

Methane Ebullition in Temperate Hydropower Reservoirs and Implications for US Policy on Greenhouse Gas Emissions

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Abstract The United States is home to 2198 dams actively used for hydropower production. With the December 2015 consensus adoption of the United Nations Framework Convention on Climate Change Paris Agreement, it is important to accurately quantify anthropogenic greenhouse gas emissions. Methane ebullition, or methane bubbles originating from river or lake sediments, has been shown to account for nearly all methane emissions from tropical hydropower reservoirs to the atmosphere. However, distinct ebullitive methane fluxes have been studied in comparatively few temperate hydropower reservoirs globally. This study measures ebullitive and diffusive methane fluxes from two eastern Washington reservoirs, and synthesizes existing studies of methane ebullition in temperate, boreal, and tropical hydropower reservoirs. Ebullition comprises nearly all methane emissions (>97%) from this study's two eastern Washington hydropower reservoirs to the atmosphere. Summer methane ebullition from these reservoirs was higher than ebullition in six southeastern U.S. hydropower reservoirs, however it was similar to temperate reservoirs in other parts of the world. Our literature synthesis suggests that methane ebullition from temperate hydropower reservoirs can be seasonally elevated compared to tropical climates, however annual emissions are likely to be higher within

tropical climates, emphasizing the possible range of methane ebullition fluxes and the need for the further study of temperate reservoirs. Possible future changes to the Intergovernmental Panel on Climate Change and UNFCCC guidelines for national greenhouse gas inventories highlights the need for accurate assessment of reservoir emissions.

Keywords Methane · Hydropower · Reservoir · Temperate · Greenhouse gas · Ebullition

Introduction

With the December 2015 consensus adoption of the United Nations Framework Convention on Climate Change (UNFCCC) Paris Agreement, it is increasingly important for environmental managers to understand and accurately quantify anthropogenic greenhouse gas (GHG) emissions. This international agreement strives to limit GHG emissions and maintain mean global temperatures within 2 °C of preindustrial levels (UNFCCC 2015). An estimated 30% of the terrestrial carbon carried by inland waters (2.2 Pg y⁻¹) (Battin et al. 2009; Aufdenkampe et al. 2011) is returned to the atmosphere in the form of methane (CH₄) (0.65 Pg y⁻¹) (Bastviken et al. 2011). Numerous studies have demonstrated that hydropower reservoirs have the potential to produce high amounts of CH₄, despite initially being considered carbon-free energy sources (Barros et al. 2011; Rudd et al. 1993; Louis et al. 2000). The United States (US) is home to 2198 dams that have a combined generating capacity of 80 GW (Uría-Martínez et al. 2015; USACE 2015). Over the past decade, US hydropower has invested \$6 billion in improvements to existing plants and increased capacity by 1.4 GW (Uría-Martínez et al. 2015). More

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recently, the 2014 Hydropower Market Report from the US Department of Energy listed 407 hydropower projects under construction and 263 hydropower projects issued authorization for construction (Uría-Martínez et al. 2015). Although the majority of these new projects further develop pre-existing, non power-producing dams, the Department of Energy is also investigating new stream-reach development, where there is an estimated 65 GW of remaining undeveloped resource potential (Kao et al. 2014). If the US plans to accurately account for its anthropogenic GHGs while continuing growth in this energy sector, it is vital to understand how hydropower reservoirs contribute to emissions.

Reservoirs differ from free-flowing rivers and naturally occurring lakes due to high sediment loads (Maeck et al. 2014; Mendonça et al. 2012), flooding of terrestrial biomass during reservoir formation and subsequent fluctuations in water elevation (Chen et al. 2009; Harrison et al. 2017), and other alterations to flow. Additionally, hydropower reservoirs differ from non-power producing reservoirs due to hydrodynamic changes caused by hydropeaking operations in which dam flows are regulated over short time periods to increase or decrease power production in response to energy demands (Ibarra et al. 2015). Sediment, organic carbon deposition, and stratification in reservoirs create anoxic environments conducive to anaerobic microbial methanogenesis (Jacinthe et al. 2012; Sobek et al. 2012). Methane from freshwater environments can be emitted to the atmosphere through diffusion or ebullition (bubbling) (Bastviken et al. 2004). Diffusion and ebullition contribute unequally to the total flux of CH₄ from reservoirs to the atmosphere (Del Sontro et al. 2011, 2010; Fearnside and Pueyo 2012; Grinham et al. 2011; Sturm et al. 2014). In oxygenated waters, aerobic methanotrophy greatly reduces diffusive CH₄ emissions but minimally impacts CH₄ ebullition due to rapid bubble transport through the water column (Bastviken et al. 2004, 2002; Harrison et al. 2017). Globally, it is estimated that 65% of CH₄ emissions from hydropower are ebullitive, yet only 52% of studies measure CH₄ ebullition (Deemer et al. 2016). Ebullitive flux of CH₄ from reservoirs to the atmosphere is therefore one of the most important and understudied pathways for GHG emissions from hydropower.

To date, a limited number of temperate hydropower reservoirs have been studied for distinct ebullitive CH₄ fluxes: Lake Wohlen in Switzerland (Del Sontro et al. 2015, 2010; Diem et al. 2012; Sobek et al. 2012), sub-tropical Little Nerang Reservoir in Queensland, Australia (Grinham et al. 2011), five consecutive hydropower reservoirs along the Saar River in western Germany (Maeck et al. 2014, 2013; Wilkinson et al. 2015), and six hydropower reservoirs in the southeastern US (Bevelhimer et al. 2016). By comparison, ebullitive CH₄ fluxes from boreal and tropical hydropower reservoirs have been moderately well-studied (e.g., Bergier et al. 2011; Del Sontro et al. 2011; Deshmukh

et al. 2014; dos Santos et al. 2006; Duchemim et al. 2006a, 1995; Galy-Lacaux et al. 1997; Huttunen et al. 2002; Keller and Stallard 1994; Kelly et al. 1997; Matthews et al. 2005; Soumis et al. 2005; Tremblay 2005; Trojanowska et al. 2009). In several cases, hydropower reservoirs with CH₄ ebullition measurements classified as “temperate” in synthesis papers using an identification system based on latitude (Barros et al. 2011; Louis et al. 2000) were labeled as “boreal” in the original authors’ assessments (Duchemim et al. 1995; Kelly et al. 1997; Tremblay 2005), and are therefore not considered to be temperate reservoirs for the purpose of this study. Additionally, some research has included diffusive GHG emissions from temperate hydropower reservoirs without distinct ebullitive CH₄ fluxes (e.g., Chen et al. 2011; Mosher et al. 2015; Soumis et al. 2004).

