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## **Inland water greenhouse gas budgets for RECCAP2: 1. State-of-the-art of global scale assessments**

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## **Keypoints**

We explore the state-of-the-art in inland water greenhouse gas emissions, discussing existing estimates and underlying methodologies

Development of models increasingly allows for assessment of spatial and temporal variability of emission fluxes

There is a persisting need for observations that capture hot-spots and hot-moments in emissions, including from small water bodies

## **Abstract**

Inland waters are important sources of the greenhouse gasses (GHGs) carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>) and nitrous oxide (N<sub>2</sub>O) to the atmosphere. In the framework of the 2<sup>nd</sup> phase of the REgional Carbon Cycle Assessment and Processes (RECCAP-2) initiative, we review the state of the art in estimating inland water GHG budgets at global scale, which has substantially advanced since the first phase of RECCAP nearly ten years ago. The development of increasingly sophisticated upscaling techniques, including statistical prediction and process based models, allows for spatially explicit estimates which are needed for regionalized assessments of continental GHG budgets such as those established for RECCAP. A few recent estimates also resolve the seasonal and/or interannual variability in inland water GHG emissions. Nonetheless, the global-scale assessment of inland water emissions remains challenging because of limited spatial and temporal coverage of observations and persisting uncertainties in the abundance and distribution of inland water surface areas. To decrease these uncertainties, more empirical work on the contributions of hot-spots and hot-moments to overall inland water GHG emissions is particularly needed.

## 1 Introduction

Inland waters (streams, rivers, lakes and reservoirs) are net-sources of greenhouse gasses (GHGs) to the atmosphere. They receive considerable amounts of reactive organic matter from terrestrial ecosystems, promoting the production of GHGs like carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>) and nitrous oxide (N<sub>2</sub>O). Inland waters are usually net-heterotrophic, meaning CO<sub>2</sub> production through respiration exceeds CO<sub>2</sub> consumption by aquatic production (Battin et al., in revision). An additional source of inland water GHG emission comes from terrestrial and wetland runoff and drainage which can be oversaturated in dissolved CO<sub>2</sub> produced by microbial and root respiration (Abril & Borges, 2019). Once this supersaturated aqueous solution enters surface waters, it can release gas to the atmosphere and contribute to inland water CO<sub>2</sub> emissions. Similarly, inland waters receive dissolved CH<sub>4</sub> and N<sub>2</sub>O from oversaturated soils and groundwater (Jurado et al., 2017; Rasilo et al., 2017). In addition, the sharp fronts between reducing and oxidizing conditions within the water column or at the interface between surface and subsurface environments (e.g. benthic and hyporheic zones) promotes the production and emissions of N<sub>2</sub>O (Marzadri et al., 2017, 2021). Moreover, autochthonous aquatic production may enhance nitrification in the water column through increased oxygen levels, while it may stimulate denitrification and methanogenesis in reducing, benthic sediments through delivery of labile organic matter. These processes play an important role in the N<sub>2</sub>O and CH<sub>4</sub> budgets of eutrophic lakes and reservoirs (DelSontro et al. 2018, Zhou et al., 2021).

While the processes driving GHG production have been known to limnologists for some time, large-scale quantification of inland water GHG emissions is still difficult and estimates are afflicted by large uncertainties. In their 5th Assessment Report (AR5, 2013), the IPCC acknowledged for the first time that inland waters are a significant contributor to the global GHG budget. At the same time, however, it was recognized that GHG fluxes from these ecosystems remain poorly constrained at the global scale. High uncertainties in flux estimates arise due to a poor spatial and temporal coverage of direct observations (Bastviken et al., 2011; Deemer et al., 2016; Regnier et al., 2013, 2022; Soued et al., 2016) and are reflected in the large range of estimated GHG fluxes reported in AR5: 0.8 - 1.2 Pg C yr<sup>-1</sup> for CO<sub>2</sub>, 8 - 73 Tg CH<sub>4</sub> yr<sup>-1</sup> for CH<sub>4</sub>, and 0.1 – 2.9 Tg N yr<sup>-1</sup> for N<sub>2</sub>O. The AR6 of the IPCC provides updated ranges for N<sub>2</sub>O (0.5 – 1.1 Tg N yr<sup>-1</sup>) and CH<sub>4</sub> (112 - 217 Tg CH<sub>4</sub> yr<sup>-1</sup>) emissions which are narrower, but still reflect significant uncertainties. This is especially true for inland water CH<sub>4</sub> emissions which remain proportionally the largest source of uncertainty in the global budget of this GHG (Canadell et al., 2021).

As part of the first phase of the REgional Carbon Cycle Assessment and Processes (RECCAP) initiative (RECCAP-1), Raymond et al. (2013) re-estimated global inland water CO<sub>2</sub> evasion suggesting that the total flux could be as high as 2.1 Pg C yr<sup>-1</sup>, which is about twice the estimates synthesized in AR5. This much higher estimate was due to a re-estimation of stream surface areas including small headwater streams which contribute disproportionately to the total water surface area and CO<sub>2</sub> emission, but which were neglected in earlier assessments that used datasets representing only larger global rivers (e.g. Cole et al., 2007). More importantly, Raymond et al. (2013) provided the first global maps of inland water CO<sub>2</sub> emissions, which allowed for the use of these estimates in regionalized, global C budgets (Bastos et al., 2020; Ciais et al., 2021; Zscheischler et al., 2017).

Since RECCAP1, a growing number of global estimates of inland water GHG emissions have been published, not only for CO<sub>2</sub> emissions (e.g. Holgerson & Raymond, 2016; Horgby et al., 2019; Lauerwald et al., 2015; Liu et al., 2022), but also for CH<sub>4</sub> (e.g. Holgerson & Raymond, 2016; Rosentreter et al., 2021; Stanley et al., 2016) and N<sub>2</sub>O (e.g. Hu et al., 2016; Lauerwald et al., 2019; Maavara et al., 2019; Marzadri et al., 2021; Soued et al., 2016; Yao et al., 2020), or for all three GHGs combined (e.g. Deemer et al., 2016; DelSontro et al., 2018). While the limited availability and quality (e.g. length and frequency of time-series), and uneven global coverage of observed emission rates (see e.g. Deemer et al. 2016) still represent a large source of uncertainty, the amount and quality of empirical data has steadily increased over the past decade. In addition, global emission estimates profited from the appearance of new, improved data sets of inland water surface areas (Allen & Pavelsky, 2018; Lehner et al., 2011; Messenger et al., 2016; Verpoorter et al., 2014). Finally, global scale estimation of inland water GHG budgets have been improved through novel upscaling techniques based on statistical (e.g. Lauerwald et al. 2015, DelSontro et al. 2018) and process based models of varying complexity (e.g. Maavara et al. 2019, Yao et al. 2020).

In the framework of the second phase of RECCAP (RECCAP-2), we present a review of existing global estimates of inland water GHG emissions. We start with a general overview of methods to achieve global scale estimates, starting from methods to measure flux rates in the field, followed by methods used to upscale flux rates to the global scale and which comprise a large range of approaches including simple upscaling based on average observed flux rates, statistical prediction and the use of process based models (section 2). Then, in three subsections respectively dedicated to estimates of emissions of CO<sub>2</sub> (section 3), CH<sub>4</sub> (section 4) and N<sub>2</sub>O (section 5), we discuss the state of the art for each of these GHGs in more detail, review all existing global estimates, and explore differences between flux assessments and their underlying methods. In addition, we highlight for each GHG persisting shortcomings and challenges for future research. The companion paper in the same issue (Lauerwald et al., submitted) then builds on the present review to derive a regionalized assessment for the 10 regions used in the RECCAP-2 project. In this companion paper, each previously published global estimate reported here was rescaled using the same new global assessment of inland water surface area, allowing for better consistency and homogeneity across all previously published values

## **2 Overview of upscaling strategies and surface area estimates used in global studies of inland water GHG emissions**

This subsection gives a brief overview of different methods that are used to obtain global scale estimates of inland water GHG emissions. These methods are here classified into three main approaches, namely direct upscaling based on observations (2.1), statistical upscaling based on functional relationships between emissions and environmental drivers (2.2), and process-based models (2.3). We also briefly review here progress in the global scale assessment of inland water surface areas (2.4), which is of vital importance for global upscaling of inland water GHG emissions.

### **2.1 Upscaling based on observations**

Large-scale estimates of inland water GHG emission fluxes  $F_{GHG}$  are usually calculated as the product of an average flux rate  $f_{GHG}$ , which can be expressed in units of mass per area and time, as derived from a set of field observations, and an estimate of the inland water surface area  $A_{IW}$  for which this flux rate is assumed to be representative (eq. 1).

$$F_{GHG} = f_{GHG} * A_{IW} \quad (\text{eq. 1})$$

Many estimates have applied this simple upscaling technique directly at the global scale using an average  $f_{GHG}$  multiplied by the total  $A_{IW}$  of one specific type of inland waters. For instance, Deemer et al. (2016) calculated the average rates of GHG emissions from reservoirs, using observations from empirical studies around the world, and multiplied those average rates by the estimated total area of reservoirs after Lehner et al. (2011). Others have first broken down the total of inland waters of one type into different subgroups, e.g. based on size of water body or stream order (Holgerson & Raymond, 2016; Humborg et al., 2010), geographic region (e.g. Aufdenkampe et al., 2011; Bastviken et al., 2011; Soued et al., 2016; Johnson et al., 2021) or both (Raymond et al. 2013, Rosentreter et al. 2021). An area-integrated flux from each subgroup was then calculated following the same eq. 1, before summing those up to a global flux.

Methods and challenges to obtain estimates of  $A_{IW}$  are presented in detail in section 2.4. In what follows, we will first focus on uncertainties associated with measuring and calculating  $f_{GHG}$ . Flux rates can either be obtained from GHG emission rates directly measured in the field (section 2.1.1), or from measurements or calculation of GHG concentration gradients and concomitant measurements or models of gas transfer velocities (section 2.1.2). Note that this study does not aim to provide a detailed review of field methods. These aspects are thus only briefly discussed, with a focus on methodological uncertainties.

### 2.1.1 Directly observed flux rates

A common method to measure aquatic GHG emission rates is the use of floating chambers, which resemble inverted plastic buckets put onto the water surface. The emission rates are then calculated based on the accumulation rate of GHGs within the floating chamber headspace. This method detects the emission rate from the small surface area of the floating chamber across the larger area over which the chamber may be moving during the deployment. Chambers may drift a few meters if tethered or over longer distances if drifting freely during the deployment. Such a well-defined footprint is advantageous for studies of local flux regulation and for distinguishing variability in space versus time. Concurrently, the small size of the footprint leads to potentially high uncertainties in the extrapolation of flux chamber measurements to large areas, without numerous representative measurements. Eddy covariance towers, though less common and only applicable in standing water bodies of a certain size have the advantage of generating net fluxes (i.e. emission or uptake) from a larger surface area (depending on height, surface roughness and wind speeds, eddy covariance towers can have a footprint of up to 3-km radius (Chu et al., 2021)), thus delivering a more representative emission rate (Podgrajsek et al., 2014). In contrast to the floating chamber method, the eddy covariance technique also allows for continuous measurements which provide better temporal resolution in emission rates. However, the flux footprint is constantly moving with wind speed and direction, making variability in time and space

challenging to distinguish. Fluxes cannot be measured at all when there is no wind (e.g. typical during night time) and complications associated with rainfall and lateral advective gas flux make accurate flux measurements challenging. Eddy covariance also relies on the performance of advanced equipment and a high level of operator expertise for adequate data filtering and QA/QC. Above all, limited eddy covariance measurements mean that global upscaling based only on this method is not yet possible, and inherent limitations make eddy covariance suboptimal for key inland water emission measurements such as fluxes from streams and along lake shores.

