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Reservoir Water-Level Drawdowns Accelerate and Amplify Methane Emission

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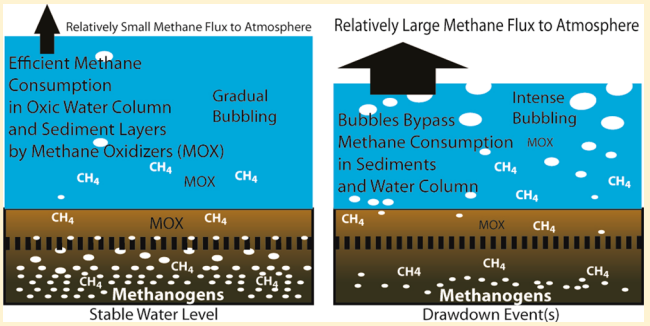
Supporting Information

ABSTRACT: Water-level fluctuations due to reservoir management could substantially affect the timing and magnitude of reservoir methane (CH_4) fluxes to the atmosphere. However, effects of such fluctuations on CH_4 emissions have received limited attention. Here we examine CH_4 emission dynamics in six Pacific Northwest U.S. reservoirs of varying trophic status, morphometry, and management regimes. In these systems, we show that water-level drawdowns can, at least temporarily, greatly increase per-area reservoir CH_4 fluxes to the atmosphere, and can account for more than 90% of annual reservoir CH_4 flux in a period of just a few weeks. Reservoirs with higher epilimnetic [chlorophyll *a*] experienced larger increases in CH_4 emission in response to drawdown (R^2 = 0.84, p < 0.01), suggesting that eutrophication magnifies the effect of drawdown on CH_4 emission. We show that drawdowns as small as 0.5 m can stimulate ebullition events. Given that drawdown events of this magnitude are quite common in reservoirs, our results suggest that this process must be considered in sampling strategies designed to characterize total CH_4 fluxes from reservoirs. The extent to which (and the mechanisms by which) drawdowns short-circuit connections between methanogenesis and methanotrophy, thereby increasing net CH_4 fluxes to the atmosphere, should be a focus of future work.



INTRODUCTION

Collectively, at the global scale, reservoirs constitute a major anthropogenic source of atmospheric CH_4, emitting 12−70 Tg y^{-1} of CH_4, roughly as much as other major anthropogenic CH_4 sources such as rice cultivation or biomass burning.[1−4] In lakes and reservoirs, CH_4 is produced primarily in anoxic sediments and can be released to overlying water via diffusion, ebullition (bubbling),[5] or, when emergent vegetation is present, via plant-mediated transport. Emissions of CH_4 from reservoirs can also occur via degassing at turbines and spillways.[3] The pathway for CH_4 emission (diffusion vs ebullition) matters because the fate of CH_4 diffusing out of sediments is quite different from that of CH_4 bubbling out of sediments. CH_4 diffusing out of sediments is generally subject to remarkably efficient microbial CH_4 oxidation (methanotrophy), a process which converts CH_4 to CO_2, a much less potent greenhouse gas on a per-molecule basis.[6] In fact, a recent review of nine lakes with both CH_4 production and oxidation measurements conservatively estimated that methanotrophs consume 50−95% (median: 90%) of all CH_4 produced in lakes and an even greater fraction of the nonebullitive CH_4 flux.[7] In contrast, CH_4 bubbling out of sediments largely bypasses methanotrophs, especially in shallow systems/sites where water column bubble dissolution is limited.[8] Hence, processes that increase the fraction of CH_4 released via ebullition (versus diffusion) can increase the magnitude of atmospheric CH_4 flux. Furthermore, although ebullition is frequently the dominant emission pathway for CH_4 emission from reservoirs, it is infrequently measured relative to diffusive emissions.[3] Hence there is a great need to better quantify and understand the drivers of CH_4 ebullition.