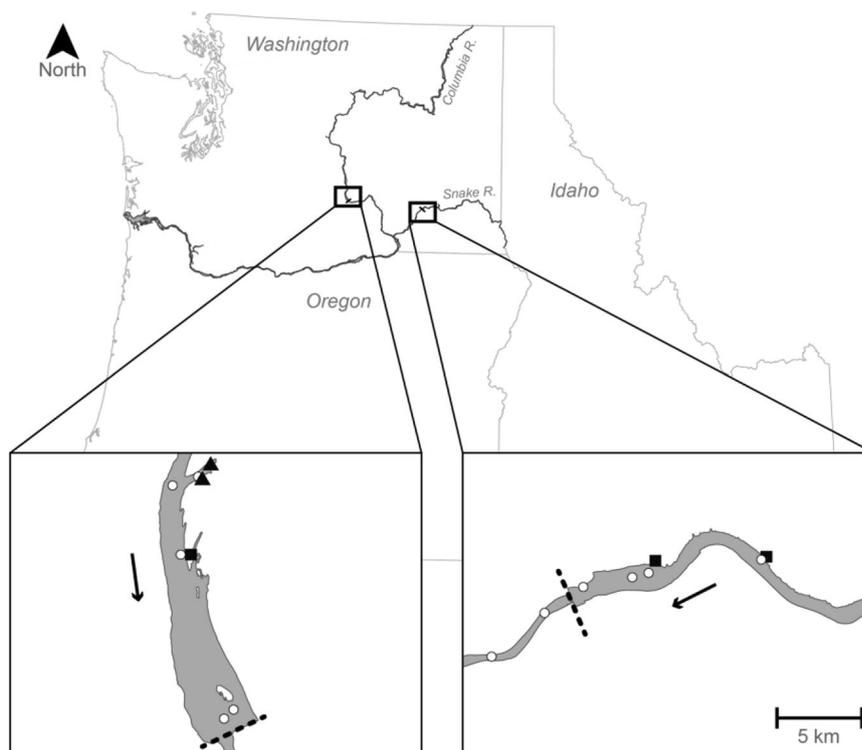
The large number of dams in the US necessitates an accurate understanding of how temperate climates impact CH₄ ebullition relative to CH₄ diffusion in reservoirs. This study aims to (1) contribute to available data by measuring both CH₄ ebullition and diffusion in two hydropower reservoirs located in temperate eastern Washington, (2) contextualize temperate hydropower reservoir CH₄ ebullition in relation to ebullition rates observed in other temperate freshwater environments and boreal and tropical hydropower reservoirs, and (3) investigate the necessity of US hydropower operators quantifying reservoir CH₄ emissions from ebullition.

Methods

Study Locations

Study locations were two run-of-the-river hydroelectric power plants (projects with limited water storage capacity that generate power based on available inflow) in temperate eastern Washington, US: Priest Rapids Reservoir on the mid-Columbia River and Lower Monumental Reservoir on the Snake River (Fig. 1). All study locations are surrounded by a combination of semi-arid shrub-steppe and irrigated agricultural land east of the Cascades Rain Shadow. For brevity, we will use the term “depositional littoral zone” to describe shallow (≤ 10 m of water) sampling sites that represent depositional environments. Two sites in the depositional littoral zones of each hydropower project were selected based on the criteria of accessibility and depth (≤ 10 m) and sampled for CH₄ ebullition during a single, September (summer) 2012 sampling campaign. For CH₄ diffusion at each hydropower plant, at least one major tributary, two depositional littoral zone sites, two main channel, and two forebay sites were sampled in order to represent all reservoir environments. Additionally, two depositional littoral zone sites and two main channel sites

Fig. 1 Priest Rapids (*left*) and Lower Monumental (*right*) sampling sites along the Columbia and Snake River, respectively. *White circles* mark locations of diffusion-only sampling. *Black triangles* mark locations of ebullition-only sampling. *Black squares* mark locations of diffusion and ebullition sampling. Additional sampling sites in the tailrace of Priest Rapids, tributaries along both rivers, and along the free-flowing Hanford Reach are not marked. *Arrows* indicate direction of flow, and *dashed lines* mark the two dams. In total, $n = 12$ for ebullition sampling and $n = 19$ for diffusion sampling



were sampled along a free-flowing, un-impounded reach (the Hanford Reach) on the mid-Columbia River. The Hanford Reach was sampled September 1–2, Priest Rapids Reservoir was sampled September 3–4, and Lower Monumental Reservoir was sampled September 6–7.

Water Quality

Water temperature, dissolved oxygen, water elevation, barometric pressure, nutrients, and dissolved organic carbon (DOC) were measured at all sites. Temperature and dissolved oxygen profiles were recorded by deploying a HACH Minisonde 5 sensor (HACH Environmental, Loveland, Colorado). Water temperatures were used to calculate diffusion according to Henry's Law (see Section 2.4). Minisonde's were also deployed at 0.5 m depth for 24 h at depositional littoral sites in Priest Rapids and Lower Monumental Reservoirs and along the Hanford Reach to measure dissolved oxygen on a diel basis.

Water elevation was monitored hourly at each of our study sites during sampling periods using HACH Minisonde 5 sensors (HACH Environmental, Loveland, Colorado). Hourly barometric pressure data were obtained from one location (Priest Rapids Reservoir), assumed to be representative for the region, and compared to mean daily data for 2012 (CBR 2013).

Nutrient (nitrate and phosphate) and DOC samples were taken from 1 m and 0.5 m from the bottom and combined into a single sample for each sample site. In the smaller

tributaries, these samples were collected from shore. Nutrients were measured in the field using a HACH DR/890 colorimeter (HACH Environmental, Loveland, Colorado). DOC samples were filtered through a Whatman GF/F, 0.45 μm glass fiber filter. Upon return to the laboratory, DOC samples were stored at 5 $^{\circ}\text{C}$ until analysis within 2 months by a total carbon analyzer (Shimadzu TOC 5000 A). Percent sediment organic carbon (SOC) was also measured at depositional littoral sites. Fine sediments (<2 mm) were sampled using a Ponar dredge and percent SOC determined using the loss on ignition method (Heiri et al. 2001).