While the majority of CO<sub>2</sub> and N<sub>2</sub>O emissions occur through diffusive flux across the air-water interface, a significant but variable fraction of aquatic CH<sub>4</sub> flux occurs as bubbles (i.e. ebullition). Ebullition occurs when CH<sub>4</sub> produced in aquatic sediments forms gas bubbles that at a certain size, due to buoyancy, evade the sediment layer and ascend through the water column. Existing emission estimates from floating chambers sometimes intentionally exclude ebullition. Other floating chamber methods include both diffusive and ebullitive emissions. Also eddy covariance towers measure the sum of both emission pathways. There are nonetheless various methods to directly quantify ebullition. However, these methods detect bubbles rather than CH<sub>4</sub> and need supplementary measurements of CH<sub>4</sub> concentration within the bubble air, usually from manually taken samples, to allow flux estimation. This point is critical as CH<sub>4</sub> concentration in bubbles can vary widely, from less than 1% to > 80% (Boereboom et al. 2012). The most common methods for directly quantifying ebullition rates is the bubble trap, an inverted funnel that collects ascending bubbles and is sometimes connected to a hydrostatic pressure sensor (Varadharajan et al., 2010) or specialized bubble size sensors (Delwiche & Hemond, 2017) to measure the timing and size distribution of ascending bubbles. Ebullition measurements based on point measurements in space and time are currently very labor intensive given the high spatiotemporal variability of ebullition fluxes (Linkhorst et al., 2020). Echosounders (Ostrovsky et al., 2008), robotic boats connected to optical methane detectors (Grinham et al., 2011) and under-ice surveys (Wik et al., 2011) have also been used to quantify ebullition rates. In addition, radar remote sensing approaches are currently being developed that could integrate over space and time for more representative measurements (Engram et al., 2020).

### 2.1.2 Estimating diffusive fluxes based on concentration gradients

The methods for directly measuring emission rates can easily be applied in deeper, slower-moving waters (floating chambers and funnel traps) or in larger water bodies (eddy covariance). However, these methods are often not feasible for smaller streams. Instead, emission flux rates can be calculated from a gradient in concentrations of a specific GHG ( $\Delta C_{GHG}$ ) in the water close to the surface and in the overlying atmosphere and a gas exchange velocity  $k_{GHG}$  (eq. 2). Note that this method only allows estimation of diffusive emissions, and is not applicable for ebullition. The gradient  $\Delta C_{GHG}$  can be calculated based on direct field measurements using headspace equilibration methods (e.g. Müller et al., 2015), or using measured headspace partial pressures and solubility constants that depend on salinity and water temperature (Weiss, 1970).

$$f_{GHG} = \Delta C_{GHG} * k_{GHG} \quad (\text{eq. 2})$$

The headspace equilibration method consists of equilibrating a known volume of sampled water and a known volume of air, with a known initial partial pressure of the GHG to be analyzed. After full equilibration, a sample of the headspace is analyzed by e.g. gas chromatography, optical gas analyzers, or other gas analysis methods, to measure the corresponding GHG partial pressure from which the  $C_{GHG}$  in the sampled surface water can be calculated. This concentration is compared with the theoretical concentration in equilibrium with the background air partial pressure of the GHG in focus to yield  $\Delta C_{GHG}$ . In the case of  $CO_2$ , concentrations can also be calculated from observations of alkalinity and pH based on chemical equilibria and the assumption that non-carbonate contributions to alkalinity are negligible, which can be questioned in some common aquatic systems [Abril et al 2015] (see section 3.2 for more discussion).

Gas exchange velocity can be assessed through direct tracer studies in which a specific tracer gas is released into the stream, and its loss is measured over a defined length. As this method is too cumbersome and costly to be applied everywhere, empirical equations have been established that relate  $k_{GHG}$  to the rate of energy dissipation at the water-air interface. Energy dissipation causes the turbulent mixing of the upper water column and thus determines the depth of the water column which interacts with the atmosphere through the process of diffusion. For streams and rivers, this energy dissipation rate can be estimated from stream flow velocity and stream channel geometry, in particular the slope of the stream channel (Natchimuthu et al., 2017; O'Connor & Dobbins, 1958; Raymond et al., 2012). More recent work has however noted a breakpoint in the energy dissipation rate at which air entrainment and bubble formation cause  $k_{GHG}$  to increase more rapidly with energy dissipation (Ulseth et al., 2019). This suggests that assuming only diffusive water-air gas exchange, as it is assumed in most studies of inland water  $CO_2$  emissions, may lead to underestimated gas transfer velocities in systems with very high hydrological energy. For lakes and reservoirs, empirical equations relate  $k_{GHG}$  to wind speed (Cole & Caraco, 1998), lake surface area (Read et al., 2012), or both (Vachon & Prairie, 2013), as the degree to which wind shear vs convective mixing dominate gas transfer dynamics generally changes as a function of waterbody size. More sophisticated modeling of  $k_{GHG}$  from lake hydrodynamics considering multiple turbulence-generating processes have also been developed (e.g. MacIntyre et al., 2021). It has been suggested that models of  $k_{GHG}$  should be locally validated whenever possible (e.g. Schilder et al. 2013).

## 2.2 Upscaling based on statistical prediction

A variety of statistical methods have been used to upscale flux measurements/estimates to the global scale. These methods can be categorized into two groups of statistical upscaling approaches: 1) methods that predict emission rates directly, and 2) methods that first predict  $A_{IW}$ ,  $\Delta C_{GHG}$  and  $k_{GHG}$  separately, and combine them using eqs. 1 and 2 to estimate the emission flux  $F_{GHG,IW}$ .

A simple example for the first group of methods is the use of emission factors (EFs), which has been applied to estimate  $N_2O$  emissions from river networks (Beaulieu et al., 2011; Kroeze et al., 2010). Averaged EFs, typically defined as the ratio of  $N_2O$  emissions to riverine N loads, were derived from a number of field studies. These EFs were then multiplied by global, spatially explicit estimates of river N loads (e.g. Mayorga et al., 2010) to estimate global riverine  $N_2O$  emissions at the same spatial resolution as the riverine N loads. This method assumes that riverine  $N_2O$  emissions simply scale linearly to riverine



N loads, which is problematic from a reaction kinetics point of view, as discussed in Maavara et al. (2019). As an alternative empirical approach, Hu et al. (2016) used findings from field studies to fit equations predicting riverine N<sub>2</sub>O emissions as a nonlinear function of dissolved inorganic N yield and catchment area, thus overcoming some of the limitations of the EF approach.

Another prominent example for the first group of methods is the study by DelSontro et al. (2018), predicting global lake CH<sub>4</sub> emissions empirically. DelSontro et al. (2018) fitted multilinear regressions equations to a database of literature studies of 166 water bodies quantifying lake CH<sub>4</sub> emissions, that predict the total (diffusive + ebullitive), annual emission flux from lake size and lake productivity (defined as chlorophyll or phosphorus concentration). The fitted regression equations were then applied to different global datasets/estimates of lake surface area and an assumed statistical distribution of lake productivity across global lakes to estimate the global-scale CH<sub>4</sub> emissions from these water bodies.

Examples for the second group of methods are the studies by Raymond et al. (2013), Lauerwald et al. (2015), and Horgby et al. (2019) that estimated CO<sub>2</sub> emissions from rivers at the global scale or for specific ecoregions (Horgby et al. 2019 focused on alpine streams). These studies all used global datasets including digital elevation models and their derivatives (stream network and channel slope) and gridded estimates of average annual river flow to estimate stream surface area and  $k_{GHG}$  spatially explicitly. While Raymond et al. (2013) combined their estimates of  $A_{IW}$  and  $k_{GHG}$  with regionalized averages of calculated  $\Delta C_{CO_2}$ , Lauerwald et al. (2015) and Horgby et al. (2019) further used multiple linear regression models to estimate riverine  $\Delta C_{CO_2}$  from different spatial drivers (like terrestrial Net Primary Productivity - NPP, climate, terrain steepness in Lauerwald et al. 2015, or elevation, soil carbon stocks, and discharge in Horgby et al. 2019). Note that combining independent estimates of  $k_{GHG}$  and  $\Delta C_{GHG}$  introduces an additional source of uncertainty, as  $\Delta C_{GHG}$  is in turn controlled by  $k_{GHG}$  and its balance with CO<sub>2</sub> resupply rates to the surface water, which is for instance evidenced by low  $\Delta C_{GHG}$  in turbulent, high alpine streams (Horgby et al. 2019).

### **2.3 Process-based models**

Process-based models of varying degrees of complexity have recently been used to assess inland water GHG emission at the global scale (Maavara et al., 2019; Marzadri et al., 2021; Yao et al., 2020). Ideally, such models represent carbon and nutrient transport and transformation processes that drive production, cycling and emission of GHGs in a water body or along a cascade of water bodies (like a sequence of stream reaches or a cascade of reservoirs along a river network). This representation requires boundary condition data at the global scale and in sufficient quality and quantity. This data requirement is a major limitation for the applicability of process-based models for inland water GHG emissions at the global scale.

A promising strategy to overcome that limitation is the explicit representation of inland waters and associated biogeochemical processes in land surface models (LSMs) that simulate the terrestrial cycling of energy, water, C, nutrients, and GHGs. Using LSMs, the biogeochemical and transport processes that drive the GHG dynamics can be simulated simultaneously for terrestrial and freshwater ecosystems, reducing the need for complex boundary conditions at the land-inland water interface. Developments in

that direction have been achieved for the LSMs DLEM (Tian et al., 2015a,b; Yao et al., 2020) and ORCHIDEE (Lauerwald et al., 2017, 2020). At global scale, LSM simulations including inland water N<sub>2</sub>O and CO<sub>2</sub> emissions have yet only been achieved with DLEM (Yao et al. 2020, Tian et al. 2015b).

When using LSMs, the simulated water fluxes and associated terrestrial C and nutrient inputs to inland waters are already afflicted by considerable uncertainties, including those arising from the overparameterization of these extremely complex models. Thus, an alternative is to use process-based models of only inland waters forced by data driven information. The global river network N<sub>2</sub>O modeling studies by Maavara et al. (2019) and Marzadri et al. (2021) follow two different strategies to overcome data limitations to constrain the models. Maavara et al. (2019) followed a metamodeling strategy, for which a box model representing all major processes of N and N<sub>2</sub>O cycling in a water body was first set-up. While this process-based model could not be applied at global scale due to data limitations to constrain each biogeochemical process, Maavara and colleagues ran the model across a realistic range of model input parameters using a Monte Carlo approach to derive simple response functions. The resulting response functions relating N<sub>2</sub>O emissions to nutrient loads and water residence times were then applied at global scale using loads and residence times derived from available global datasets. Marzadri et al. (2021) applied their process-based model of river N<sub>2</sub>O emissions directly at global scale, which required spatially resolved model inputs comprising a detailed set of parameters describing stream hydro-morphology and water quality, which in that form did not yet exist at global scale. To overcome that limitation, machine learning techniques were applied to derive these input datasets from other, available geodata. These input data were then used to feed a process-based model that parametrizes N<sub>2</sub>O emissions as a function of river size by means of two Damköhler numbers representing the ratio between a characteristic time of transport and a characteristic time of reaction. The proposed hybrid model (machine learning + process based) allows consideration of the contribution of surface (e.g. water column) and subsurface (e.g. benthic and hyporheic zones) processes to N<sub>2</sub>O emissions (Marzadri et al., 2021).

For aquatic CH<sub>4</sub> emissions, process-based modeling efforts have been mostly dedicated to lake and reservoir systems. For example, an online, open-source predictive model framework “G-res” has recently been developed to provide global, spatially explicit estimates of the form and magnitude of reservoir CH<sub>4</sub> and CO<sub>2</sub> emissions (Harrison et al., 2021; Prairie et al., 2021). G-res uses a series of calibrated empirical models that integrate local (reservoir-specific) and regional (watershed attributes) information to predict GHG emissions (Prairie et al. 2021). The model has been applied to 4,727 reservoirs to estimate global emissions (Harrison et al. 2021). Tan and Zhuang (2015a, 2015b) have developed and applied a process based model to estimate CH<sub>4</sub> emissions from lakes at pan-arctic scale. That model produces gridded output, resolves seasonal and interannual variability and permits for projections of long-term trends following global change scenarios.

## **2.4 Available data and previous estimates of global inland water surface area**

The first digital global map of inland water surface areas that was used for inland water GHG emission estimates was the Global Lake and Wetland Database (GLWD) by Lehner and Döll (2004). GLWD was derived from a compilation of different global and regional inventories. While GLWD is not globally

consistent with regard to detail and reliability of the data sources, it represented the best available dataset for more than a decade and was used in numerous studies of inland water GHG emissions (e.g., by Raymond et al. 2013 for lakes and reservoirs, by Aufdenkampe et al. 2011 for all water bodies). Since then, our ability to estimate the global surface area of rivers, lakes, and reservoirs has progressed significantly. This progress has been driven by advances in satellite remote sensing, image processing methods, and geospatial analysis techniques. Several freely available global hydrography datasets have recently become available that can be used to estimate surface area and distribution of inland water bodies. Here we discuss a selection of high-resolution, freely available datasets that can be useful for global-scale evaluations of greenhouse gas emissions from inland water bodies.