Methane is produced in anoxic lake sediments via microbial decomposition of organic matter, and production rates depend on organic carbon availability, redox conditions, and temperature.[9,10] If the partial pressure of all dissolved gases in porewater exceeds ambient pressure and water surface tension, free gas is formed.[11−13] Continuous CH_4 production causes bubbles to form, grow and create fractures or disc-shaped cavities within sediments.[13,14] With continuing gas production in sediments, these gas bubbles can grow further, coalesce, and migrate vertically through the sediment until they are released into the water column and, subsequently, to the atmosphere.[11,13] Decreases in hydrostatic pressure (or other physical disturbances) can lower compressive sediment stress and cause bubbles to overcome their confinement, expand by deforming

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Table 1. Characteristics of Study Reservoirs

reservoir name	surface area (ha)	max depth (m)	drawdown description	magnitude of drawdown (m)	average pace of drawdown (m d^{-1})	average surface[Chl a] (mg m^{-3})	trophic status[a]
Cle Elum	1940	101	large summer drawdown	25	0.45	0.24	oligotrophic[36]
Kachess	1837	125	large late-summer drawdown	7	0.12	0.40	oligotrophic
Foster	425	33	fall drawdown + hydropower reregulation	6	0.30	1.35	mesotrophic[37]
Lacamas	127	17	short late-summer drawdown	2	0.14	5.38	eutrophic[38]
J. C. Boyle	154	14	hydropower peaking	0.5	0.66	6.27	eutrophic[39]
Keno	634	10	constant water level	0	na	12.03	eutrophic[39]

[a]Trophic status was taken as reported by system-specific studies when available (citations included in table) or, otherwise, based on U.S. EPA National Lakes Assessment chlorophyll a criteria and data from this study.[61]

the surrounding sediments, and hasten their rise to the surface.[11] The cohesive strength of sediments, and hence their sensitivity to changes in hydrostatic pressure, is thought to vary as a function of organic content and pore pressure, with greater organic matter content generally associated with greater sediment cohesive strength.[15]

On their way upward through sediments (and the water column), bubbles are subject to diffusive exchange with the surrounding environment. There is a widely reported positive relationship between ebullition rates and bubble CH_4 concentrations, which is generally attributed to N_2 stripping by CH_4 bubbles.[16,17] The idea here is that bubbles moving through sediments can strip out N_2 (the most abundant of the dissolved gases at atmospheric equilibrium) from pore waters, leaving CH_4 to make up a large fraction of the partial pressure in remaining bubbles. Although the flattened bubbles thought to occur in cohesive sediments[12] are likely to exhibit more efficient diffusive exchange than spherical bubbles would, larger bubbles generally have lower surface area:volume ratios and therefore result in less efficient gas exchange. In addition, longer residence times for bubbles in sediments would result in lower emission efficiencies for CH_4 produced in sediments as greater time in sediments provides greater opportunity for methanotrophy to occur.

Prior studies of factors controlling ebullition events from aquatic sediments have highlighted many potential drivers, including wind events and associated bottom shear,[15,18] variations in atmospheric pressure,[19−21] oxygen concentrations, water temperature, organic matter input,[22−24] and seasonal[22,25] and tidal[26] decreases in water level. The trophic status of reservoirs also should influence sediment CH_4 production and atmospheric emissions.[27] The theory here is that higher nutrient loads support higher rates of primary production, which in turn provide the organic carbon substrate and favorable (hypoxic) conditions necessary to support rapid rates of CH_4 production. Trophic status also influences the availability of autochthonous carbon, which has been found to promote higher rates of methanogenesis than allocthonous carbon.[27] Recent studies of single systems, as well as regional and global syntheses, suggest an important link between primary production and CH_4 emission in lakes and reservoirs,[3,5,28−30] with more nutrient enriched systems generally exhibiting higher rates of CH_4 emission.

Although fluctuations in pressure (either barometric or hydrostatic) can clearly influence the timing of methane release from sediments,[19,31,32] and researchers have argued that reservoir drawdowns should affect CH_4 ebullition,[11,33,34] there have been very few studies of this phenomenon in reservoirs. In fact, an extensive literature search yielded only two studies investigating the relationship between reservoir water level and ebullition, both of which focused on a single reservoir with a small (<0.5 m in both cases) range of stage height fluctuation. The first study reported synchronous bubble releases across multiple sites within a single, slightly regulated kettle-hole lake that were linked equally to variations in atmospheric pressure and reservoir water level via time series analysis.[22] This study measured cumulative bubble volume, but CH_4 concentrations in bubble gases were not measured, so CH_4 fluxes could not be directly computed. The second, more recent study reported pulsed releases of CH_4-rich bubbles from sediments in a regulated portion of the Saar river (Germany), which correlated with navigation-associated water level fluctuations (total range in water level <0.3 m).[35] Neither study examined the potential interactive effects of water level drawdown and trophic status on atmospheric CH_4 emissions, but it stands to reason that more eutrophic systems may have larger sediment CH_4 stores to be released during drops in hydrostatic pressure.