Ebullition

CH_4 ebullition was sampled in the depositional littoral zone using four, submerged inverted funnels at each of the three study locations ($n = 12$ total) (Del Sontro et al. 2010; Strayer and Tiedje 1978). Inverted funnels were constructed of vinyl with minimal seams and no openings along their interior collection surfaces. The funnels channeled bubbles from a circular, 0.79 m^2 opening at a depth of 2 m into a sealed syringe at their terminus. According to Ostrovsky et al. (2008), CH_4 bubbles collected at this depth in an unstratified water column undergo <5% dissolution before reaching the surface, and Del Sontro et al. (2010) demonstrated that dissolution from 2 m to the surface was "negligible" in Lake Wohlen. Therefore, we assume that CH_4 ebullition sampled at this depth represents emission at the surface. Two inverted funnels were deployed at each site in

the depositional littoral zone ≤ 10 m water depth), allowed to drift >2 m from their moorings on the riverbed, vented of entrained gases, and left for 24 h. Following this deployment period, 12 mL of sample gas was collected with a syringe from each funnel and injected into a pre-evacuated 10 mL serum bottle (i.e., over-pressurized) sealed with a 10-mm thick butyl rubber stopper and crimped aluminum ring. Each serum bottle was then inverted inside an outer 50 mL centrifuge tube filled with deionized water as an additional safeguard against atmospheric invasion. Samples were stored at 5 °C until analysis within 2 months by gas chromatograph (SRI 8610 C FID with methanizer). Ebullition was calculated using the following equation:

$$F_E = \frac{[\text{CH}_4] \cdot V}{t_d \cdot A_f}, \quad (1)$$

where F_E is flux through ebullition ($\text{mg m}^{-2} \text{d}^{-1}$), $[\text{CH}_4]$ is the concentration of CH_4 (mg L^{-1}), V is the volume of CH_4 collected by a funnel (L), t_d is the deployment time (d), and A_f is the cross sectional area of a funnel (m^2).

Scaling ebullitive CH_4 fluxes requires additional data on the extent of the depositional littoral zone in a given reservoir. We classified the depositional littoral zone as areas that were shallow (≤ 10 m) and represented depositional environments. We estimated the total such area using bathymetric data and removed shallow water areas in the main channel likely to experience higher water velocities (Fig. 2). The extent of the depositional littoral zone ranges from approximately 1.8 km^2 in Priest Rapids to 5.9 km^2 in Lower Monumental Reservoir (Fig. 2).

Diffusion

Water from 0.01 m depth ($n = 19$ total) and ambient air from 1 m above the water's surface ($n = 19$ total) were

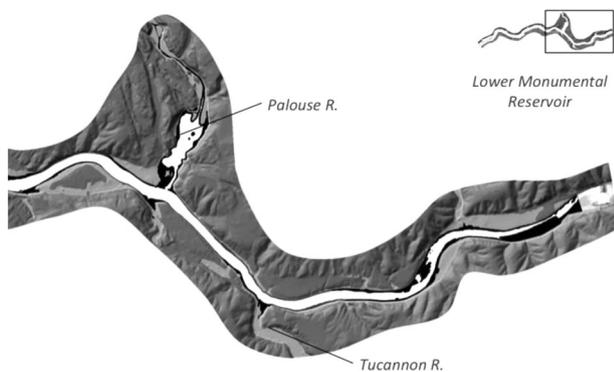


Fig. 2 Depositional littoral zone (black) along the upstream reach of Lower Monumental reservoir on the Snake River (white), based on bathymetric data. The depositional littoral zone was similarly classified along the downstream reach of Lower Monument Reservoir and Priest Rapids Reservoir

sampled at each study location. Water samples were equilibrated with syringes following Kling et al. (2000). A volume of 12 mL of sample gas was injected into a pre-evacuated 10 mL serum bottle (i.e., over-pressurized) sealed with a 10-mm thick butyl rubber stopper and crimped aluminum ring. Each serum bottle was then inverted inside an outer 50-mL centrifuge tube filled with deionized water as an additional safeguard against atmospheric invasion. Samples were stored at 5 °C until analysis within 2 months by gas chromatograph (SRI 8610 C FID with methanizer). Flux of dissolved CH_4 through diffusion across the water's surface (F_D ; $\text{mg m}^{-2} \text{d}^{-1}$) was calculated using in-stream partial pressures and the thin boundary-layer equation:

$$F_D = k \cdot K_H(p\text{Gas}_w - p\text{Gas}_a), \quad (2)$$

where k is the gas transfer velocity (cm h^{-1}), $p\text{Gas}_w$ is the partial pressure of a gas in water (μatm), $p\text{Gas}_a$ is the partial pressure of a gas in water at equilibrium with ambient air (μatm), and K_H is a water temperature-dependent Henry's constant ($\text{mmol kg}^{-1} \text{atm}^{-1}$) (Wilhelm et al. 1977). For large, high order rivers such as the Columbia and Snake, there is evidence that water temperature and wind speed are the dominant physical controls of gas transfer velocity (Alin et al. 2011). This may be a function of their broader width, or fetch (Vachon and Prairie 2013), which also describes run-of-the-river reservoirs such as Priest Rapids and Lower Monumental. The gas transfer velocity, k (cm h^{-1}) was therefore determined using the following relationships:

$$k = k_{600} \left(\frac{Sc}{600} \right)^{-0.66} \quad (3)$$

$$Sc = a - bT + cT^2 - dT^3 \quad (4)$$

where Sc is the Schmidt number, T is the water temperature ($^{\circ}\text{C}$), a , b , c , and d are dimensionless constants for CH_4 (Crusius and Wanninkhof 2003; Wanninkhof 1992), and k_{600} (cm h^{-1}) is the gas transfer velocity at a Schmidt number of 600. k_{600} was calculated using wind speed following Cole and Caraco (1998):

$$k_{600} = 2.07 + 0.215(U_{10})^{1.7}, \quad (5)$$

$$U_{10} = U_1 \left[1 + \frac{(C_{d10})^{0.5}}{\kappa} \ln \left(\frac{10}{1} \right) \right], \quad (6)$$

where U_1 is the wind speed measured 1 m above the water surface (m s^{-1}), C_{d10} is a mean drag coefficient 10 m above the water's surface (dimensionless), and κ is the von Karman constant (0.41; dimensionless) (Crusius and Wanninkhof 2003). U_{10} was calculated using the C_{d10} value determined for "frictionless" lakes (1.3×10^{-3}) (Crusius and Wanninkhof 2003). Wind speed was measured 1 m above the water's surface at the time of sampling each site using a Kestrel 2000 Wind Meter (Nielsen-Kellerman, Boothwyn, PA).