A few global inland water body datasets have been developed using optical remote sensing data. The JRC GSW datasets from Pekel et al. (2016) and the GSWD from Pickens et al. (2020) are two global 30-m-resolution datasets of open surface water extent, created from the Landsat archive. These datasets are multitemporal and highly consistent but they do not distinguish between different water body types (e.g. rivers, lakes, etc.). Classifying water body type is necessary in evaluations of GHG exchange because of differing exchange rates and processes occurring in different aquatic environments. The Global River Widths from Landsat (GRWL) database (Allen & Pavelsky, 2018) contains exclusively river surface areas derived from Landsat imagery.

In addition to these image-based datasets, global topography-based datasets derived from digital elevation models (DEMs) have been used for representing the global extent and distribution of streams and rivers. These include hydrologically conditioned gridded raster datasets like HydroSHEDS (Lehner et al., 2008) and MERIT Hydro (Yamazaki et al., 2019) or vectorized flowline datasets derived from these gridded datasets including HydroRIVERS (Lehner & Grill, 2013) or MERIT Hydro–Vector (Lin et al., 2021). These DEM-based datasets can be used to infer the location and size of narrow rivers and streams too small to be visible from freely available satellite datasets. These datasets can also be used to infer other characteristics of river networks including stream order, slope, upstream area, and topology, which are of potential value for estimating amount and turbulence of riverflow, which in turn are important drivers of GHG emissions. Other hydrography datasets innovatively combine DEM-based datasets with other sources of data to produce novel information including machine-learning based estimates of river surface area (Lin et al., 2020) and the extent of non-perennial rivers (Lin et al., 2021; Messenger et al., 2021).

For standing open water bodies like lakes and reservoirs, attempts have also been made to identify water bodies from satellite imagery using automated algorithms. A prominent example is the Global Water Body (GLOWABO) dataset (Verpoorter et al., 2014). Due to the unsupervised classification method and missing evaluation of ground truth, this dataset is however highly uncertified and likely contaminated with wrongly assigned riverine, coastal or temporal water bodies. In addition, inventory based datasets have further been developed, including the Global Reservoir and Dam database (GRanD) (Lehner et al., 2011) and the HydroLAKES database (Messenger et al., 2016) which gives water surface areas of standing waters distinguishing lakes from reservoirs. Note that HydroLAKES also includes the information from GRanD and GLWD, which makes these products partly redundant. The advantage of

inventory based datasets as GRanD and HydroLAKES is the avoidance of contamination with other water bodies and additional attributes such as names, estimates of water volume and residence time, height and purpose of dam for reservoirs, etc. In particular the distinction between lakes and reservoirs is of major importance for the assessment of inland water GHG emissions. Reservoirs as artificial water bodies deserve special attention, as they represent an anthropogenic source of GHGs and a potential lever for controlling future emissions. However, we have to expect an under-classification of reservoirs in inventory datasets such as HydroLAKES, as water bodies for which this information was not available have been categorized as natural lakes by default (Messenger et al. 2016). Smaller hydropower projects which outnumber large hydropower projects by approximately 11:1 (Couto & Olden, 2018) may not always be inventoried and accounted for in regional and global datasets. Recently, new datasets of dams and reservoirs have been created combining remote sensing-based datasets with other sources of information, e.g., GOODD (Mulligan et al., 2020) and GeoDAR (Wang et al., 2021), continuously increasing the numbers of reservoirs that are taken up into inventories.

Although considerable progress has been made recently in developing global hydrography datasets, much less work has been done to apply these datasets to estimate global surface area of inland water bodies. For the surface area of rivers, three notable global estimates have been produced by Downing et al. (2012) of 485,000 and 682,000 km<sup>2</sup>, Raymond et al. (2013) of 487,000 and 761,000 km<sup>2</sup>, and Allen & Pavelsky (2018) of 773,000±79,000 km<sup>2</sup>. Downing et al. (2012) based their estimate on >400 observations of stream width, data on number and length of streams from HydroSHEDS dataset, and statistical scaling relating stream number, width and length to stream order. Raymond et al. (2013) combined the stream network of HydroSHEDS with gridded runoff data to obtain a distribution of stream lengths and discharge per stream order of medium to large rivers, to which they then applied empirical, hydraulic equations predicting stream width from discharge. Finally, they used stream-order based scaling laws to estimate stream surface areas for smaller streams. Allen and Pavelsky (2018) used their remote-sensing based GRWL database for surface areas of medium to large rivers, which they complemented with topography- and statistical-based estimates for streams narrower than 90m to headwater streams as defined by Allen et al. (2018). The GRWL dataset is to date the most complete and reliable dataset of its kind.

For the surface area of lakes and reservoirs, three notable global estimates have been made by Downing et al. (2006), Verpoorter et al. (2014), and Messenger et al. (2016). Downing et al. (2006) used surface areas from standing water bodies > 10 km<sup>2</sup> from GLWD (Lehner & Döll, 2004) and extrapolated the surface area to smaller water bodies down to 0.001 km<sup>2</sup> assuming power-law relationships (Pareto-law distributions) between water body size and frequency. Verpoorter et al. (2014) used their remote-sensing derived GloWaBo database which includes lakes as small as 0.002 km<sup>2</sup>. Messenger et al. 2016 derived their estimate from their inventory based HydroLAKES database, which contains water bodies >0.1 km<sup>2</sup>. Due to this restriction with regard to minimum lake size, Messenger et al. (2016) obtained the lowest of the three global surface area estimates for standing waters with 2.7 x 10<sup>6</sup> km<sup>2</sup>. The estimate of Downing et al. (2006) is substantially higher with 4.2 x 10<sup>6</sup> km<sup>2</sup>, while for water bodies larger than 0.1 km<sup>2</sup>, their estimate of 2.9 x 10<sup>6</sup> km<sup>2</sup> is quite comparable to HydroLAKES. The estimate by Verpoorter et al. (2014) is even higher with 5 x 10<sup>6</sup> km<sup>2</sup>, likely due in part to overestimation of lake areas through

contamination with other water bodies. A reliable map of smaller bodies of standing water, such as ponds, which are thought to contribute substantially to the total water surface area and disproportionately to GHG emissions (Holgerson and Raymond 2016; Rosentreter et al. 2021), is still not achievable.

### 3 Inland water CO<sub>2</sub> budget

#### 3.1 Overview of existing estimates

**Table 1.** Global estimates of inland water CO<sub>2</sub> emissions. For each estimate, the total water surface area ( $\Sigma A_{water}$ ), the total CO<sub>2</sub> emission flux ( $\Sigma CO_{2\ em}$ ) and the area weighted average emission rate ( $\Sigma CO_{2\ em}/\Sigma A_{water}$ ) are reported.

Reference	$\Sigma CO_{2\ em}/\Sigma A_{water}$ [g CO <sub>2</sub> m <sup>-2</sup> yr <sup>-1</sup> ]	$\Sigma A_{water}$ [10 <sup>6</sup> km <sup>2</sup> ]	$\Sigma CO_{2\ em}$ [Pg CO <sub>2</sub> yr <sup>-1</sup> ]	Method
<i>Rivers</i>				
DLEM (Tian et al. 2015b)	3531	0.64	2.24	Model
Liu et al. 2022 <sup>&amp;</sup>	9900	0.672	7.33±0.73 <sup>b</sup>	Machine learning
Lauerwald et al. 2015 <sup>&amp;</sup>	5771	0.55- 0.67	2.38 (1.77- 3.10) <sup>a</sup>	Statistical prediction
Raymond et al. 2013 <sup>&amp;</sup>	10644	0.62	6.6 (5.7-7.5) <sup>a</sup>	Upscaling from observations + Statistical prediction
Aufdenkampe et al. 2011	5009	0.31- 0.51	2.05	Lumped estimate
Tranvik et al. 2009			2.02	Literature review
Cole et al. 2007	1492	0.74	0.84	Literature review
<i>Streams and small rivers</i>				
Liu et al. 2022 <sup>&amp;</sup>	23962	0.202	4.84	Machine learning
Marx et al. 2017			3.41	Literature review
Lauerwald et al. 2015 <sup>&amp;</sup>	7348	0.14- 0.26	1.16 (0.78- 1.61) <sup>a</sup>	Statistical prediction
<i>Mountain streams</i>				
Horgby et al. 2019	17490	0.035	0.61	Statistical prediction
<i>Large rivers</i>				
Liu et al. 2022 <sup>&amp;</sup>	4946	0.47	2.31	Machine learning
Lauerwald et al. 2015 <sup>&amp;</sup>	3942	0.41	1.22 (0.96- 1.54) <sup>a</sup>	Statistical prediction
<i>Lakes and reservoirs</i>				
DelSontro et al. 2018	414	3.23-	1.99-3.30	Upscaling from

		5.36		observations
-"	416	4.42	1.84 (1.72-1.98) <sup>a</sup>	Statistical prediction
-"	360	5.36	1.93 (1.80-2.06) <sup>a</sup>	Statistical prediction
-"	276	3.23	0.89 (0.83-0.96) <sup>a</sup>	Statistical prediction
Raymond et al. 2013 <sup>&amp;</sup>	392	3	1.17 (0.22-3.08) <sup>a</sup>	Upscaling from observations + Statistical prediction
Aufdenkampe et al. 2011	638	2.80-4.54	2.35	Upscaling from observations
<i>Lakes (including lakes with dams)</i>				
DLEM (Tian et al. 2015b)	312	2.4	0.77	Model
Holgerson and Raymond 2016	348	5.98	2.14	Upscaling from observations
Tranvik et al. 2009			1.94	Literature review
Cole et al. 2007	257	2	0.4	Literature review
<i>Reservoirs</i>				
DLEM (Tian et al. 2015b)	312	0.27	0.08	Model
Deemer et al. 2016	451	0.3	0.14 (0.12-0.16) <sup>a</sup>	Upscaling from observations
Cole et al. 2007	686	1.5	1.03	Literature review
<sup>a</sup> lower and upper 90% (Raymond et al. 2013; Lauerwald et al. 2015; Deemer et al. 2016) or 95% (DeSontro et al. 2018) CI <sup>b</sup> standard error <sup>c</sup> min and max estimate <sup>&amp;</sup> estimate accounts for effects of seasonal ice cover				

Global estimates for the aquatic CO<sub>2</sub> emission range from 0.84 to 7.33 Pg CO<sub>2</sub> yr<sup>-1</sup> for streams and rivers, from 0.40 to 2.09 Pg CO<sub>2</sub> yr<sup>-1</sup> for lakes, from 0.08 to 0.14 Pg CO<sub>2</sub> yr<sup>-1</sup> for reservoirs (excluding the estimate by Cole et al. (2007), which is discussed at the end of this section), and from 1.27 to 2.35 Pg CO<sub>2</sub> yr<sup>-1</sup> for estimates that lumped lakes and reservoirs together (Table 1). In general, considerable discrepancies exist in particular between early estimates that relied mostly on lumped estimates of average CO<sub>2</sub> concentrations,  $k_{GHG}$  and water surface area, and more recent estimates relying on more complete concentration datasets, more sophisticated upscaling approaches and spatially resolved water surface area estimates. For streams and rivers, the earliest estimates (Cole et al., 2007, p. 207; Cole & Caraco, 2001) were crude and most likely underestimate riverine CO<sub>2</sub> emissions because of their reliance on data from large rivers, which tend to show lower areal CO<sub>2</sub> emission rates than smaller and more upstream systems, as large rivers tend to be less heterotrophic, receive less important inputs of CO<sub>2</sub> enriched groundwater, and show less turbulent stream flow which leads to lower gas exchange velocities (Raymond et al. 2013). Relying on an extensive database for  $pCO_2$ , new scaling laws for  $k_{GHG}$  and stream hydraulic geometry that allowed for spatially resolved estimates for stream surface areas at the global

scale, Raymond et al. (2013) presented the first spatially explicit estimate for the aquatic CO<sub>2</sub> flux and reports a river CO<sub>2</sub> evasion rate that is 3–8 times higher than the earlier lumped estimates (Aufdenkampe et al., 2011; Cole et al., 2007; Tranvik et al., 2009). Moreover, they demonstrated the importance of small headwaters which contribute disproportionately to the total emission flux. More recent advancements in stream and river CO<sub>2</sub> evasion estimates involve development of data-driven statistical models to resolve temporal and finer spatial scale variations of the riverine CO<sub>2</sub> flux (Lauerwald et al., 2015; Liu et al., 2022). For instance, relying on direct C<sub>CO2</sub> measurements and seasonally varying estimates for *k* and river surface area, Liu et al. (2022) demonstrated CO<sub>2</sub> emission from global streams and rivers varied between 411 to 766 Tg CO<sub>2</sub> yr<sup>-1</sup> per month, i.e. by a factor of ~ 2, with the highest global emissions during northern summer in July.

