantly different than "normal" (NORM) sites (Kruskal-Wallis test $\chi^2 = 460.1$, df = 12, *P* < 0.0001). Pairwise Wilcoxon comparisons adjusted to





account for multiple comparisons (Benjamini and Hochberg, 1995) indicated that springs (SP) and delta channels (DC) did not differ from NORM sites (P > 0.4) and 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 CH₄ concentrations than NORM sites. Fluxes among channel type were also significantly different (Kruskal-Wallis test $\chi^2 = 143.8$, df = 12, P < 0.0001), and similar to concentration, fluxes in delta channels, permafrostinfluenced channels (PI), and springs were similar to NORM channels. Pairwise comparisons indicated that all other site types differed from NORM sites. Further, fluxes at floodplain sites were significantly higher (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.





420 Figure 12: CH₄ concentration (top row) and diffusive flux (bottom row) versus concurrent measures of dissolved oxygen (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 CH₄, 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).





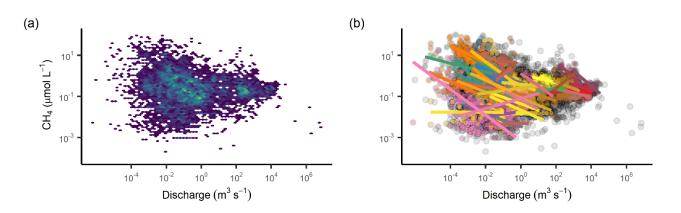
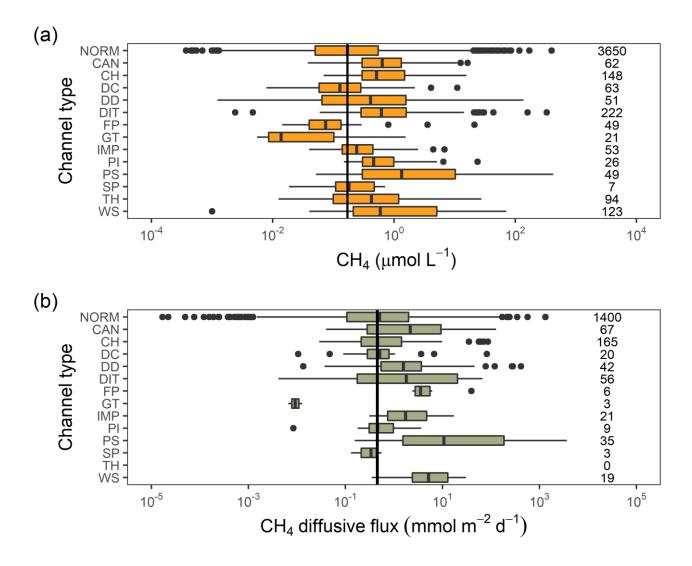


Figure 13: CH_4 concentration versus concurrent measures of discharge for (a) all sites and (b) sites with >30 observations (57 sites) with trend lines denoting within-site relationships between concentration and discharge. Each site is represented by a separate colour. Because of the log scale for CH_4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







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Fig. 14: Boxplots of site-averaged CH₄ 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 CH₄ 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.



445

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 across broad spatial scales in lotic ecosystems. Similarly, constructing GRiMeDB provided us with an unprecedented opportunity to identify tendencies in when, where, and

450 how CH₄ 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.

455 **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 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
- 465 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</p>
- 470 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
- 475 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, in press), and that the global north stores massive quantities of C in soils (Hugelius et al., 2014), more extensive coverage of CH₄ across Arctic drainage systems is warranted.





480 Although the spatial coverage of CH₄ 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) CH₄ 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 CH₄ 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

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 CO₂ 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 CH₄ 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

used as a basic sampling unit and results are presented as averages of replicates collected at different points within the study 505 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 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-

and during database construction. For example, geomorphologically distinct units (e.g., an individual riffle or pool) are often

510 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., 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).

515

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 though many studies of lotic CH₄ dynamics are framed in terms of understanding the contribution of these ecosystems to the

- 520 rapidly increasing atmospheric CH₄ pool, flux measurements lag far behind those of concentration, and the vast majority (ca. 85%) of flux data quantify only the diffusive pathway. 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 30-90% of total CH₄ emissions; Baulch et al., 2011; Crawford et al., 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
- ⁵²⁵ 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; Robison et al., 2021) requires good replication of bubble traps that need to be deployed over multiple days to generate reliable measurement. Given the logistic challenges and labourintensive 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
- 530 calculated from dissolved CH₄ and *k* (i.e., the 'chamber [concentration + *k*]' method in Fig. 9) 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. For example, this strategy is arguably inappropriate for situations in which the chamber gas content increases in a linear fashion, consistent with the occurrence of diffusive flux alone. 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
- 535 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