Literature Synthesis

Web of Science and Google Scholar were searched for published studies pertaining to ebullition in hydropower reservoirs and in temperate fresh waters. All reservoirs included are power-producing. Studies presenting results in bubble volume per time, mass per time, combined ebullition and diffusion, or any other values that could not be converted to ebullitive CH_4 flux in units of $\text{mg CH}_4 \text{m}^{-2} \text{d}^{-1}$ were excluded. Mean daily ebullitive CH_4 fluxes reported by each study were grouped as follows for statistical and graphical comparison: Temperate rivers and streams, temperate lakes, temperate hydropower reservoirs, boreal hydropower reservoirs, and tropical hydropower reservoirs. Mean daily ebullitive CH_4 fluxes reported by reservoir studies were also related to latitude. Where mean (i.e., replicated) daily ebullitive CH_4 fluxes exist in a study for the same water body at different sampling sites or times, these mean values were included in comparisons as separate data points.

Statistics

Statistical comparisons between CH_4 ebullition and diffusion at different study locations were made with Wilcoxon–Mann–Whitney tests ($\alpha = 0.05$). Wilcoxon–Mann–Whitney tests were also used to compare mean daily CH_4 ebullition from temperate rivers, temperate lakes, temperate reservoirs, tropical reservoirs, and boreal reservoirs in the literature synthesis. Spearman's rho analyses (ρ ; $\alpha = 0.05$) were used to determine whether CH_4 ebullition was correlated to water temperature, dissolved oxygen, DOC, or SOC. Spearman's rho analysis was also used to determine whether mean daily CH_4 ebullition was correlated to latitude in the literature synthesis. When scaling CH_4 ebullition and diffusion to reservoir surface areas, uncertainties were propagated following Taylor (1997).

Results

Water Quality

Data gathered on reservoir characteristics are summarized in Table 1. Both reservoirs and the Hanford Reach were oxic and ranged from mesotrophic to eutrophic (Dodds et al 1998). Dissolved oxygen in the depositional littoral zone varied on a diel basis from 8 mg L^{-1} to 12 mg L^{-1} , consistent with alternately high rates of primary production during the day and respiration at night (Fig. 3). Nutrient concentrations ranged from 0 to $0.47 \text{ mg phosphate (PO}_4^{3-}) \text{ L}^{-1}$ and from 0.6 to $1.9 \text{ mg nitrate (NO}_3^-) \text{ L}^{-1}$ along the Hanford Reach, 0.02 to $1.64 \text{ mg PO}_4^{3-} \text{ L}^{-1}$ and

Table 1 Reservoir characteristics and mean water temperature, dissolved organic carbon (DOC), and percent sediment organic carbon (SOC) (\pm standard error, SE) measured by this study

Study location	Year constructed	Latitude (degrees)	Reservoir surface area (km^2)	Water residence time (d)	Mean annual discharge ($\text{m}^3 \text{s}^{-1}$)	Mean water temperature ($^\circ\text{C}$)	Mean DOC concentration (mg L^{-1})	Mean SOC (%)
Priest Rapids Columbia River	1961	46.6431876	31.3	0.8	3115	18.7 ± 0.3	1.5 ± 0.2	6 ± 4
Lower Monumental Snake River	1969	46.5623627	26.7	6.0	1410	18.6 ± 0.3	1.7 ± 0.1	3.1 ± 0.5
Hanford Reach Columbia River						18.7 ± 0.6	1.25 ± 0.05	4 ± 2

Surface area, residence time, and discharge values were gathered from the Oak Ridge National Laboratory National Hydropower Asset Assessment Project Database (2013) and the U.S. Geological Survey National Water Information System (2013; <http://nwis.waterdata.usgs.gov/usa/nwis/>), and the Columbia River Data Access in Real Time (2013; www.cbr.washington.edu/dart/river.html)

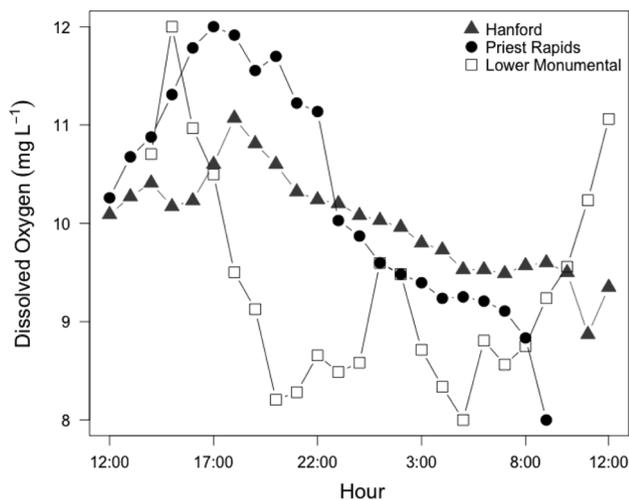


Fig. 3 Diel dissolved oxygen (mg L^{-1}) measured in the depositional littoral zone of Priest Rapids Reservoir, Lower Monumental Reservoir, and along the Hanford Reach

0.8 to $5.5 \text{ mg NO}_3^- \text{ L}^{-1}$ in the Priest Rapids Dam reservoir, and 0.28 to $2.75 \text{ mg PO}_4^{3-} \text{ L}^{-1}$ and 0.1 to $4.3 \text{ mg NO}_3^- \text{ L}^{-1}$ in the Lower Monumental Dam reservoir. Water temperatures ranged from 17 to 20°C .

During the approximately 24 h that ebullition was measured in Priest Rapids Reservoir, water elevation initially increased by 10 cm, then decreased by 10 cm. Water elevation increased 6 cm while inverted funnels were deployed at Lower Monumental. Change in water elevation was most dramatic along the Hanford Reach, decreasing 34 cm during the measurement period. Barometric pressure increased incrementally from 747 mm Hg on September 1 to 753 mm Hg on September 7 during our sampling campaign, which was slightly higher than the 2012 mean barometric pressure (748.5 mm Hg; CBR 2013).