A process-based model has also been developed (DLEM, Tian et al. 2015b), which predicts a much lower emission rate than recent data-driven approaches (2.24 versus 6.60-7.33 Pg CO<sub>2</sub> yr<sup>-1</sup>) (Table 1). The DLEM estimate is however close to the estimate by Lauerwald et al. (2015) (2.38 Pg CO<sub>2</sub> yr<sup>-1</sup>) that only accounted for emissions from medium-sized to large rivers (i.e., 3rd order and above). The large discrepancy (i.e., 2.24 versus 6.60-7.33 Pg CO<sub>2</sub> yr<sup>-1</sup>) however argues for the importance of the smallest streams in global CO<sub>2</sub> emission from fluvial networks (Marx et al., 2017). In line with this, Liu et al. (2022) estimated emission from the medium-to-large rivers (corresponding roughly to stream order 3 and above as in Lauerwald et al. 2015) of ~ 2.31 Pg CO<sub>2</sub> yr<sup>-1</sup>, while roughly two thirds of the total riverine emissions (~ 5 Pg CO<sub>2</sub> yr<sup>-1</sup>) are predicted to be emitted by smaller streams (extrapolated to a minimum stream width of 0.3 meters).

For lakes, there is a much larger variation in estimates of water surface area than in average emission rates between different studies (Table 1). In particular, estimates that relied on earlier global lake inventories (Raymond et al., 2013) report lower surface area and total emissions than more recent estimates based on newer lake inventories and extrapolated surface area to account for the smallest water bodies (DelSontro et al., 2018; Hastie et al., 2018; Holgerson & Raymond, 2016). Despite employment of scaling laws (e.g., with lake size and nutrient status) that account for spatial variability due to system size and autotrophic productivity in more recent estimates (DelSontro et al., 2018; Holgerson & Raymond, 2016; Raymond et al., 2013), there seems to be only small difference with regard to global average lake CO<sub>2</sub> emission rates per water surface area between those newer estimates (348–414 g CO<sub>2</sub> m<sup>-2</sup>yr<sup>-1</sup>) and those of the early crude estimates (257 g CO<sub>2</sub> m<sup>-2</sup> yr<sup>-1</sup>, Cole et al. 2007). Additionally, though earlier estimates relied more on C<sub>CO2</sub> calculated from pH and alkalinity (Raymond et al. 2013; Cole et al. 2007), more recent estimates used more often direct measurements (Holgerson and Raymond et al. 2016; DelSontro et al. 2018). Differences in lake CO<sub>2</sub> evasion estimates are more driven by variation in estimates of lake area than by areal emission rates. Estimates of global average emission rates per water surface area for lakes and reservoirs (392–638 g CO<sub>2</sub> m<sup>-2</sup>yr<sup>-1</sup>) are about one order of magnitude lower than those for streams and rivers (1,492–10,644 g CO<sub>2</sub> m<sup>-2</sup>yr<sup>-1</sup>, Table 1).

In comparison to lakes, reported global average emission rates per water surface area for reservoirs are slightly higher (312–686 versus 257–348 g CO<sub>2</sub> m<sup>-2</sup>yr<sup>-1</sup> for reservoirs and lakes, respectively) (Table 1). This may in part be due to the different geographic distribution of both types of standing water bodies;

with lakes being particularly abundant in high latitudes where average emission rates tend to be lower (Aufdenkampe et al. 2011). Nonetheless, the current estimates place total CO<sub>2</sub> evasion from reservoirs more than one order of magnitude lower than that from lakes (see Table 1, when excluding the estimate by Cole et al. 2007), following its low share in the global surface area of standing water bodies. However, the inventory for global reservoirs (which is growing) is far from complete and thus surface area might pose the largest uncertainty for CO<sub>2</sub> evasion from reservoirs. Note that Cole et al. (2007), based on data from St. Louis et al. (2000), estimated a reservoir CO<sub>2</sub> emission of 1.03 Pg CO<sub>2</sub> yr<sup>-1</sup>, i.e. about one order of magnitude higher than the other estimates listed in Table 1. This number is based on a first-order estimate of the total surface area of reservoirs including smallest systems such as farm ponds. This estimated total area is about 5 times larger than that of reservoirs accounted for in recent inventories. While this first order estimate is an eye-opener for the underestimate related to the exclusion of these small systems, it is also highly uncertain and represents an expert opinion rather than a reproducible number. Note further that Cole et al. (2007) estimated a much lower CO<sub>2</sub> evasion rate from lakes, for which they rely on a much more conservative estimate of surface area which excludes smaller systems. In that regard, their emission estimate for standing waters is not consistent. Note finally that other estimates of CO<sub>2</sub> emissions from reservoirs vs. lakes might be underestimated, as in inventories, where the required information is missing, reservoirs might wrongly have been classified as lakes (see discussion in section 2.4).

### **3.2 Persisting shortcomings and future challenges**

#### **3.2.1 Process understanding**

The most prominent gap in the understanding of the processes that drive inland water CO<sub>2</sub> emissions is the question of where the emitted CO<sub>2</sub> is sourced from. A part of the emitted CO<sub>2</sub> may be produced in-situ from the oxidation of allochthonous organic carbon, while another part might stem from inflows of water supersaturated in CO<sub>2</sub> produced during respiration in upland soils and wetlands. Further, this respiration comprises both heterotrophic respiration of plant and soil organic matter as well as autotrophic root respiration. Knowledge about the source of the aquatic CO<sub>2</sub> emissions is of paramount importance for the integration of these fluxes in the overall C budget of continents, as highlighted in the perspective article by Abril & Borges (2019). While earlier studies assumed that the net-CO<sub>2</sub> emissions are entirely the product of heterotrophic respiration and could thus be regarded as a fraction of terrestrial net-primary production (Richey et al., 2002), the contributions of autotrophic root respiration demands consideration of these fluxes as part of total ecosystem respiration that counterbalances gross primary production (Abril & Borges, 2019; Lauerwald et al., 2020).

For streams and rivers, it is assumed that most of the emitted CO<sub>2</sub> is sourced from CO<sub>2</sub> produced by respiration in upland soils and wetlands (Abril et al. 2014; Liu et al., 2022). The relative importance of these external CO<sub>2</sub> inputs are highest in headwaters and decrease downstream (Finlay, 2003; Horgby et al., 2019; Liu et al., 2022; Marx et al., 2017). Moreover, it was shown that due to the very high oversaturation of emerging groundwater, a large part of the emission already takes place over a few hundred meters downstream of the freshwater source (Johnson et al., 2008). It would thus be required



to monitor smallest headwaters directly to well capture those hot spots of aquatic CO<sub>2</sub> emission, for which however monitoring data are not available in sufficient quantity (Marx et al., 2017). Assessment of groundwater CO<sub>2</sub> inputs to inland waters would further require knowledge about groundwater C content and residence time (to quantify the outflows), for which data is limited as well (Downing & Striegl, 2018). Stable C isotopes have been used to estimate source contribution of riverine C loads and CO<sub>2</sub> emissions for single aquatic systems (Telmer & Veizer, 1999). But observational data are not yet sufficient for large-scale assessment. Also, these studies do not often include the uppermost parts of the river network where large amounts of external CO<sub>2</sub> inputs are evading to the atmosphere.

In addition, most existing studies of freshwater CO<sub>2</sub> emissions have not yet attempted to include estimates of aquatic net ecosystem production (NEP), that is the difference between aquatic production and respiration, and thus they currently assess inland waters net-CO<sub>2</sub> emissions rather as a black box that is fed by (semi-)terrestrial C inputs. The recent study by Battin et al. (in revision) has nevertheless demonstrated that inclusion of NEP estimates can help to disentangle autochthonous CO<sub>2</sub> production from allochthonous CO<sub>2</sub> inputs even at global scale. This study corroborates the assumption that most of the aquatic CO<sub>2</sub> evasion is derived from external CO<sub>2</sub> inputs. However, also availability of aquatic NEP data is limited and does not allow yet for spatially explicit estimates at global scale. More importantly, diurnal variations in NEP may entail similar variation in pCO<sub>2</sub> and air-water CO<sub>2</sub> exchange. Moreover, predominant sampling during daytime, when CO<sub>2</sub> emissions are lower than at night, may lead to important biases in flux estimations. Gómez-Gener et al. (2021) recently argued that global estimates based on daytime measurements are biased as night time emissions are on average ~30% higher.

To better understand temporal variability and potential “hot-moments” of inland water CO<sub>2</sub> emissions, more process understanding would be required with regard to CO<sub>2</sub> cycling during periods of ice-cover, spring ice-melt, spring freshet, lake-turnover, and extreme events like floods for which observations are generally rare. Only one of the studies included in our synthesis actually accounts for seasonality (Liu et al. 2022), while the other studies completely ignore seasonality in hydrodynamics, including spring freshet. In our study, we use only simplified correction factors for seasonal ice-cover (Denfeld et al., 2018). Further, the estimates of lake and reservoir CO<sub>2</sub> emissions synthesized in our study do not account for contributions during lake-turnover, when emission rates are thought to be highest in boreal to Arctic systems (Sepulveda-Jauregui et al., 2015).

In contrast to, for instance, the estimate by Raymond et al. (2013), we do not account for intermittent drying of inland waters. Few existing studies suggest that during dry periods, exposed beds might show similar CO<sub>2</sub> emission rates as from the water surface when inundated (Keller et al., 2020). A correction might thus not be adequate unless the emissions from seasonally dry beds are taken explicitly into account for terrestrial respired CO<sub>2</sub> flux. On the other hand, no estimate of CO<sub>2</sub> emissions from temporarily flooded areas is available at the global scale. More systematic investigations of flux rates from both temporarily dry falling inland water beds and temporarily flooded areas would help to refine estimates of inland water CO<sub>2</sub> budgets, and to better integrate them into continual CO<sub>2</sub> budgets while avoiding gaps and overlaps with terrestrial and wetland ecosystems.

Though some studies (Deemer et al., 2016; DelSontro et al., 2018; Raymond et al., 2013) have linked CO<sub>2</sub> variability in lakes and reservoirs to predictors such as waterbody size, mean annual precipitation, and ecosystem productivity, the controls on within-system CO<sub>2</sub> spatial and temporal variations are not well understood and effective scaling relationships are still in need to better represent CO<sub>2</sub> evasion from lakes and reservoirs. Further, characterizations of spatial variability within water bodies is rather scarce, and the representativeness of the sampling site within an aquatic system is a large source of uncertainty (Colas et al., 2020). Finally, our estimates of reservoir CO<sub>2</sub> emissions do not account for fluxes from dam outlets, where deep, hypolimnetic water enriched in CO<sub>2</sub> is released. River reaches directly downstream of dams have been reported to show increased pCO<sub>2</sub> while this excess CO<sub>2</sub> is being emitted rapidly over a few tenths of river-km (Calamita et al., 2021; Guérin et al., 2006; Teodoru et al., 2015). However, more systematic observations from these parts of the river system are needed to quantify this source of CO<sub>2</sub> flux at global scale, and to complete the assessment of reservoir CO<sub>2</sub> emissions.