Here, we take a multireservoir comparative approach to investigate how reservoir management and characteristics affect CH_4 emissions by examining CH_4 ebullition dynamics in several reservoirs subject to a variety of drawdown types (flood prevention, hydropower peaking, and maintenance-related) and spanning a range of trophic statuses. This allows us to examine how reservoir management and characteristics affect CH_4 emissions. Second, we directly compare two adjacent (in-series) reservoirs that differ primarily with respect to water-level management, allowing us to examine the impact of drawdowns on CH_4 flux magnitude, in addition to timing. Third, three full years of data from one of our study reservoirs allowed us to quantify the contribution of drawdown-associated CH_4 fluxes relative to annual CH_4 emissions, a result with important implications for efforts to quantify CH_4 emissions from reservoirs.

MATERIALS AND METHODS

To examine the relationship between water level drawdown and CH_4 ebullition we monitored water column CH_4 concentrations, CH_4 ebullition fluxes, and variables likely to control CH_4 ebullition in six Pacific Northwest U.S. reservoirs, spanning a range of management regimes, trophic statuses, and morphologies (Table 1, Supporting Information (SI) Figures S1 and S2). Two of the study reservoirs (Cle Elum and Kachess) are oligotrophic systems[36] located high in the Yakima River Basin (~700 m elevation), and are managed primarily as irrigation storage reservoirs, with pronounced summer drawdowns that occur over periods of approximately two months (7 and 25 m drawdowns for Kachess and Cle Elum reservoirs, respectively; Table 1). Foster Reservoir is located in the

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Santiam River drainage, is mesotrophic,[37] and is managed as a hydropower reregulation reservoir, a reservoir designed to reduce the impact of flow disturbance caused by hydropower operations at an upstream dam. During this study, Foster Reservoir experienced a 6 m drawdown over a 20 day period in late autumn. Lacamas Lake is a small (127 ha), relatively shallow (8 m average depth), eutrophic reservoir which is managed primarily for recreation.[38] This reservoir experiences an annual September drawdown of 1.5−2.0 m over a 7−12 day period so that dam owners can perform dam maintenance. Finally, Keno and J. C. Boyle reservoirs are adjacent eutrophic systems,[39] situated in series, with Keno located just upstream of J. C. Boyle. These two eutrophic reservoirs differ primarily with respect to their water level management regime, allowing us to examine the effect of drawdowns on CH_4 emissions in two otherwise similar reservoirs. Keno reservoir is managed to maintain a constant water level all year with no drawdowns, whereas J. C. Boyle experiences a daily ∼0.5 m drawdown in order to provide hydropower during periods of peak energy demand. The measurement period for most of these reservoirs ranged from weeks to months, but in Lacamas Lake CH_4 fluxes were measured during multiple drawdown events spanning five years (Table 2).

Table 2. Number of Sampling Sites and Sampling Dates for Each Study Reservoir

reservoir name	no. of sampling sites	dates of trap deployment and associated CH_4 sampling	period between sampling events
Cle Elum	4	May 14, 2013 to Oct 22, 2013	20−30 days
Kachess	4	Jun 5, 2013 to Sept 24, 2013	27−29 days
Foster	4	Aug 6 to Nov 12, 2013	2−16 days
Lacamas	4−13*	Aug 9, 2011 to Oct 7, 2011, Sept 1, 2012 to Oct 26, 2012, and May 31, 2013 to Jan 7, 2016	<1 day during drawdown) - 55 days (winter); (every 5 min for volume)
J. C. Boyle	4	Aug 12−19, 2013	0.05−2 days (every 5 min for volume)
Keno	4	Aug 13−18, 2013	0.2−1.3 days (every 5 min for volume)