Ebullition

CH_4 ebullition was not correlated to water temperature, dissolved oxygen, DOC, or SOC in Lower Monumental Reservoir, Priest Rapids Reservoir, or along the Hanford Reach. CH_4 ebullition varied widely in each reservoir. In Lower Monumental reservoir, CH_4 ebullition ranged from 7.5 to $984.3 \text{ mg m}^{-2} \text{ d}^{-1}$ (mean \pm standard error, or standard deviation/square root n ; $400 \pm 200 \text{ mg m}^{-2} \text{ d}^{-1}$) ($n = 4$). In Priest Rapids reservoir, CH_4 ebullition ranged from 179.4 to $744.1 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$ ($400 \pm 100 \text{ mg m}^{-2} \text{ d}^{-1}$). By contrast, CH_4 ebullition from the Hanford Reach ranged from 0.3 to $2.4 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$ ($1.1 \pm 0.5 \text{ mg m}^{-2} \text{ d}^{-1}$) ($n = 4$). Thus, ebullitive CH_4 fluxes were significantly greater from both reservoirs than along the Hanford Reach ($p = 0.01$) where change in water elevation was greatest. If CH_4 ebullition was simply a function of decreasing water

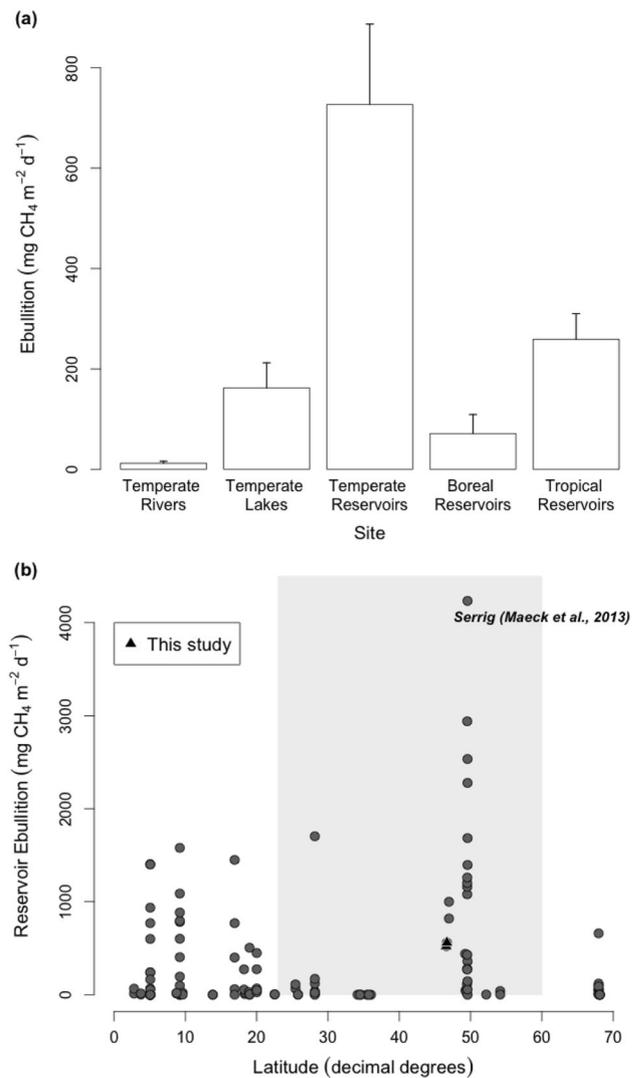


Fig. 4 **a** Mean daily ebullitive CH_4 fluxes compiled from the literature for temperate rivers and streams ($n = 7$), temperate lakes ($n = 32$), temperate hydropower reservoirs ($n = 38$), boreal hydropower reservoirs ($n = 17$), and tropical hydropower reservoirs ($n = 66$). **b** Temperate hydropower reservoirs, shaded in gray, have significantly greater mean daily ebullitive CH_4 fluxes than tropical or boreal hydropower reservoirs

elevation and hydrostatic pressure, greater ebullitive CH_4 fluxes would have been observed along the Hanford Reach. Ebullitive CH_4 fluxes measured in eastern Washington were within the range reported by other studies of temperate hydropower reservoirs (Fig. 4b; Table 2). According to the literature synthesis, mean daily ebullitive CH_4 fluxes from temperate hydropower reservoirs ($700 \pm 200 \text{ mg m}^{-2} \text{ d}^{-1}$) ($n = 38$) tend to be greater than mean daily ebullitive fluxes from temperate rivers and streams ($12 \pm 4 \text{ mg m}^{-2} \text{ d}^{-1}$; $p = 0.02$) ($n = 7$), temperate lakes ($160 \pm 50 \text{ mg m}^{-2} \text{ d}^{-1}$; $p = 0.04$) ($n = 32$), boreal hydropower reservoirs ($70 \pm 40 \text{ mg m}^{-2} \text{ d}^{-1}$; $p = 0.01$) ($n = 17$), and tropical hydropower reservoirs ($260 \pm 50 \text{ mg m}^{-2} \text{ d}^{-1}$; $p = 0.01$) (n

Table 2 Study methods, locations, and mean daily CH₄ ebullitive flux measurements compiled from the literature

Category	Authors	Method	Study	Location	CH ₄ Ebullition (mg m ⁻² d ⁻¹)
Temperate hydropower reservoirs	Del Sontro et al. 2010	Bubble trap	Switzerland	Wohlen	1000.0
		Optical detector	Australia	Little Nerang-1	1704.1
	Little Nerang-2			37.1	
	Little Nerang-3			9.1	
	Little Nerang-4			120.8	
	Little Nerang-5			0.3	
	Little Nerang-6			170.9	
	Little Nerang-7			22.0	
	Maeck et al. 2013	Echosounder	Germany	Saarbruecken-8	439.5
				Lisdorf	48.1
				Rehlingen	32.1
				Mettlach ^a	113.9
				Serrig-1	0.0
				Serrig-2	1684.2
				Serrig-3	2534.3
				Serrig-4	4234.6
				Serrig-5	2277.7
				Serrig-6	1395.5
	Maeck et al. 2014	Inverted funnel	Germany	Serrig-1	359.3
				Serrig-2	56.1
				Serrig-3	146.0
	Del Sontro et al. 2015	Echosounder	Switzerland	Wohlen	820.0
				Wilkinson et al. 2015	Germany
	Serrig-2	2940.0			
	Serrig-3	280.0			
	Serrig-4	1160.0			
	Serrig-5	430.0			
Serrig-6	1200.0				
Mettlach-1	270.0				
Mettlach-2	1260.0				
Bevelhimer et al. 2016	Inverted funnel	USA	Allatoona	0.3	
			Douglas	0.0	
			Fontana	0.0	
			Guntersville	0.4	
			Hartwell	0.0	
			Watts Bar	0.0	
			This study	Inverted funnel	USA
Priest Rapids	559.9				
Temperate lakes	Strayer and Tiedje 1978	Inverted funnel	USA	Wintergreen	337.0
				Mattson and Likens 1990	Inverted funnel
	Mirror-2	68.0			
	Casper et al. 2000	Inverted funnel	UK	Priest Pot	199.0
				Bastviken et al. 2004	Floating chamber ^a
	Crampton	3.1			
	East Long	9.7			
	Hummingbird	4.2			