### **3.2.2 Spatial and temporal resolution**

Spatially and temporally resolved estimates of inland water GHG emissions at global scale can help to better understand the role of these fluxes in the overall GHG budget, to include these fluxes in regional budgets, and to evaluate them directly against observations. The realization of spatially and temporally resolved estimates is however limited by the availability of observations and by the utilized estimation techniques. For rivers, global empirical, spatially explicit estimates have already been achieved at several different resolutions, specifically, 231 regions (Raymond et al. 2013), gridded at 0.5 degrees (Lauerwald et al. 2015), and for individual river reaches (Horgby et al., 2019; Liu et al., 2022). For lakes and reservoirs, the regionalized estimate based on 231 regions by Raymond et al (2013) is the only existing spatially explicit estimate of CO<sub>2</sub> emissions at the global scale. Hastie et al. (2018) achieved a spatially explicit, pan-boreal estimate of lake and reservoir CO<sub>2</sub> emissions at 0.5 degree resolution. As the only process-based model approach at global scale, spatially explicit simulations with the land surface model DLEM have been achieved at 0.5 degree resolution for rivers and reservoirs (Tian et al. 2015b). ORCHILEAK - the inland water branch of the land surface model ORCHIDEE has so far only been applied at the continental scale of Europe (Gommet et al., 2022) and in few large scale basins across the world (Bowring et al., 2020; Hastie et al., 2021; Lauerwald et al., 2020).

With regard to temporal resolution, most of the empirical studies published so far represent climatologies of average annual fluxes, often without precise specification of the time frame covered by the observations (Regnier et al., 2022). The only exception at global scale is the study by Liu et al. (2022) which presents a climatology of average monthly emission fluxes from rivers, thus representing the typical seasonal cycle of riverine emissions. The process-based model DLEM simulates time-series of riverine and reservoir CO<sub>2</sub> emission which reflect both seasonal and interannual variability. In general, a physically-based model approach appears to be the most promising strategy to obtain seasonal and interannual variations in response to climate variability, for present day but also for scenario dependent future projections (Tian et al., 2015b; Hastie et al., 2021; Lauerwald et al., 2020).

While empirical studies have highlighted the importance of diurnal variation in water-air CO<sub>2</sub> exchange, temporal variations at this time-scale are not yet possible to include in estimates. Process-based models

like DLEM (Tian et al. 2015b) or ORCHILEAK (Lauerwald et al., 2017) represent aquatic CO<sub>2</sub> emissions as net-emissions driven by allochthonous inputs of CO<sub>2</sub> and net-instream respiration. A simulation of the diurnal variations would however require the representation of autochthonous aquatic production, which is not yet possible.

### 3.2.3 Data Requirements

As for all GHGs, data required to improve inland water CO<sub>2</sub> emission estimates include in first place direct observation of emission rates. Many earlier estimates relied heavily on partial pressures of CO<sub>2</sub> (pCO<sub>2</sub>) calculated from pH and alkalinity (Lauerwald et al., 2015; Raymond et al., 2013), which has been demonstrated to be a significant source of error leading to an overestimation of pCO<sub>2</sub> particularly in freshwaters with low buffer capacity against acidification (Abril et al., 2015; Golub et al., 2017; Hunt et al., 2011; Liu et al., 2020). Liu et al. (2022), relied on direct pCO<sub>2</sub> observations and suggested that average pCO<sub>2</sub> in global streams and rivers obtained by Raymond et al. (2013) is by 30% too high.

However, as alkalinity and pH are easier to measure, a vast amount of data is available from a large number of studies and in datasets from environmental agencies, with greater spatial and temporal coverage (Hartmann et al., 2014) than that of direct observation. Nevertheless, as potential biases are hard to correct at large scales, a clear preference should be given to directly observed pCO<sub>2</sub> values. Liu et al. (2022) synthesized 5,910 direct pCO<sub>2</sub> observations from 63 studies, which represents about the latest inventory of available data for stream and river systems at global scale. Delsontro et al. (2018) synthesized literature data for 7824 lakes and reservoirs. The number of direct pCO<sub>2</sub> observations is still limited, but steadily growing.

Likely for logistical reasons, most observations are from developed countries which contribute most to the research of inland water GHG budgets. For this reason, systems from temperate climate regions are better represented than tropical or high-latitude systems in remote areas (e.g., compare Liu et al. 2022, Deemer et al. 2016). However, it is these remote areas that play a potentially important role, considering the extensive lake areas in Boreal to Arctic regions, and the large river systems of the humid tropics. There has been notable progress in sampling tropical (Africa (Borges et al., 2019; Borges et al., 2015), Amazon (Abril et al., 2014; Rasera et al., 2013), and SE Asia (Wit et al., 2015)) and high latitude systems (Siberia: (Karlsson et al., 2021; Serikova et al., 2019), Alaska: (Sepulveda-Jauregui et al., 2015)). Despite these advancements, more observations from these poorly monitored areas would help to improve estimates of global inland water CO<sub>2</sub> emissions.

Further, small water bodies require more attention in sampling campaigns. Holgerson & Raymond (2016) have highlighted the potentially important contribution of small lakes and ponds to global inland water CO<sub>2</sub> emissions. However, a regionalized estimate was not yet possible as observations of emission rates are still scarce, and more importantly, as no spatially explicit dataset exists yet that would represent such small water bodies. Datasets that present the smallest water bodies (< 1 km<sup>2</sup>) reliably would help to better integrate these important CO<sub>2</sub> sources into regionalized, global estimates.

Finally, increasing the number, variety and representativeness of investigated systems is only one step to reduce uncertainties in large scale estimates of inland water CO<sub>2</sub> emissions. Temporal and small scale spatial variations with small stream networks (Natchimuthu et al., 2017) and with lakes (Natchimuthu et al., 2017) and reservoirs (Colas et al., 2020) are substantial, and the choice of one or few sampling locations and a limited measurement period lead to large uncertainties and may introduce biases in the flux estimate for the whole waterbody. Improved investigation of CO<sub>2</sub> budgets of single systems requires measurements at various locations within a stream network or water body. In addition, the observations should be taken over a time period long enough to assess seasonal and inter-annual variability, and at a high enough frequency to assess short term variations, including diurnal variations. In particular, datasets covering longer time periods such as those assembled for the US (Jones et al., 2003), China (Ran et al., 2021) and the boreal biome (Lapierre et al., 2013) are crucially needed to evaluate the extent to which trends simulated by LSMs are realistic (Regnier et al., 2022). The development and deployment of automated data loggers is a promising strategy for achieving this objective (Bastviken et al., 2015).

## **4 Inland water CH<sub>4</sub> budget**

### **4.1 Overview of existing estimates**

Global estimates of aquatic CH<sub>4</sub> emission range from 1.5 to 30 Tg CH<sub>4</sub> yr<sup>-1</sup> for streams and rivers, from 42 to 151 Tg CH<sub>4</sub> yr<sup>-1</sup> for lakes, from 9.8 to 52 Tg CH<sub>4</sub> yr<sup>-1</sup> for reservoirs, and from 16 to 331 Tg CH<sub>4</sub> yr<sup>-1</sup> for estimates that lumped lakes and reservoirs together (Table 2). The range in these emission estimates is generally more dramatic than for either CO<sub>2</sub> or N<sub>2</sub>O (see sections 3 and 5, respectively), with the exception of global CO<sub>2</sub> emission estimates from rivers and streams.

Some of the variation in global CH<sub>4</sub> emission estimates is due to large differences in the waterbody surface areas applied. For example, the earliest estimate of CH<sub>4</sub> emissions from reservoirs used a very rough estimate of surface area, multiplying the surface area of reservoirs in the *World Register of Dams* by a factor of four under the assumption that this would better represent the total surface area including small reservoirs and farm ponds not included in that register (St. Louis et al. 2000). This approach resulted in a surface area that is approximately three times larger than any subsequent estimate. Conversely, the earliest estimate from streams and rivers was conservative in that it applied a surface area for larger rivers only (quantifiable from global maps as the GLWD; Bastviken et al. 2011), resulting in approximately a factor of two reduction compared to subsequent estimates that also account for smaller rivers and streams. While most global estimates have ignored ice cover, Johnson et al. (2021) produced an estimate of reservoir emissions that accounted for this effect and which resulted in a CH<sub>4</sub> emission of 10 Tg CH<sub>4</sub> yr<sup>-1</sup> that is half or less of any previous assessment. Harrison et al. (2021) incorporated ice cover correction into their global reservoir emission estimate, resulting in similarly low emissions from reservoir surfaces (9.8 Tg CH<sub>4</sub> yr<sup>-1</sup>), but still yielded a higher total flux due to the inclusion of reservoir turbine degassing (22 Tg CH<sub>4</sub> yr<sup>-1</sup>). Rosentreter et al. (2021) also incorporated an ice cover correction into their assessment of global river (30 Tg CH<sub>4</sub> yr<sup>-1</sup>), lake (151 Tg CH<sub>4</sub> yr<sup>-1</sup>), and reservoir (24 Tg CH<sub>4</sub> yr<sup>-1</sup>) emissions (estimates upscaled from mean emission rates), but in addition also an ice melt overturn correction that reduced the impact of ice cover. Moreover, their corrections did not result in a substantial lowering of the global flux due to increases in the magnitude of areal emission rates applied.

The mean areal emission rates applied to upscaling efforts vary by approximately 2, 3, and 10-fold for reservoirs, lakes, and rivers respectively. In general, there is a temporal trend wherein older datasets have lower average areal emission rates than newer datasets. Part of this trend is due to the treatment of ebullition measurements in older emission estimates. Some global estimates summarized diffusive-only estimates of methane emission (Holgerson and Raymond 2016, Stanley et al. 2016) while others combined diffusive only areal fluxes with ebullitive + diffusive estimates without differentiating one from the other (St. Louis et al. 2000). More recent estimates (Rosentreter et al. 2021, Johnson et al. 2021, 2022) only included studies that estimated both ebullition and diffusion together. Increasing average areal emission estimates may also be due to the increased likelihood of sampling right-skewed data as sample size for water bodies increases (see Wik et al., 2016). For example, a recent dataset of lake and reservoir CH<sub>4</sub> emissions contains some of the highest mean areal fluxes, with about 65% of the estimates contained therein published since 2015 (Rosentreter et al. 2021).

Variation in binned areal emissions (e.g. by latitude, size, and chlorophyll-a) are even larger. For example, Rosentreter et al. (2021) reported an average areal CH<sub>4</sub> flux from the smallest lakes (<0.001km<sup>2</sup>) that is nearly an order of magnitude higher than from lakes in the 0.1-1 km<sup>2</sup> size category, making these smallest systems responsible for 38% of the total lake CH<sub>4</sub> emissions (Rosenteter et al. 2021; Table 2). In addition, Bastviken et al. (2011) reported areal reservoir CH<sub>4</sub> emissions from tropical regions that are an order of magnitude larger than in boreal regions (Bastviken et al. 2011), although follow-up work suggests that this discrepancy may have more to do with a lack of boreal ebullition estimates (Deemer et al. 2016) and the fact that latitude is only a weak predictor for reservoir CH<sub>4</sub> emission (Deemer and Holgerson 2021, Johnson et al. 2021). Conversely, Rosentreter et al. (2021) report average areal CH<sub>4</sub> emissions from rivers that varied by a factor of about four by latitudinal bin, with the subtropical region (10-25 degrees absolute latitude) producing the highest areal emissions and the temperate region (25-40 degrees absolute latitude) producing the lowest areal emissions (Rosentreter et al. 2021).

While a variety of upscaling methods have been used to estimate inland water CH<sub>4</sub> emission, there does not appear to be any directional bias in the resulting estimates, that is one type of approach does not seem to systematically produce higher or lower emissions than other approaches. Many early estimates and some more recent estimates have applied the simplest empirical upscaling wherein a single areal flux was applied to a global surface area of lakes and/or reservoirs (St. Louis et al. 2000, Deemer et al. 2016 DelSontro et al. 2018), and rivers (Stanley et al., 2016). Other estimates have binned lakes and reservoirs CH<sub>4</sub> fluxes based on latitude (Bastviken 2011), waterbody surface area (Holgerson and Raymond 2016; Bastviken et al. 2004), primary productivity (e.g. chlorophyll a concentration; DelSontro et al. 2018), or has used some combination of these approaches (Rosentreter et al. 2021). For rivers, binning has so far only been based on latitude (Bastviken 2011, Rosentreter et al., 2021). Finally, the most recent efforts to model lake and reservoir CH<sub>4</sub> flux have used a gridded approach that considers a variety of factors likely to influence the spatial variations in CH<sub>4</sub> emission including temperature, nutrients, and latitudinal variation in emission factors (Stavert et al. 2021, Johnson et al. 2021, Harrison et al. 2021).