- 540 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 GRiMe 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
- 545 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 have led to differences in averages and measures of variability for CH₄ concentrations and fluxes compared our previous efforts. Median values for all three CH₄ 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 higher temporal resolution of 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 global-scale variability of fluvial concentrations and fluxes, and future database updates should be used to examine this relationship.
- 560 Despite the slight lowering of median values, supersaturated concentrations and positive fluxes are the norm for CH₄ as well as for CO₂ and N₂O. However, it is likely that CH₄ concentrations and fluxes below detection limits (BDLs) are under-reported, as is common with environmental data in general (Stow et al., 2018), so current averages may be slight overestimations of true population medians. Even given the modest number of zero or undetectable CH₄ 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 565 these observations are excluded, median CH₄ concentrations for all other observations increases from 1.49 to 1.51 μ mol 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 μmol L⁻¹. Although 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
- 570 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

575 variables that have been identified as potential predictors or drivers of CH₄ production, concentration, or flux (Figs. 11-13), and these plots suggest both the presence and absence of relationships. For example, increasing CH₄ 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 across the entirety of the GRiMeDB data. Similarly, increases in discharge have been linked to declines in gas concentration, likely due to source limitation (i.e., dilution) of terrestrial supply



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- (Aho et al., 2021; Gómez- Gómez-Gener et al., 2021a) and/or greater water turbulence, which increases gas exchange and in turn can reduce supersaturated CH₄ 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 CH₄ concentrations or used to extrapolate site-specific gas measurements to larger (even global) scales (e.g., Bastviken et al., 2011; Li et al., 2020),
 evidence for such relationships is 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 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
- 590 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, ditch and canal construction, oil and

- gas extraction, or passage through wetlands. Such signals persist at the global scale (Fig. 14), highlighting widespread human enhancement of CH₄ emissions from lotic ecosystems. Not all targeted sites are CH₄-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
 600 conditions, which allows higher rates of CH₄ oxidation in the water column. Indeed, oxidation has been shown to represent a
- significant CH₄ 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 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) has increased rapidly in the past decade. Recent upscaling efforts analyses (Rosentreter et al., 2021; Liu et al., 2022; Rocher-Ros et al., in review) have, for example, 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, we anticipate that their pairing with GRiMeDB data will result in significant improvements in the strength and certainty of continental and global-scale models explaining CH₄ distribution and quantifying fluvial emissions to the atmosphere.





5. Data and code availability

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GRiMeDB and its associated metadata are available from the Environmental Data Initiative (Stanley et al., 2022): 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.

620

6. Conclusion

The data gathered in GRiMeDB highlights many new opportunities, both through analysis of CH₄ and supporting data in the database, and by revealing gaps that currently exist across fluvial CH₄ studies. The most conspicuous data limitations include deficits in measurements of non-diffusive flux pathways in underrepresented arid, tropical, and arctic biomes. Challenges

- 625 associated with quantifying ebullition discussed above also emphasize the need for more intercomparisons among the various flux methods. Regardless of pathway, flux is a difficult process to quantify and can be highly sensitive to methods or gas exchange model choices and there are few methodological comparisons available to inform these decisions (Raymond et al., 2012; Lorke et al., 2015). 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
- 630 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). Unfortunately, GHG's, particularly CH₄ and N₂O, 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.

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

- 640 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 in estuaries where distinct processes such as tidal cycles and elevated sulphate 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
- 645 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.





650 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 CH4 data
Data_DOI_primary	DOI or hyperlink for CH4 data posted in a data repository
Data_DOI_supporting	DOI or hyperlink for separate datasets providing supporting data



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 <i>de novo</i> 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 A2. Column titles and content description for the GRiMeDB Sites Table.



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 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 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 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 if data are aggregated 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 for the sampling event
CH4_SD	Standard deviation of the mean CH ₄ concentration
CH4median	Median CH ₄ concentration
CO2min	Minimum measured CO_2 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
CO2max	Maximum measured CO_2 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
CO2mean	Mean or sole reported CO ₂ concentration for the sampling event
CO2_SD	Standard deviation of the mean concentration
CO2median	Median CO ₂ concentration
N2Omin	Minimum measured N ₂ O 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