Table 2 continued

Category	Authors	Method	Study	Location	CH ₄ Ebullition (mg m ⁻² d ⁻¹)
				Morris	59.6
				North Gate	3.3
				Paul	9.8
				Peter	16.3
				Roach	0.3
				Tuesday	4.3
				Ward	36.5
				Ontario	2.0
	Martinez and Anderson 2013	Echosounder	USA	Elsinore-1	1539.8
				Elsinore-2	256.6
				Elsinore-3	449.1
				Elsinore-4	208.5
				Elsinore-5	288.7
				Elsinore-6	208.5
				Elsinore-7	352.9
				Elsinore-8	433.1
Category	Authors	Method	Study	Location	CH ₄ Ebullition (mg m ¹² d ⁻¹)
Temperate Lakes	Martinez and Anderson 2013	Echosounder	USA	Elsinore-9	80.2
				Elsinore-10	128.3
				Elsinore-11	64.2
				Elsinore-12	112.3
				Elsinore-13	64.2
				Elsinore-14	160.4
	Bartosiewicz et al. 2015	Inverted funnel	Canada	Jacques-1	66.0
				Jacques-2	10.0
Temperate Rivers	Wilcock and Sorell 2008	Floating chamber ^a	New Zealand	Whakapipi	2.0
				Toenipi-1	15.0
				Toenipi-2	10.0
				Toenipi-3	18.0
	Crawford et al. 2014	Inverted funnel	USA	Allequash-1	9.6
				Allequash-2	30.5
	<i>This study</i>		USA	Hanford	1.4
Boreal Hydropower Reservoirs	Huttunen et al. 2002	Inverted funnel	Finland	Lokka-1	58.0
				Lokka-2	47.0
				Lokka-3	101.0
				Lokka-4	55.0
				Lokka-5	660.0
				Lokka-6	16.0
				Lokka-7	2.6
				Lokka-8	120.0
				Lokka-9	4.4
				Lokka-10	89.0
				Porttipahta-1	0.0
				Porttipahta-2	0.7

Table 2 continued

Category	Authors	Method	Study	Location	CH ₄ Ebullition (mg m ¹² d ⁻¹)
Tropical hydropower reservoirs	Duchemin et al. 2006a, b	Bubble trap	Canada	Porttipahta-3	2.1
				Porttipahta-4	0.2
				Robert Bourassa-1	40.0
				Robert Bourassa-2	1.8
	Teodoru et al. 2012	Inverted funnel	Canada	Eastmain	2.0
	Keller and Stallard 1994	Inverted funnel	Panama	Gatun-1	17.0
				Gatun-2	404.0
				Gatun-3	783.0
				Gatun-4	195.0
				Gatun-5	795.0
				Gatun-6	98.0
				Gatun-7	602.0
				Gatun-8	1579.0
				Gatun-9	0.0
				Gatun-10	0.0
	Galy-Lacaux et al. 1997	Inverted funnel	French Guiana	Petit Saut-1	1404.0
				Petit Saut-2	936.0
				Petit Saut-3	0.0
				Petit Saut-4	0.0
				Petit Saut-5	0.0
				Petit Saut-6	240.0
				Petit Saut-7	600.0
				Petit Saut-8	0.0
				Petit Saut-9	0.0
				Petit Saut-10	0.0
	Galy-Lacaux et al. 1999	Inverted funnel	French Guiana	Petit Saut-1	1400.0
				Petit Saut-2	770.0
Petit Saut-3				240.0	
Petit Saut-4				0.0	
Petit Saut-5				164.0	
Petit Saut-6				66.0	
Petit Saut-7				55.0	
Petit Saut-8				0.0	
Duchemin et al. 2000	Inverted funnel	Brazil	Curua Una-1	12.0	
			Curua Una-2	65.0	
Joyce and Jewell 2003	Inverted funnel	Puerto Rico	Loiza-1	8.0	
			Loiza-2	24.0	
		Panama	Gatun-1	5.0	
			Gatun-2	1088.0	
			Gatun-3	884.0	
		Category	Authors	Method	Study
Tropical hydropower reservoirs	Tremblay 2005	Inverted funnel	Brazil	Miranda	18.5
				Tres Marias	55.9

Table 2 continued

Category	Authors	Method	Study	Location	CH ₄ Ebullition (mg m ⁻² d ⁻¹)
				Barra Bonita	3.1
				Segredo	1.9
				Xingo	19.6
				Samuel	13.6
				Tucurui	2.5
				Itaipu	0.6
				Serra da Mesa	66.3
	dos Santos et al. 2006 ^b	Inverted funnel	Brazil	Miranda	29.2
				Tres Marias	273.1
				Barra Bonita	4.8
				Segredo	1.7
				Xingo	1.9
				Samuel	19.3
				Tucurui	13.2
				Itaipu	0.5
				Serra da Mesa	111.0
	Bergier et al. 2011	Floating chamber ^a	Brazil	Corumba-1	0.0
				Corumba-2	4.0
				Corumba-3	505.0
	Del Sontro et al. 2011	Echosounder	Zambia	Lufua	1450.0
				Gache Gache-1	770.0
				Gache Gache-2	60.0
				Charara-1	400.0
				Charara-2	1.0
	Deshmukh et al. 2014	Inverted funnel	Laos	Nam Thuen-1	26.0
				Nam Thuen-2	67.0
				Nam Thuen-3	449.0
				Nam Thuen-4	274.0
				Nam Thuen-5	48.0

When fluxes existed for a water body at multiple sampling sites or times, the fluxes are listed as separate values

^a Distribution and variance in gas transfer velocities used to calculate separate ebullitive and diffusive CH₄ fluxes following Bastviken et al. (2004)

^b Values also reported in Rosa et al. (2003)

= 66) (Fig. 4a). Mean daily ebullitive CH₄ fluxes were therefore not correlated to a monotonic increase or decrease in latitude (Fig. 4b). This is consistent with a recent synthesis of total CH₄ emissions (i.e., combined diffusion and ebullition) from hydropower by Deemer et al. (2016), though not a synthesis by Barros et al. (2011), who showed a negative correlation between CH₄ diffusion and latitude.