**Table 2:** Existing global estimates of lake, reservoir, and river CH<sub>4</sub> flux. For each estimate, the total water surface area ( $\Sigma A_{water}$ ), the total CH<sub>4</sub> emission flux ( $\Sigma CH_{4em}$ ) and the area weighted average emission rate ( $\Sigma CH_{4em}/\Sigma A_{water}$ ) are reported.

Reference	$\Sigma CH_{4em}/\Sigma A_{water}$ g CH <sub>4</sub> m <sup>-2</sup> yr <sup>-1</sup>	$\Sigma A_{water}$ [10 <sup>6</sup> km <sup>2</sup> ]	$\Sigma CH_{4em}$ Tg CH <sub>4</sub> yr <sup>-1</sup>	Method
<i>Rivers</i>				
Bastviken 2011	4	0.36	1.5	Upscaling from observations
Stanley et al. 2016	41.4*	0.65	27*	Upscaling from observations
Rosentreter et al. 2021	66.5	0.77	30	Upscaling from observations
<i>Lakes and Reservoirs</i>				
DelSontro et al. 2018	61.7	3.23-5.36	199-331	Upscaling from observations
-"	33.7	4.42	149 (95-236) <sup>a</sup>	Statistical prediction
-"	34.5	5.36	185 (119-295) <sup>a</sup>	Statistical prediction
-"	32.2	3.23	104 (67-165) <sup>a</sup>	Statistical prediction
Holgerson and Raymond 2016	2.7*	5.98	16*	Upscaling from observations
Stavert et al. 2021	32.4	2.93	95	Statistical prediction
<i>Lakes (including lakes with dams)</i>				
Bastviken et al. 2004	0.12-122.9	2.8	8-48	Upscaling from observations
Bastviken et al. 2011	19.2	3.7	72	Upscaling from observations
Rosentreter et al. 2021 <sup>&amp;</sup>	54.1	3.71-5.69	151	Upscaling from observations
Johnson et al. 2022 <sup>&amp;</sup>	15	2.8	42 ± 18 <sup>b</sup>	Model
<i>Reservoirs</i>				
St. Louis et al. 2000	35	1.5 <sup>s</sup>	52	Upscaling from observations
Bastviken 2011	40.1	0.5	20	Upscaling from observations
Deemer et al. 2016	58.5	0.31	17 (12-30) <sup>a</sup>	Upscaling from observations
Rosentreter et al. 2021 <sup>&amp;</sup>	63.8	0.26-0.58	24	Upscaling from observations
Harrison et al. 2021 <sup>&amp;</sup>	28.3(62.9) <sup>#</sup>	0.35	9.8 (22) <sup>#</sup>	Model
Johnson et al. 2021 <sup>&amp;</sup>	33.3	0.3	10	Model

<sup>a</sup> lower and upper 90% (Raymond et al. 2013; Lauerwald et al. 2015; Deemer et al. 2016) or 95% (DelSontro et al. 2018) CI

<sup>b</sup> standard error

<sup>c</sup> min and max estimate

& estimate accounts for effects of seasonal ice cover

\* only diffusive emissions

# includes emissions from turbines

## 4.2 Persisting shortcomings and future challenges

### 4.2.1 Process understanding:

Significant progress has been made towards describing the drivers of lake and reservoir CH<sub>4</sub> flux, which may help improve our understanding of the spatial and temporal variability in emissions in the future. Specifically, small, shallow, productive, and low latitude lakes and reservoirs have been found to show higher areal methane emissions than larger, deeper, less productive, high latitude systems (Deemer and Holgerson 2021). In northern systems, methane emissions are often further binned by lake type, with yedoma, peat, and glacial lakes exhibiting different patterns and magnitudes of emission (Wik et al. 2016; Matthews et al. 2020; Kuhn et al. 2021). Less is known about the key drivers of river CH<sub>4</sub> flux. Two of the three existing global estimates of river and stream CH<sub>4</sub> flux use latitude to bin emissions, but the latitudinal trend does not appear to describe much of the spatial variability (Rosentreter et al. 2021). The earlier dataset compiled by Stanley and others contained many estimates from anthropogenically-impacted rivers and streams (Stanley et al. 2016), and could be one explanation for the high global emission estimate despite only considering diffusive fluxes. Still, the effect of nutrient enrichment and productivity on river methane emissions has not been established the way it has been for lakes and reservoir methane emissions (Beaulieu et al. 2019).

Temperature is generally considered an important predictor of aquatic CH<sub>4</sub> emission and relationships between temperature and CH<sub>4</sub> flux have been used to scale seasonal emissions from reservoirs (Prairie et al. 2021; Johnson et al. 2021; Harrison et al. 2021). Such temperature-corrections address biases in many flux observations where measurements are focused during the spring-to-fall period whereas lower emissions during the ice-free winter period are typically not recorded. While there is compelling cross-ecosystem evidence of increasing CH<sub>4</sub> emission with increasing temperature (Yvon-Durocher et al., 2014) there are also examples of systems where CH<sub>4</sub> oxidation is able to keep pace or surpass CH<sub>4</sub> production at higher temperatures (Duc et al. 2010; Shelley et al., 2015). A recent synthesis of CH<sub>4</sub> oxidation in lakes and reservoirs showed that CH<sub>4</sub> oxidation efficiency declines with ecosystem productivity (e.g. trophic status, D'Ambrosio & Harrison, 2021). Another recent study showed experimental evidence that CH<sub>4</sub> oxidation may be phosphorus-limited in northern lakes, also providing further evidence of interactions between lake CH<sub>4</sub> dynamics and nutrient levels (Sawakuchi et al. 2021). Future work could improve our process understanding of methane emission dynamics by disentangling

the role of temperature and productivity in driving both total emission and the balance between methane production and consumption.

Within a single waterbody, CH<sub>4</sub> emissions generally vary substantially in space and time (Wik et al. 2016), and this variation is likely more substantial than for either CO<sub>2</sub> or N<sub>2</sub>O. This spatial and temporal variability has been shown to cause bias in upscaling, where too few measurements in either space or time can lead to underestimation of fluxes (Wik et al. 2016). While the regionalization exercise carried out in our companion paper (Lauerwald et al., this issue) begins to address seasonality by applying an ice cover and ice melt correction, future work should aim to better constrain temporal variability in methane fluxes within single water bodies. Temporal variability can arise from seasonal dynamics such as ice melt (Denfeld et al., 2018), fall turnover (Mayr et al., 2020), seasonal water level changes (Varadharajan et al., 2010), or in response to phytoplankton blooms (Waldo et al., 2021). Diel variation can also be important. Daytime sampling might overestimate CH<sub>4</sub> flux in lakes (Sieczko et al., 2020), but may underestimate it in wetlands (Anthony & MacIntyre, 2016; Godwin et al., 2013; Poindexter et al., 2016). Episodic events can also be the source of large temporal variation such as water level drops in reservoirs (Harrison et al., 2017), storm-driven drops in hydrostatic pressure (Mattson & Likens, 1990) or increases in wind shear stress (Joyce & Jewell, 2003). For rivers, elevated discharge can lead to higher methane fluxes, especially in small high-gradient streams where methane is sourced predominantly from groundwater (Natchimuthu et al., 2017). Spatial variability in aquatic methane fluxes can arise for both biological and physical reasons. In lakes and reservoirs, higher fluxes are observed due to elevated organic matter processing in inlets (DelSontro et al., 2011) and near the shores (Natchimuthu et al. 2016; Peixoto et al. 2015), to the accumulation of organic matter behind run-of-river dams (Maeck et al., 2013), or due to more general heterogeneity of the sediment matrix and associated seeps (Walter Anthony & Anthony, 2013). In rivers, physical features such as waterfalls can be particularly important sites for CH<sub>4</sub> emissions (Natchimuthu et al., 2017). At larger scales, high gradient headwater streams comprising <1% of catchment stream surface area can contribute 30% of catchment emissions, emphasizing the need to sample throughout a catchment rather than attempting to capture network-wide flux via single measurements at river mouths (Natchimuthu, Wallin, et al., 2017).

#### **4.2.2 Spatial and Temporal Resolution**

At global scale, gridded estimates of inland water CH<sub>4</sub> emissions exist for reservoirs (Johnson et al., 2021), lakes (Johnson et al. 2022) and lakes and reservoirs (Stavert et al., 2022). For rivers, disaggregating global fluxes over broad latitudinal zones (Bastviken et al. 2011; Rosentreter et al. 2021) seems still the best possible practice. Most existing global estimates for lakes and rivers represent climatologies of annual fluxes that do not resolve the seasonal and interannual variability, and longer-term trends. Using a relatively simple, process based model, Johnson et al. (2021, 2022) were able to represent the seasonality in lake and reservoir CH<sub>4</sub> emission forced by temperature and ice-cover as drivers. Long-term trends in lake CH<sub>4</sub> emissions due to climate change have been predicted for the hol-arctic/boreal region using a more complex process-based model (Tan & Zhuang, 2015, 2015). Other complex, process-based models of lake CH<sub>4</sub> cycling have been developed (e.g. Lake 2.0, Stepanenko et al., 2016), but have not been applied at large-scales. In contrast to CO<sub>2</sub> and N<sub>2</sub>O, no efforts to model river CH<sub>4</sub> emissions at regional to global scales have been published yet, which may partly be due to the



relative small role of rivers in inland water CH<sub>4</sub> emissions as well as to the complexity of processes involved and the scarcity of data for model calibration and evaluation.

### 4.2.3 Data Requirements

One critical uncertainty for the inland water CH<sub>4</sub> budget is the inability to resolve the location and total surface area of the smallest lakes. Waterbodies <0.001 km<sup>2</sup> have been recently estimated to comprise 37% of the lentic methane flux (Rosentreter et al. 2021). Given high variability in areal emissions from these smallest lakes it is also important to increase effort in sampling these systems to reduce uncertainty. In addition to very small lakes, sampling effort should be increased for large lakes (>1km<sup>2</sup>) and small reservoirs (<1km<sup>2</sup>) (Deemer & Holgerson, 2021). Given the additional importance of depth and productivity in regulating lentic CH<sub>4</sub> flux, spatially resolved information about the depth, chlorophyll *a*, oxygen and dissolved organic carbon concentration of lakes and reservoirs will also help improve regional budgets (and overall upscaling efforts). More generally, systematic, long-term monitoring programs are needed which account for the high spatio-temporal variability in areal emission rates, in particular for ebullition, to better constrain the emissions even for individual, monitored systems. Long time-series of observations may finally help to better constrain the evolution of CH<sub>4</sub> production and emission in response to environmental change and climate extremes like droughts and heatwaves. This need for more and better observational data can hardly be satisfied with conventional methods, but would require the deployment of automatized observation systems and the use of remote sensing data, for which more research and development is needed.

For rivers, many estimates of CH<sub>4</sub> emission rely on pairing concentration data with estimates of gas transfer ( $k_{GHG}$ ) especially in low order streams (see section 2.1 for further discussion). These low order systems have been observed to contribute disproportionately to CH<sub>4</sub> emissions at the catchment network scale despite very low CH<sub>4</sub> concentrations (Natchimuthu et al., 2017), highlighting the need to constrain local values of  $k_{GHG}$  and/or perfect a universal physical model. The development and application of other empirical methods to directly measure GHG flux from low order streams would also help constrain emissions from these systems.