In each reservoir, we estimated ebullition using at least 4 inverted funnel traps (SI Figure S3)[22] deployed for extended periods, hanging 1.5 m below the water surface at profundal, intermediate depth, inlet littoral, and noninlet littoral sites. In Lacamas Lake, 6 and 13 traps were deployed in 2013 and 2014−2015, respectively. Gases were sampled at regular intervals (hours to weeks; Table 2) determined by the rate of bubbling within each reservoir, and concentrations were subsequently measured on a Hewlett-Packard 5890 gas chromatograph equipped with a flame ionization (FID) detector.[38] Ebullition fluxes (mg CH_4 m^{-2} y^{-1}) were calculated as the product of accumulated gas volume and concentration, divided by funnel aperture cross-sectional area, and sampling interval. Control traps injected with 130 mL of 50% CH_4 standard were deployed along with sampling traps in each reservoir to account for potential diffusive loss of gas from traps left out over extended periods. These control traps were identical to sampling traps except that a plexiglass sheet was hung below control trap funnels to prevent bubbles from diluting or enriching the control gas.

These control traps were sampled every time the noncontrol traps were sampled. Measured concentrations of standards held for up to 55 days in control traps demonstrated no detectable loss of CH_4 (SI Figure S4), perhaps due to the high volume of standard (generally >70 mL) relative to the surface area in contact with reservoir waters (3.98 cm^2) and the low solubility of CH_4. It is possible that small volumes of gas held in traps would be subject to greater proportional dissolution and oxidation, but we did not observe this effect for the volumes tested (40−100 mL). Sampling events where gas volume in traps was greater than zero and less than 40 mL accounted for just 2.1% of total gas volume collected during this study. At the deepest point in each reservoir (SI Figure S2), vertical profiles were sampled for dissolved [CH_4], temperature, dissolved oxygen, and [chl *a*]. Dissolved [CH_4] was estimated via headspace equilibration as in Harrison and Matson (2003),[40] with gas concentrations measured on a GC equipped with an FID, as described above for bubble fluxes.[38] Temperature, dissolved O_2, and [Chl *a*] were measured using a Hach DSX Sonde. Chl *a* measurements were cross calibrated with acetone-extracted Chl *a*, measured on a Turner Designs AU Fluorometer. Hypolimnetic CH_4 accumulation for Lacamas Lake, Cle Elum Reservoir, and Kachess Reservoir was estimated from profile data as the change in volume-weighted CH_4 mass over time as in Deemer et al. (2011).[41] Diffusive CH_4 flux was estimated using a standard thin boundary layer model that predicted gas flux (F; for example, mmol CH_4 m^{-2} d^{-1}) according to

$$F = k(C_{sur} - C_{eq})$$

where k is the piston velocity (m d^{-1}) and C_{sur} and C_{eq} are measured surface water concentration and calculated air-equilibrium concentration, respectively, for CH_4.[42] Piston velocity (k) was estimated to vary as a function of surface water temperatures measured at the same time as surface water CH_4 concentrations and mean daily windspeed as in Musenze et al. (2014)[43] and similar to Cole and Caraco (1998)[44] according to

$$k = \left(\frac{S_c}{600}\right)^{-0.5}\left[2.07 + 0.215\left(\frac{\ln\left(\frac{10}{Z_0}\right)}{\ln\left(\frac{Z}{Z_0}\right)}\right)^{1.7} U_z^{1.7}\right]$$

where S_c is the Schmidt number for CH_4 at measured surface water temperature calculated as in Waninkhof et al., (1992),[45] Z_0 is the roughness height (taken as 0.1 m), Z is the height at which wind speed was measured, and U_z is the average daily wind speed during the day when surface water concentration was measured.

Lake-wide ebullition fluxes were estimated by area-weighting fluxes from two zones (profundal and littoral, defined as greater and less than 4 m depth, respectively) in each reservoir, using a minimum of 2 traps in each zone for each reservoir. In Lacamas Lake, diffusive and ebullitive CH_4 fluxes as well as hypolimnetic accumulation were measured biweekly over two and a half years (June 2013 to January 2016) and both prior to and during two additional annual drawdown events (in 2011 and 2012). In addition to funnel trap estimates, we also performed hydroacoustic transects in each reservoir using a 120 Hz, split-beam transducer to develop qualitative insight into temporal and spatial distribution of bubbles[34] (see Supplement for additional detail). These transects were executed before and during drawdown in each study reservoir in 2013.