Diffusion

Dissolved CH₄ was oversaturated relative to atmospheric concentrations at most study locations (Table 3). CH₄ diffusion was significantly lower than CH₄ ebullition across

study locations ($p < 0.001$). Mean diffusive CH₄ flux was 0.4 (± 0.2) mg CH₄ m⁻² d⁻¹ from Lower Monumental reservoir, 0.9 (± 0.6) mg CH₄ m⁻² d⁻¹ from Priest Rapids reservoir, and 3 (± 1) mg CH₄ m⁻² d⁻¹ along the Hanford Reach. CH₄ diffusion along the free-flowing Hanford Reach was significantly greater than CH₄ diffusion in either reservoir ($p = 0.04$). Dissolved CH₄ concentrations and diffusive CH₄ fluxes did not vary significantly across different reservoir environments, such as the main channel, depositional littoral zone, and forebay.

When these diffusive fluxes are scaled to the total surface areas of Lower Monumental (26.7 km²) and Priest Rapids (31.3 km²) reservoirs, they amount to 10 (± 6) kg CH₄ d⁻¹

Table 3 Dissolved CH₄ concentrations, windspeed at 10 m above the water's surface with a drag coefficient (U_{10} ; see Section 2.4), gas transfer velocity with a Schmidt number of 600 (k_{600}), and daily CH₄ diffusive flux measurements in Priest Rapids and Lower Monumental Reservoirs and along the Hanford reach

Study location	Dissolved CH ₄ concentration (μmol L ⁻¹)	U_{10} (m s ⁻¹)	k_{600} (cm h ⁻¹)	Diffusive CH ₄ flux (mg m ⁻² d ⁻¹)
Priest Rapids				
Forebay 1	0.00	0.00	2.07	-0.03
Forebay 2	0.64	0.00	2.07	4.60
Mainstem 1	0.04	1.70	2.60	0.29
Mainstem 2	0.01	3.55	3.92	0.15
Littoral 1	0.00	1.24	2.38	-0.03
Littoral 2	0.13	2.64	3.19	1.44
Tributary 1	0.00	1.76	2.63	-0.04
Lower Monumental				
Forebay 1	0.01	0.79	2.21	0.08
Forebay 2	0.03	0.03	2.07	0.20
Mainstem 1	0.05	1.97	2.75	0.42
Mainstem 2	0.01	0.00	2.07	0.04
Littoral 1	0.01	1.52	2.51	0.12
Littoral 2	0.00	2.37	3.00	-0.02
Tributary 1	0.24	0.00	2.07	1.76
Tributary 2	0.12	0.00	2.07	0.77
Hanford Reach				
Mainstem 1	0.01	4.07	4.40	0.16
Mainstem 2	0.14	4.07	4.40	2.15
Littoral 1	0.18	2.12	2.84	1.85
Littoral 2	0.84	0.91	2.25	6.72

and 30 (± 20) kg CH₄ d⁻¹, respectively. Ebullitive CH₄ fluxes from Lower Monumental and Priest Rapids scaled to the surface area of the depositional littoral zones in these reservoirs amount to 900 (± 500) kg CH₄ d⁻¹ and 1000 (± 300) kg CH₄ d⁻¹, respectively. Consequently, over 97% of daily CH₄ emissions are shown here to originate from just 22% of the surface area in Lower Monumental reservoir and 6% of surface area in Priest Rapids reservoir.

Discussion

Temperate Hydropower Reservoirs

CH₄ ebullition in Priest Rapids and Lower Monumental reservoirs was 3–4 orders of magnitude higher than CH₄ diffusion in either reservoir, which highlights the potential for misrepresentation of CH₄ fluxes from reservoirs when ebullition is not measured. Despite nearly identical sampling methods, Bevelhimer et al. (2016) measured much lower CH₄ ebullition in six southeastern US hydropower

reservoirs (0.0–15.7 mg m⁻² d⁻¹). Although this regional difference in measurements can be attributed to many factors, including catchment land use, it highlights both the range in CH₄ ebullition possible and need for further study at different spatial and temporal scales. Notably, the scope of this study and others is limited due to sampling during the daytime and temperate summer, only. Thus, our diffusion results (sampled once during 24 h) cannot be considered representative of other times of the day or of other seasons. Our results are more comparable to Del Sontro et al. (2010, 2015), Maeck et al. (2012, 2013), and Wilkinson et al. (2015) (Table 2). Del Sontro et al. (2010) found that CH₄ ebullition in temperate hydropower reservoir Lake Wohlen was greatest when water quality conditions were similar to the conditions during our sampling period (i.e., oxic with temperatures exceeding 17 °C during the temperate summer). When comparing our results to other studies, it is important to distinguish that our study sampled ebullition in the shallow depositional littoral zone, only. It is widely acknowledged that shallow depths produce greater ebullition fluxes, in part due to lower hydrostatic pressure (Bastviken et al. 2004; Galy-Lacaux et al. 1999; Keller and Stallard 1994; Mattson and Likens 1990). Grinham et al. (2011) found that up to 97% of the total CH₄ flux in their study reservoir was produced by only 1.8–7.0% of the entire surface area, and over 95% of ebullition occurred in depths less than 12 m during the day and 6 m during the night, which supports our sampling strategy.

Tropical and Boreal Hydropower Reservoirs

Wide ranges in ebullitive fluxes from hydropower have been observed in all three climate zones, which are unsurprising given the variations in methodology, sampling design, and temporal stochasticity of ebullition. GHG emissions from tropical reservoirs have long been a concern for hydropower growth (Barros et al. 2011; dos Santos et al. 2006; Fearnside and Pueyo 2012; St Louis et al. 2000). Boreal and temperate hydropower reservoirs have generally been grouped as a less urgent concern, with evidence from boreal hydropower reservoirs supporting low ebullitive fluxes (e.g., Duchemin et al. 2006a; Huttunen et al. 2002). Measurements of CH₄ ebullition in temperate hydropower reservoirs have only occurred over the past 9 years; the discussion has not since shifted to include concerns about high CH₄ emissions from temperate hydropower reservoirs. Ebullition measured by this study in eastern Washington was not higher than ebullition measured in tropical hydropower reservoirs. However, the literature synthesis indicates that temperate hydropower reservoirs as a group have significantly greater mean daily ebullitive CH₄ fluxes than tropical hydropower reservoirs.