N2Omax	Maximum 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
N2Omean	Mean or sole reported N_2O concentration for the concentration for the sampling event
N2O_SD	Standard deviation of the mean N2O concentration
N2Omedian	Median N ₂ O concentration
WaterTemp_degC	Water temperature in degrees C measured concurrently with CH4
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
рН	pH
DO_mgL	Dissolved oxygen in mg L ⁻¹
DO_percentsat	Percent saturation of dissolved oxygen
Q	Discharge measured at the time of sample collection
NO3	NO ₃ or NO ₂ +NO ₃ measured concurrently with CH ₄
NH4	NH ₄ measured concurrently with CH ₄
TN	TN or TDN measured concurrently with CH ₄
SRP	SRP or PO ₄ measured concurrently with CH ₄
TP	TP or TDP measured concurrently with CH ₄
DOC	DOC or TOC measured concurrently with CH ₄
Comments	Any additional relevant information regarding data
new_CH4_unit	Current common units for all CH ₄ concentrations
new_CO2_unit	Current common units for all CO ₂ concentrations
new_N2O_unit	Current common units for all N ₂ O 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

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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 ₂ O 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 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 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 CH4 data entered are averages from >1 date
Diffusive_CH4_Flux_Min	Minimum measured CH ₄ diffusive flux if data are aggregated or are from diel or data- dense spatial studies
Diffusive_CH4_Flux_Max	Maximum measured CH ₄ diffusive flux if data are aggregated or are from diel or data- dense spatial studies
Diffusive_CH4_Flux_Mean	Mean or sole reported CH4 diffusive flux for the sampling event
Diffusive_CH4_Flux_SD	Standard deviation of the mean CH4 diffusive flux
Diffusive_CH4_Flux_Median	Median CH ₄ diffusive flux
SampleCount_Diffusive	Number of samples or observations corresponding to the mean or median diffusive CH4 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

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 $k = gas \ exchange \ coefficient$ $C_w = CH_4 \ concentration \ measured \ in \ water$ $C_{eq} = CH_4 \ concentration \ in \ water \ in \ equilibrium \ with \ the \ atmosphere$
	other
	methods other than those described above
Eb_CH4_Flux_Min	Minimum measured CH_4 ebullitive flux if data are aggregated or are from diel or data- dense spatial studies
Eb_CH4_Flux_Max	Maximum measured CH ₄ ebullitive flux if data are aggregated or are from diel or data-dense spatial studies
Eb_CH4_Flux_Mean	Mean or sole reported CH4 ebullitive flux 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
SampleCount_Eb	Number of samples or observations corresponding to the mean or median ebullitive CH4 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 CH4 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



	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
Total_CH4_Flux_Max	Maximum measured total CH ₄ flux
Total_CH4_Flux_Mean	Mean or sole reported total CH4 flux for the sampling event
Total_CH4_Flux_SD	Standard deviation of the mean total CH4 flux
Total_CH4_Flux_Median	Median measured total CH ₄ flux
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 CH4 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)



Table A4. Continued	
	other methods other than those described above
CO2_Flux_Min	Minimum measured CO ₂ flux if data are aggregated or are from diel or data-dense spatial studies
CO2_Flux_Max	Maximum measured CO_2 flux if data are aggregated or are from diel or data-dense spatial studies
CO2_Flux_Mean	Mean or sole reported CO ₂ diffusive flux for the sampling event
CO2_Flux_SD	Standard deviation of the mean CO ₂ flux
CO2_Flux_Median	Median CO ₂ flux
N2O_Flux_Min	Minimum measured N_2O flux if data are aggregated or are from diel or data-dense spatial studies
N2O_Flux_Max	Maximum measured N ₂ O flux if data are aggregated or are from diel or data-dense spatial studies
N2O_Flux_Mean	Mean or sole reported N_2O diffusive flux for the sampling event
N2O_Flux_Stddev	Standard deviation of the mean N ₂ O flux
N2O_Flux_Median	Median N ₂ O flux
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 CH4 flux data
new_Eb_CH4_Flux_unit	Current common units for all ebullitive CH ₄ flux data
new_Total_Flux_unit	Current common units for all total CH4 flux data
new_CO2_Flux_unit	Current common units for all CO ₂ flux data
new_N2O_Flux_unit	Current common units for all N2O flux data
orig_Diffusive_Flux_unit	Original units for diffusive CH ₄ flux
orig_Eb_CH4_Flux_unit	Original units for ebullitive CH4 flux used
orig_Total_Flux_unit	Original units for total CH4 flux
orig_CO2_Flux_unit	Original units for CO ₂ flux
orig_N2O_Flux_unit	Original units for N ₂ O flux

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Appendix B. Citations for data sources in GRiMeDB. Citations are not provided for unpublished datasets.

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Supplement.

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1675 The supplement related to this article is available at

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 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.

1685 Competing Interests.

The authors declare that they have no conflict of interest.

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