Sampling density and duration in this study were comparable to those in many recent published studies. For example,

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Kemenes et al. (2007)[46] quantified ebullition over 20 min periods at monthly intervals using 10−14 sites, and Maeck et al. (2014)[35] had funnel traps deployed at three sites over a 6 month duration. Similar to Maeck and colleagues (2014),[35] we deployed gas traps for a much greater period of time than is common (weeks-to-months as opposed to the typical minutes-to-hours) and in a variety of habitat types. Sampling and flux calculations were designed to avoid overemphasis of inlet hotspots. Ebullition traps in each reservoir were intentionally placed so as to maximize both the variability captured and sample representativeness. For example, in each reservoir, we utilized at least one shallow site near the inlet, one shallow outlet site, and two comparatively deep (>4 m depth) sites. From traps with pressure transducers (deployed in Keno, J. C. Boyle, and Lacamas) we collected hundreds of observations of gas volume. We present data from only traps with a minimum of five sampling events apiece, but most traps were sampled much more frequently, with collection events (instances when gas samples were collected from traps and returned to the lab for subsequent concentration measurements) occurring at intervals ranging from 0.05 days to several weeks and total sampling time ranging from 5 days up to several calendar years (Table 2).

RESULTS AND DISCUSSION

Drawdown Affects Timing of CH_4 Emission. Several lines of evidence implicate water level drawdown as a crucial control on timing of CH_4 emissions in our study reservoirs. First, there was a strong temporal correspondence between drawdown events and increases in ebullition rates across all study reservoirs experiencing drawdowns ≥0.5 m (i.e., five of the six study systems). In all reservoirs (and in all years for Lacamas Lake, the one system in our study which was monitored for multiple years) we observed pulses of total ebullition, CH_4 emission via ebullition, CH_4 emission via diffusion, and total CH_4 emission (ebullition plus diffusion) associated with water level drawdown (Figures S5, 1, 2, and 3). In every case, simple average and area-weighted-mean whole-lake ebullition rates at least tripled between predrawdown and drawdown periods, and, in the most extreme case (Lacamas in 2013), increased by more than a factor of 6000 (Figure 1a). Furthermore, there were no instances where ebullition rates decreased in any of our study reservoirs (or even individual traps) during drawdown events.

We also observed increases in average surface water CH_4 concentrations in all study reservoirs during reservoir drawdown ($P < 0.05$ in all cases by one-way ANOVA; SI Figure S6), and increases ($P < 0.1$ in all cases) in average hypolimnetic CH_4 concentrations during drawdown in all but one study reservoir (Foster Reservoir; SI Figure S6), which showed no significant change. In reservoirs where it was possible to calculate dissolved CH_4 mass and dissolved CH_4 accumulation rates during predrawdown and drawdown periods (Lacamas, Kachess, and Cle Elum Reservoirs), we observed increases in hypolimnetic CH_4 mass during drawdowns (SI Figure S7). One explanation for these observed increases is that bubbling events load the water column with CH_4 as gases in bubbles exchange with the water column during ascent. An alternative explanation for observed patterns in hypolimnetic CH_4 loading during drawdowns is that increased bubble densities in sediments enhanced diffusion of sparingly soluble gases from sediments into the water column, as has been suggested could occur by at least one core incubation study.[47] Regardless of mechanism,