Importantly, the literature synthesis compares mean daily ebullitive CH₄ fluxes sampled during summer from temperate and boreal hydropower reservoirs (see Maeck et al. 2013 for an exception) to mean daily ebullitive CH₄ fluxes sampled year round from tropical reservoirs. High CH₄ ebullition in temperate hydropower reservoirs may be related to organic carbon loading and seasonal temperature fluctuations. Temperate and boreal fresh water sediments receive large annual inputs of organic carbon (Aitkenhead and McDowell 2000) during autumn (Filstrup et al. 2009), when temperatures fall and microbial methanogenesis becomes limited (Yvon-Durocher et al. 2014). This may lead to accumulations of antecedent organic carbon until the following summer, when temperate hydropower reservoirs are typically studied, and peak temperatures may cause a pulse of respiration and CH₄ ebullition. In some cases, CH₄ ebullition may also be trapped under ice through the temperate and boreal winter and released suddenly during the spring thaw (Walter et al. 2006). By contrast, year-round inputs of organic carbon to tropical fresh water sediments and its subsequent respiration proceed at relatively constant rates sustained by high temperatures (Castillo et al. 2004; Yvon-Durocher et al. 2012). Thus, while temperate and boreal CH₄ ebullition can be seasonally elevated compared to tropical climates, annual CH₄ emissions due to ebullition are likely to be higher within tropical climates.

Temperate Lakes and Rivers

Mean daily ebullitive CH₄ fluxes from temperate hydropower reservoirs were significantly higher than mean daily ebullitive CH₄ fluxes from temperate rivers and lakes. This emphasizes that hydropower structures alter natural systems by creating environments that can increase sediment and organic carbon deposition behind dams, which may foster CH₄ ebullition in certain conditions (Maeck et al. 2013; Thornton et al. 1990). Reliance on the GHG flux literature from temperate lakes to supplement the paucity of information on temperate hydropower reservoirs (Goldenfum 2012) would therefore be inaccurate.

Temperate Hydropower Reservoirs without Ebullition Measurements

Several studies have quantified CH₄ fluxes from temperate hydropower reservoirs without the inclusion of ebullition measurements or without the separation of ebullition from total flux measurements. Temperate study locations have included six hydropower reservoirs in the western US (Soumis et al. 2004), the Three Gorges Reservoir in subtropical China (Chen et al. 2011; Zhao et al. 2013), Douglas Lake, a hydropower reservoir in Tennessee, US (Mosher et al. 2015), and sub-tropical Little Nerang Reservoir in

Queensland, Australia (Grinham et al. 2011). The studies that do not account for ebullition identified CH₄ emissions ranging from 0 to 9.5 mg CH₄ m⁻² d⁻¹ (Chen et al. 2011; Mosher et al. 2015; Soumis et al. 2004; Zhao et al. 2013). Such estimates may be misleadingly small when considering the high CH₄ emissions seen in temperate hydropower reservoirs that have reported ebullition values, including this study. Furthermore, tropical hydropower reservoirs have been shown to emit more than 85% of CH₄ through ebullition from a small fraction of their surface area (Del Sontro et al. 2010; dos Santos et al. 2006; Maeck et al. 2013; Ramos et al. 2006). Similarly, Grinham et al. (2011) included results from an optical detector that targeted a distinct CH₄ ebullition flux (Table 2) and results from static chambers that captured both diffusive and ebullitive fluxes. The static chambers used by Grinham et al. (2011) were dominated by ebullition fluxes in their most-shallow sites and commonly measured CH₄ fluxes greater than 2000 mg CH₄ m⁻² d⁻¹, with a maximum flux of 22,000 ± 4000 mg CH₄ m⁻² d⁻¹, which is one of the highest CH₄ fluxes recorded in the literature.

Importance to the US

With the continued growth and expansion of hydropower in the US and the recent consensus adoption of the Paris Agreement under the UNFCCC, it is clear that hydropower will continue to be a valuable and important renewable energy source in the US. The emphasis for hydropower growth is currently more focused on existing structures, which is less likely to increase GHG emissions than the creation of new dams and their accompanying reservoirs (Barros et al. 2011; Uría-Martínez et al. 2015). The US Environmental Protection Agency (EPA) compiles the national inventory of GHGs submitted to the UNFCCC, which requires that its member countries follow the IPCC 2006 Guidelines for National Greenhouse Gas Inventories in its report preparation (UNFCCC 2014; USEPA 2015). The IPCC 2006 guidelines do not yet require the inclusion of GHG sources or sinks associated with hydropower, but they contain an appendix entitled, “CH₄ Emissions from Flooded Land: Basis for Future Methodological Development” (Duchemin et al. 2006b). Hydropower reservoirs are included in the characterization of “flooded land,” and thus the appendix methodologies can be applied to hydropower reservoir emissions. The exclusion of flooded land emissions from national inventories is attributed to a lack of comprehensive information (Duchemin et al. 2006b). With increasingly comprehensive information available, GHG emissions from hydropower may be required in future emissions inventories for member countries of the UNFCCC. The methodology suggested by the IPCC requires the inclusion of CH₄ ebullition for emissions calculations if (1) data are available or (2) if flood lands are estimated to have “a significant influence on the country’s total inventory of

greenhouse gases in terms of the absolute level, the trend, or the uncertainty in emissions and removals” (Duchemin et al. 2006b; Herold et al. 2006). Therefore, the US may soon be required to include ebullitive CH₄ emissions information from its hydropower reservoirs. Few measurements and the lack of a standardized protocol in the US makes this difficult, particularly in non-power producing reservoirs, for which there are even fewer data available (see Beaulieu et al. 2014 for an exception).

Conclusion

Although understudied, temperate hydropower reservoirs can act as an important source of GHG emissions from fresh waters through CH₄ ebullition. CH₄ ebullition in this study’s eastern Washington reservoirs was up to three orders of magnitude higher than ebullition in southeastern US hydropower reservoirs (Bevelhimer et al. 2016), but is similar to other studies of temperate hydropower reservoirs (Fig. 4b). This emphasizes the possible range of CH₄ emissions through ebullition and the need for more widespread scrutiny of hydropower emissions from temperate climates (Fearnside and Pueyo 2012). The path forward for the US is still unclear, though understanding the causes and accurately quantifying CH₄ emissions from hydropower reservoirs will help the country better understand its GHG emissions and comply with future guidelines by the IPCC and UNFCCC. The impacts of fully characterizing GHG emissions from hydropower are potentially far-reaching and may raise new policy questions concerning carbon offset and cap-and-trade programs, engineering for new hydropower projects, and future renewable energy balances.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no competing interests.

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