## 5 Inland water N<sub>2</sub>O budget

### 5.1 Overview of existing estimates

N<sub>2</sub>O emissions from inland waters are poorly constrained at the global scale, which is visible in the largely divergent global estimates listed in Table 3: 0.05 - 3.3 Tg N<sub>2</sub>O yr<sup>-1</sup> for streams and rivers and 0.1-0.6 Tg N<sub>2</sub>O yr<sup>-1</sup> for lakes and reservoirs. Most existing global estimates of riverine N<sub>2</sub>O emissions are based on modeled N loads from watersheds and emission factors (EFs), in stark contrast to CO<sub>2</sub> and CH<sub>4</sub> global estimates, which are calculated mainly by empirically upscaling local observations. N<sub>2</sub>O is produced as an intermediate product in denitrification, i.e. the reduction of nitrate to N<sub>2</sub>, but also as a by-product in the process of nitrification, i.e. the oxidation of ammonium to nitrate (Canfield et al., 2010). The amount of N<sub>2</sub>O produced and emitted due to these processes depends on environmental and hydrological factors including water temperature, N availability and speciation, water body depth, oxygen availability, pH, and labile carbon concentrations (Clough et al., 2007; Hu et al., 2019; Outram &

Hiscock, 2012; Rosamond et al., 2012; Venkiteswaran et al., 2014). EFs can be defined as average ratios of N<sub>2</sub>O emission to denitrification and nitrification fluxes. However, it is difficult to quantify nitrification and denitrification fluxes for entire river systems, and even more so at the global scale. Therefore, EFs have traditionally been established by linking N<sub>2</sub>O emissions directly to riverine N loads, implicitly assuming a certain fraction of riverine N loads to be nitrified and denitrified. Mosier et al. (1998) assumed that N leached to the river network was denitrified once and nitrified twice along the river network. Further assuming that 0.25% of both nitrified and denitrified N is emitted as N<sub>2</sub>O, they concluded that 0.75% of the total N leached to the river is emitted as N<sub>2</sub>O. Applying that percentage as EF directly to riverine N loads, they estimated a global riverine N<sub>2</sub>O emission of 1.1 Tg N<sub>2</sub>O yr<sup>-1</sup>. The methodology and EFs established by Mosier et al. (1998) also served to assess the river N<sub>2</sub>O emissions in the 5<sup>th</sup> Assessment Report of the IPCC.

In a similar approach, Seitzinger and Kroeze (1998) and Seitzinger et al. (2000) estimated N<sub>2</sub>O emissions from only the dissolved inorganic fraction (nitrate, nitrite and ammonium) of N (DIN) leached to rivers. Applying EFs of 0.3% and 3% relative to riverine DIN load they estimated a global riverine N<sub>2</sub>O emission of 1.7 (range 0.3-2.9) Tg N<sub>2</sub>O/yr. Over the following decade, these two EF approaches, i.e. the one of Seitzinger and Kroeze (1998) and the IPCC approach derived from Mosier et al. (1998), were updated, yielding consistently large emissions fluxes. Kroeze et al. (2005) estimated 2 Tg N<sub>2</sub>O/yr, and later Kroeze et al. (2010) revised their estimate to 0.5-3.3 Tg N<sub>2</sub>O/yr, both using modeled DIN loads and EFs of 0.3% and 3%. De Klein et al. (2006) predicted a global riverine N<sub>2</sub>O emission of 0.6 Tg N<sub>2</sub>O/yr, while Beaulieu et al. (2011) calculated an emissions flux of 1.1 Tg N<sub>2</sub>O/yr, both using the IPCC approach.

Studies conducted over the last 5-7 years (Hu et al., 2016; Maavara et al., 2019; Marzadri et al., 2021; Yao et al., 2020) consistently calculate N<sub>2</sub>O emissions for rivers that are substantially lower than those of the decades before. Hu et al (2016)'s empirical approach estimated global riverine N<sub>2</sub>O emissions of 51 (19 – 105) Gg N<sub>2</sub>O/yr. Further, the authors report EFs relative to riverine DIN loads of 0.16% to 0.19% to be realistic, suggesting the EFs used by Kroeze and Seitzinger (1998) to be unrealistically high. Maavara et al. (2019)'s spatially resolved stochastic-mechanistic river-continuum model is the first to explicitly represent N transformation processes, and results agreed well with Hu et al.'s predictions, with a global flux of 72-78 Gg N<sub>2</sub>O/yr. Moreover, Maavara et al. (2019) estimated that only 7% and 9% of the total N loads are respectively denitrified and nitrified in the global river network. Thus, the assumption behind the IPCC AR5 approach that all N leached to rivers is once denitrified and twice nitrified also appears to be unrealistic and responsible for gross overestimations.

The studies by Yao et al. (2020) and Marzadri et al. (2021) are complementary as they provide estimates that also account for small streams that contribute disproportionately to the overall riverine N<sub>2</sub>O emissions, but which were ignored in earlier estimates. Marzadri et al. (2021), using a machine learning based approach, reach an estimate of about 114 Gg N<sub>2</sub>O yr<sup>-1</sup>, of which about half is contributed by headwater streams. Note that this is only a near-global estimate which excludes high latitudes > 60°N, which can however be assumed to be small contributors to the global emission due to low N loads and low surface areas of the corresponding river systems (Maavara et al. 2019). Yao et al. (2020), using the land surface model DLEM, estimate riverine N<sub>2</sub>O emissions at even higher values of 458 ±92 Gg N<sub>2</sub>O yr<sup>-1</sup>,

of which 80% stems from small stream emissions up to stream order 3. In their simulations, emissions from these small streams are largely fed by N<sub>2</sub>O inputs from groundwater and saturated soils, which are not accounted for in the other studies. Marzadri et al. (2021), although not representing groundwater N<sub>2</sub>O inputs, still estimate that about ⅓ of total riverine N<sub>2</sub>O emissions is contributed by small streams of orders 3 and lower (see Table 3). In these small streams, nitrification-denitrification processes occur mainly within hyporheic and benthic zones, whereas in larger rivers, the contribution of water column exceeds that of subsurface environments in contributing to N<sub>2</sub>O production (Marzadri et al. 2021). The estimates for larger rivers only by Yao et al. (2020) and Marzadri et al. (2021) agree better with those by Hu et al. (2016) and Maavara et al. (2019).

For lakes and reservoirs, the first global estimates were only published recently. Soued et al. (2016) and DelSontro et al. (2018) gave estimates for the entirety of lakes and reservoirs, without distinguishing between both types of systems while Deemer et al. (2016) estimated N<sub>2</sub>O emission from reservoirs only. Maavara et al. (2019), in their stochastic-mechanistic model of N<sub>2</sub>O emissions from river networks, included explicit emission estimates for reservoirs. Lauerwald et al. (2019) then adapted that model to estimate N<sub>2</sub>O emission from both reservoirs and lakes.

Soued et al. (2016) performed a simple upscaling based on averaged observed N<sub>2</sub>O emissions rates for three latitudinal zones, which yielded with 985±465 Gg N<sub>2</sub>O yr<sup>-1</sup> the highest of the emission fluxes from lakes and reservoirs listed in Table 3. A major limitation of this study was the poor global coverage of observations. While they used data from 298 systems worldwide, they had observations from only six systems for their low latitude estimate, all belonging to the reservoir-class from the study of Guérin et al. (2006). This fact is critical as in their upscaling, lakes and reservoirs from that zone contributed about 80% of their global estimate of N<sub>2</sub>O emissions. Moreover, some of these reservoirs showed extremely high emission rates due to the fact that soils and biomass had not been removed before dam closure, which contributed massively to GHG production and emission (Guérin et al., 2006). It is thus highly probable that these reservoirs are not representative for low latitude lakes and reservoirs, as later discussed in detail in Lauerwald et al. (2019).

Similar to their estimates of lake and reservoir CO<sub>2</sub> and CH<sub>4</sub> emissions (see sections 3 and 4), DelSontro et al. (2018) followed two distinct methodological approaches to obtain their global estimates: a direct upscaling approach based on a global average of observed N<sub>2</sub>O emission rates (252-346 Gg N<sub>2</sub>O yr<sup>-1</sup>), and a statistical approach using a lake/reservoir size classes and classes of chlorophyll-a concentrations as predictors (409-597 Gg N<sub>2</sub>O yr<sup>-1</sup>). Note that the second approach did not lead to a spatially explicit estimate, as only global, statistical distributions of size classes and chlorophyll-a concentrations were used for upscaling. Further, the statistical upscaling equation had a very low predictive power with an R<sup>2</sup> below 0.1.

Deemer et al. (2016) performed a simple upscaling to estimate N<sub>2</sub>O emissions from reservoirs only, obtaining a global flux of about 47 Gg N<sub>2</sub>O yr<sup>-1</sup>. Despite the very different approach, Maavara et al. (2019) and Lauerwald et al. (2019) estimated global N<sub>2</sub>O emissions from reservoirs that are comparable to those by Deemer et al. (2016) (see table 3). For the entirety of lakes and reservoirs, Lauerwald et al. (2019) by far the lowest global estimate of 99±64 Gg N<sub>2</sub>O yr<sup>-1</sup>, and which is only about one tenth of what was

estimated by Soued et al. (2016). Comparing their spatially explicit estimate to regional estimates based on direct upscaling, Lauerwald et al. (2019) found that their model results are reasonable. Moreover, they estimated that lakes, although contributing more than 90% of the global surface area of standing water bodies, contribute only about half of the emission flux as a result of their much lower average emission rates. This indicates that it is problematic to lump together lakes and reservoirs in global upscaling exercises.

**Table 3:** Global scale estimates of inland water N<sub>2</sub>O emissions. For each estimate, the total water surface area ( $\Sigma A_{water}$ ), the total N<sub>2</sub>O emission flux ( $\Sigma N_2O_{em}$ ) and the area weighted average emission rate ( $\Sigma N_2O_{em}/\Sigma A_{water}$ ) are reported.

Reference	$\Sigma N_2O_{em}/\Sigma A_{water}$ mg N <sub>2</sub> O m <sup>-2</sup> yr <sup>-1</sup>	$\Sigma A_{water}$ 10 <sup>6</sup> km <sup>2</sup>	$\Sigma N_2O_{em}$ Gg N yr <sup>-1</sup>	Method
<i>Rivers</i>				
Seitzinger & Kroeze 1998 and Seitzinger et al. 2000			1650 (300 – 2940) <sup>c</sup>	Emission factors
Kroeze et al. 2005			1975	Emission factors
Mosier et al. 1998			1100	Emission factors
De Klein et al. 2006			550	Emission factors
Kroeze et al. 2010			470–3300	Emission factors
Beaulieu et al. 2011			1070	Emission factors
Hu et al. 2016			51 (19-105) <sup>a</sup>	Statistical prediction
Maavara et al. 2019			72–78 <sup>c</sup>	Model
Yao et al. 2020			458±92 <sup>b</sup>	Model
" , stream orders 1-3			387±93 <sup>b</sup>	Model
" , stream orders ≥4			71±23 <sup>b</sup>	Model
Marzadri et al. 2021			114	Machine learning+Model
" , stream orders 1-3			76	Machine learning+Model
" , stream orders ≥4			38	Machine learning+Model
<i>Lakes and reservoirs</i>				
DelSontro et al. 2018	78	3.23-5.36	252-424	Upscaling from observation
DelSontro et al. 2018	106	4.42	470 (300- 710) <sup>a</sup>	Statistical prediction
DelSontro et al. 2018	112	5.36	600 (380- 860) <sup>a</sup>	Statistical prediction
DelSontro et al. 2018		3.23	410 (250- 600) <sup>a</sup>	Statistical prediction
	127			
Soued et al. 2016	235	4.20	985±465 <sup>b</sup>	Upscaling from observation
Lauerwald et al. 2019	34	2.93	98±64 <sup>c</sup>	Model
<i>Lakes (including lakes with dams)</i>				

Lauerwald et al. 2019	17	2.68	46±29 <sup>c</sup>	Model
<i>Reservoirs</i>				
Deemer et al. 2016	152	0.31	47 (31-110) <sup>a</sup>	Upscaling from observations
Maavara et al. 2019	148-250 <sup>c)</sup>	0.45	67–112 <sup>c</sup>	Model
Lauerwald et al. 2019	185	0.25	52±33 <sup>c</sup>	Model

<sup>a</sup> lower and upper 90% (Raymond et al. 2013; Lauerwald et al. 2015; Deemer et al. 2016) or 95% (DeSontro et al. 2018) CI

<sup>b</sup> standard error

<sup>c</sup> min and max estimate

## 5.2 Persisting shortcomings and future challenges

### 5.2.1 Process understanding

While a basic understanding of processes involved in aquatic N<sub>2</sub>O cycling exists from a certain number of field studies, the quantification of these processes in large scale estimates is still difficult due to their complexity and the unavailability of sufficient datasets to support their assessment. For this reason, empirical EFs have long been used to estimate riverine N<sub>2</sub>O emissions directly from N loads, assuming a constant fraction of N loads to be nitrified and denitrified within the rivers, independent of size of the river network, its ecoclimatological setting, and anthropogenic pressure. While newer, model-based studies proved the worth of calculating more precise estimates of nitrification and denitrification fluxes taking into account physical constraints such as water residence time and temperature (Maavara et al. 2019, Yao et al. 2020, Marzadri et al. 2021), the actual production and/or emission of N<sub>2</sub>O related to these processes is still based on simple, empirical factors. As N<sub>2</sub>O is formed only as a by-product of nitrification and as an intermediate product in the denitrification process, the actual fraction of N<sub>2</sub>O produced from these processes is highly variable and not yet possible to reproduce based on mechanistic formulations.