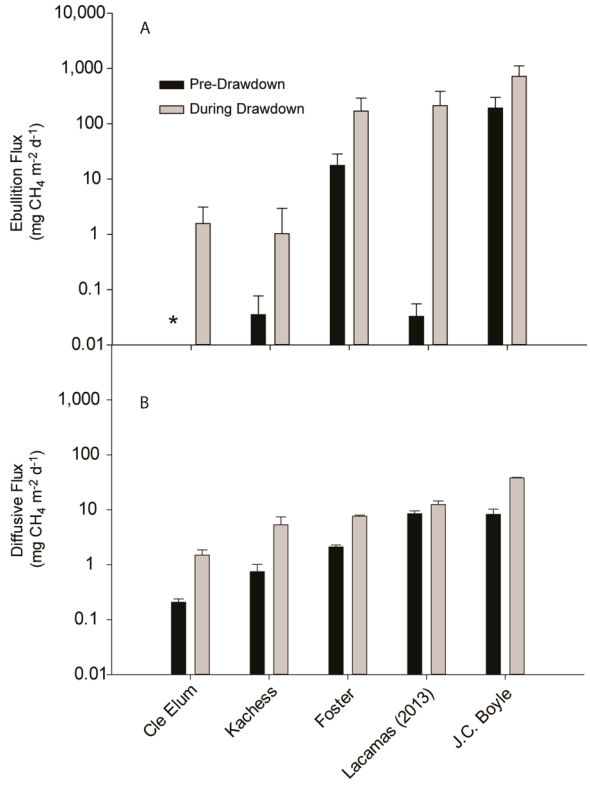

Figure 1. Average CH_4 ebullitive (A) and diffusive (B) fluxes (mg CH_4 m^{-2} day^{-1}) from 5 reservoirs during predrawdown versus during-drawdown conditions. For Panel A, $n = 4$ funnel traps in all cases except Lacamas, where $n = 6$, and error bars represent one standard error of measurements between all traps. For Panel B, $n = 2$−14 time points, depending on the system, and error bars represent one standard deviation of modeled fluxes. * denotes zero flux during the predrawdown period in Cle Elum Reservoir.

hypolimnetic CH_4 accumulation rates in Lacamas increased 250−4,200% during drawdown events, compared to predrawdown accumulation rates during the five years for which we have data (SI Figure S7). Diffusive CH_4 losses to the atmosphere were also elevated during drawdown periods (Figure 1b). Thin boundary layer model-estimated diffusive fluxes were smaller than ebullition fluxes during drawdown periods in all reservoirs, but were larger than ebullition fluxes during nondrawdown periods in oligotrophic reservoirs and in Lacamas Lake (Figure 1a and b). When both ebullition and diffusive fluxes are considered (ignoring hypolimnetic CH_4 accumulation), all reservoirs in this study experienced at least a 3.6-fold increase in CH_4 emissions during drawdown. In all reservoirs, CH_4 accumulation measurements from traps were qualitatively consistent with hydroacoustic data. Bubble densities in the hydroacoustic surveys were much greater following drawdowns than prior to drawdown events, and the highest bubble densities in the hydroacoustic surveys occurred at locations where the greatest gas volumes were collected in traps (SI Methods and Figures S8 and S9).

Ebullition rates varied substantially between traps in each reservoir. During predrawdown periods, coefficients of variation (CVs) for mean rates of CH_4 ebullition between traps ranged from zero in Cle Elum Reservoir, where no ebullition was detected, to 200% in Kachess Reservoir. During drawdown CVs for mean CH_4 ebullition rates ranged from 117% (in J. C. Boyle Reservoir) to 200% (in Kachess Reservoir). A Monte Carlo