For a better assessment of inland water N<sub>2</sub>O cycling, a better distinction between processes in the water column and in the sediments is required. That this distinction is indeed possible in global scale assessments of river N<sub>2</sub>O emissions was demonstrated by Marzadri et al. (2017, 2021). In particular, the top-layer of aquatic sediments is often a zone of sharp transition between nitrifying (oxic) and denitrifying (anoxic) conditions, and thus a hot spot of N cycling and N<sub>2</sub>O production. Here, anoxic water rich in ammonium mixes with oxygen-rich waters, promoting nitrification, while in turn nitrate produced from nitrification diffuses down and fuels denitrification in the anoxic zone (Beaulieu et al., 2015). The relative importance of water column vs. sediment processes may change along the river network. Following the conceptual model by Marzadri et al. (2017), denitrification of emergent, ammonium rich groundwater in streambed sediments is the dominant source of N<sub>2</sub>O in headwaters, but its importance decreases downstream, until finally denitrification in the lower water column dominates. Further, it was shown that dissolved N<sub>2</sub>O inputs from groundwater and waterlogged soils feed an overproportional contribution of headwaters to riverine N<sub>2</sub>O emissions (Billen et al., 2020; Yao et al., 2020). Note that the global assessments of river N<sub>2</sub>O emissions by Marzadri et al. (2021) and Maavara et al. (2019) do not account for groundwater N<sub>2</sub>O inputs which represent an important part of inland water N<sub>2</sub>O emissions.

For lakes and reservoirs, the importance of processes in the benthic zone has been implicitly taken into account by the use of “hydraulic load” to scale denitrification rates (Harrison et al., 2009). Hydraulic load has been defined as the ratio of water inflow to water surface area, which is identical to the ratio of average lake or reservoir depth over water residence time (Harrison et al. 2009). The process of denitrification is assigned an “apparent settling velocity” which expresses rates of nitrification or denitrification in the benthic zone relative to water column depth. The deeper the average lake or reservoir, the longer it takes until the whole volume is nitrified or denitrified. However, this approach does not take into account the actual shape of the lake/reservoir bed and the proportions of shallow, littoral zones, where emitted  $N_2O$  is mainly produced in the bed sediments (Liikanen et al., 2003; Zhu et al., 2015), vs. the deeper zones, where processes in the water column are the dominant source of  $N_2O$  (Mengis et al., 1997). While streams and rivers are usually well mixed, deeper lakes and reservoirs may be temporally stratified, with important consequences for  $N_2O$  cycling, which have so far not been taken into account in large scale assessments. During stratification, only the top layer (epilimnion) is exchangeable with the atmosphere and thus well oxygenated. Then, nitrification in the epilimnion is the main source of  $N_2O$  emissions (Beaulieu et al. 2015, Mengis et al. 1997). In anoxic parts of the lower layer (hypolimnion), denitrification prevails, which can be a source or sink of  $N_2O$ , depending on the availability of nitrate for reduction (Beaulieu et al. 2015, Mengis et al. 1997). As this anoxic water may also be rich in ammonium from the in-situ decomposition of organic matter, mixing with more oxygenated, epilimnetic waters during lake turn-over may represent a “hot moment” for nitrification and  $N_2O$  emissions (Beaulieu et al., 2015; Roland et al., 2017). However, a quantitative assessment of this hot-moment at large scales is yet not possible due to the lack of observational data. Moreover, while a certain number of studies report measurements of  $N_2O$  concentrations in the shallow, easy to reach epilimnion, studies investigating the deeper profile of  $N_2O$  concentrations through the hypolimnion are scarce (Mengis et al. 1997, Beaulieu et al. 2015).

Further, also resolving the horizontal zonation would help to better assess the overall  $N_2O$  budget of a lake. Within larger lakes, shallow littoral zones have been shown to contribute disproportionately to lake  $N_2O$  emissions relative to their areal extent (Zhu et al 2015). Here, benthic sediments contribute most to  $N_2O$  production, while in the deeper, pelagic zone,  $N_2O$  is produced in the water column, and more specifically, under stratified conditions, in the epilimnion. Yet, most observations are constrained to pelagic zones, which dominate lakes and reservoirs with regard to surface area, but not necessarily emissions. Further, strong horizontal gradients in  $N_2O$  emissions rates may be formed towards the points of riverine inflows of reactive N (Miao et al., 2020). However, few studies conduct systematic sampling which could reveal and account for these internal spatial variations.

In general, observational studies are skewed towards temperate, eutrophic systems in developed countries, which are easily accessible for sampling and which represent potentially important  $N_2O$  sources related to water quality issues caused by agricultural non-point sources and sewage water injections. In boreal regions where N loads are usually lower, it was demonstrated that a substantial proportion of aquatic systems is undersaturated with  $N_2O$  and thus rather act as sinks for this GHG (Kortelainen et al., 2020; Soued et al., 2016). Further, as for  $CO_2$  and  $CH_4$ , observations of smaller water bodies are generally underrepresented. Interestingly, though small, agricultural ponds could be hypothesized to be strong GHG emitters, a study of 101 such systems across the US (Webb et al., 2019) has shown that about two thirds of these systems are on the contrary  $N_2O$  sinks. Overall, this imbalance in observed systems might have introduced a bias in upscaling towards overestimation of fluxes. Moreover, most estimation approaches, particularly the use of EFs, do not permit for representing inland waters as  $N_2O$  sinks. Finally, samples from temperate and high latitude systems are skewed towards summer months, while the full seasonal cycle is only rarely covered in observational studies. Kortelainen

et al. (2020) have demonstrated in their study on Finnish lakes that there is a strong seasonality in N<sub>2</sub>O concentrations and emission rates, with much higher values in winter when low autotrophic production allows for higher nitrate concentrations. A flux estimate based on summer-time observations only would thus have led to an underestimation by a factor of four. Assuming that similar seasonal patterns are to be observed in other temperate to high latitude systems, and that in particular lake turnover as hot moment of N<sub>2</sub>O emissions is not well captured in observations, we can hypothesize that the non-representativeness of sampling times might have introduced a negative bias in upscaling exercises. Note finally that the non-representativeness of observations does not only affect estimates based on direct upscaling of average emission rates. With the lack of representative observational data for calibration and validation, also model-based studies will remain of limited validity.

### **5.2.2 Spatial and temporal resolution**

For riverine systems, global, spatially explicit estimates of N<sub>2</sub>O emissions have been achieved by numerous studies. The spatial resolution ranges from large river basins (Hu et al., 2016), over gridded estimates (Yao et al. 2020) to estimates per stream segment (Marzadri et al., 2021). For lakes, Soued et al. (2016) have resolved N<sub>2</sub>O emissions for three broad latitudinal bands and the only published spatially explicit, gridded estimate is that of Lauerwald et al. (2019). Most of these studies represent a climatology of average annual fluxes. Only the process-based model by Yao et al. (2020) allows for spatio-temporally resolved simulation results which cover seasonality, interannual variability and long-term temporal trends. So far, this model includes rivers and reservoirs. For lakes, a global scale, process based model that permits for temporally varying N<sub>2</sub>O emissions is still missing. In line with what was discussed in the preceding subsection, such a model would need to couple lake physics and biogeochemistry.

### **5.2.3 Data requirements**

To achieve better global estimates of inland water N<sub>2</sub>O emissions, more observational data is needed, in particular from systems that are so far underrepresented like lakes of high latitude and tropical areas, in general oligotrophic systems that are not susceptible to yield high N<sub>2</sub>O emissions and may even be sinks, the smallest systems including natural and farm ponds, and ponds used for aquaculture. In general, more systematic observational programs permitting the quantification of seasonality and the impact of seasonal ice cover, lake turn-over and algae blooms to annual emissions are needed to avoid biased upscaling of annual flux estimates. Finally long-time series are needed to assess the long-term evolution of inland water N<sub>2</sub>O emissions and to evaluate process based models.

To support the application of more advanced upscaling approaches in the estimation of inland water N<sub>2</sub>O budgets, including process based models, better data on environmental drivers and boundary conditions are required. That includes the representation of reactive N species to inland waters. While global estimates of total N and DIN inputs to the river network exist (Mayorga et al., 2010), it would be even better to have information on the more specific inputs of nitrate, ammonium, and dissolved N<sub>2</sub>O to set the boundary conditions for processes involved in N<sub>2</sub>O production, reduction and emissions. Further, the model representation of N and N<sub>2</sub>O cycling in inland water would profit from datasets on bed morphology of the water body and properties of bed sediments. Marzadri et al. (2021) have used a machine learning approach to estimate all these boundary conditions for application of their model of stream N<sub>2</sub>O production. While this seems a promising strategy, this approach could be steadily improved with new findings from field observations and improved datasets of predictor variables. Also for lakes

and reservoirs, only estimates of volume and average lake depth are available (Messenger et al. 2016), which could be steadily improved using a similar strategy. Finally, for the better assessment of lake and reservoir N<sub>2</sub>O budgets, physical processes such as stratification, mixing and ice cover would need to be represented dynamically. For example, process based models of lake physical processes have been developed and even implemented into land surface models for global scale application (Subin et al., 2012) and the outputs of such models may be included in future lake and reservoir N<sub>2</sub>O models.

## **6 Conclusions and outlook**

The number of global scale estimates of inland water GHG emissions is constantly increasing, at an accelerated step. For CO<sub>2</sub> and CH<sub>4</sub>, we see a tendency for increasing numbers in estimates of global scale fluxes following the inclusion of water bodies which contribute significantly to the overall water surface area, and disproportionately to overall emissions. For water bodies above a certain size (e.g. stream orders, lake size), estimates of average emission rates seem to converge in latest estimates. Major discrepancies persist however with regard to the assumed water surface area and the statistical distribution of water body size classes, in particular for small lakes and impoundments (<10 ha), and ponds. For riverine N<sub>2</sub>O emissions on the contrary, we find newer estimates to be lower than older estimates, following a change in methodologies moving away from application of emission factors towards more process oriented modeling. For lake N<sub>2</sub>O emissions, discrepancies in assumed water surface area and distribution of lake size classes still play a role as well, but not as strongly as for CO<sub>2</sub> and CH<sub>4</sub> because small water bodies do not appear to contribute disproportionately to the total emission flux.

There is ongoing work to improve spatially explicit datasets of inland water surface areas that will help improve global scale estimates. For streams and rivers, global scale estimates have recently been largely improved combining high resolution remote sensing of water surface areas and statistical prediction for headwater streams which are too narrow to be detected (GRWL). A similar strategy may also be the solution for lakes and reservoirs, combining data from inventories and remote sensing. Inventories are more reliable but less comprehensive as they exclude smallest water bodies and are susceptible to geographical biases due to differences between national data sources. Remote sensing is able to detect smaller water bodies but is prone to contaminations with wrongly attributed water surface areas if unsupervised algorithms are applied and checks for ground truth in sufficient quantity and quality are not possible.

More importantly, improving inland water GHG emissions estimates requires more fieldwork to improve quantity and quality of observational data. In particular, we need more data from systems in remote areas of the high latitudes and the tropics, and systematic measurements with time-series of sufficient length and frequency of observations to better capture seasonal and inter-annual variability in fluxes as well as long-term trends in response to environmental change. In addition, more attention has to be paid to hot-spots and hot-moments of inland water GHG emissions, which likely contribute a substantial fraction of overall emissions.



For upscaling and predictions to achieve better global scale estimates, recent developments of machine learning based approaches and process-oriented models seem promising. These approaches help to better constrain the spatial-temporal variability in global scale estimates, which helps to better include inland water GHG emissions in regionalized budget efforts such as RECCAP-2, but also in top-down approaches based on atmospheric inversions, which will help to further reduce uncertainties in global estimates.

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