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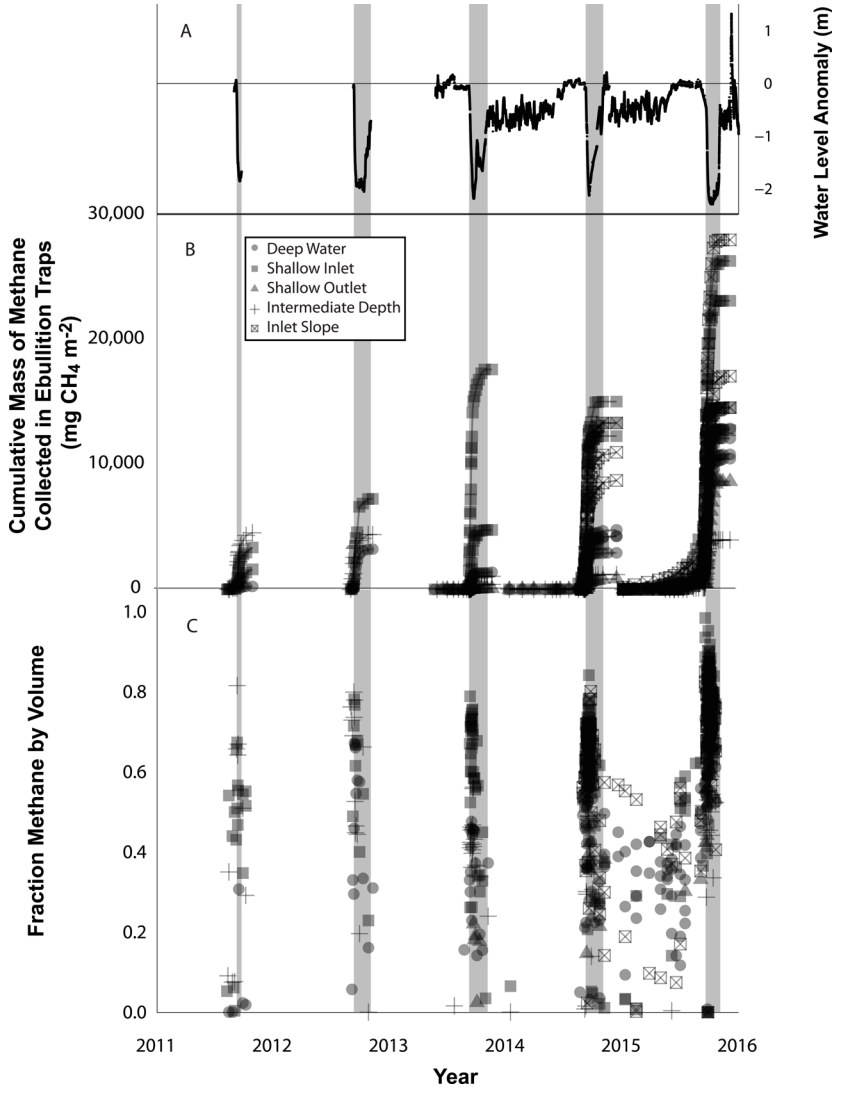

Figure 2. Time series showing (A) change in the water surface elevation anomaly in Lacamas Lake (secondary *Y*-axis and black lines; annual reservoir drawdown events are also indicated by shaded gray bars), (B) cumulative bubbled mass of CH_4 in traps located at four sites in Lacamas Lake in 2011 and 2012, six in 2013, and 13 sites in 2014−2016; lines represent individual bubble traps, and points represent sampling events where traps were emptied and gas concentrations were measured, and (C) the fraction of sampled gas as CH_4 for each time a trap was sampled (n = 717).

subsampling analysis of subsequent additional, higher density sampling in Lacamas Lake (Figure 2) allowed us to determine that sampling with two shallow and two deep ebullition traps (the design implemented in most reservoirs in this study) yields rate estimates within ±60% of rates calculated using 13 traps >95% of the time. Hence, it appears that the large signal due to drawdown made it possible to detect a significant drawdown effect in Lacamas Lake despite substantial spatial variation between traps. If spatial variability of ebullition rates in other study reservoirs is similar to (or less than) that observed in Lacamas Lake, then the sampling density employed in this study was sufficient to reliably detect the large and consistent observed drawdown effect, which was greater than 3-fold in all study reservoirs.

Drawdowns and Magnitude of Ebullition Events. A comparison of two reservoirs differing primarily in water level management suggests that drawdowns affect not just the timing but also the *magnitude* of CH_4 ebullition (J. C. Boyle vs Keno; Figure 3). These reservoirs occur in series, within 9 river km of each other and experience very similar levels of nutrient loading.[39] In J. C. Boyle reservoir, daily drawdowns timed to meet hydropower demand were associated with pulsed release of CH_4-rich bubbles from sediments. In contrast, over the same period, just upstream in Keno reservoir (which is managed to maintain a constant water level), no such pulsed bubbling events were observed. Instead, where ebullition was measured in Keno, rates were fairly constant over the duration of our experiment (Figure 3). The net effect of drawdowns over the period of measurement was to increase total CH_4 ebullition by a factor of 3.5 in J. C. Boyle relative to Keno (Figure 3). During the experimental period bubble CH_4 concentrations averaged 54% in Keno Reservoir and 74% in JC Boyle reservoir, so differences in CH_4 concentration of bubbles only accounted for a small portion of the total difference in ebullition between the two reservoirs. The remainder of the difference in observed ebullition between the two reservoirs was due to differences in bubbling rates, which averaged 341 mL $m^{-2}d^{-1}$ in Keno and 445 and 1624 mL m^{-2} d^{-1} during nondrawdown and drawdown periods in JC Boyle, respectively. It is conceivable that differences in short-term whole-reservoir ebullition rates

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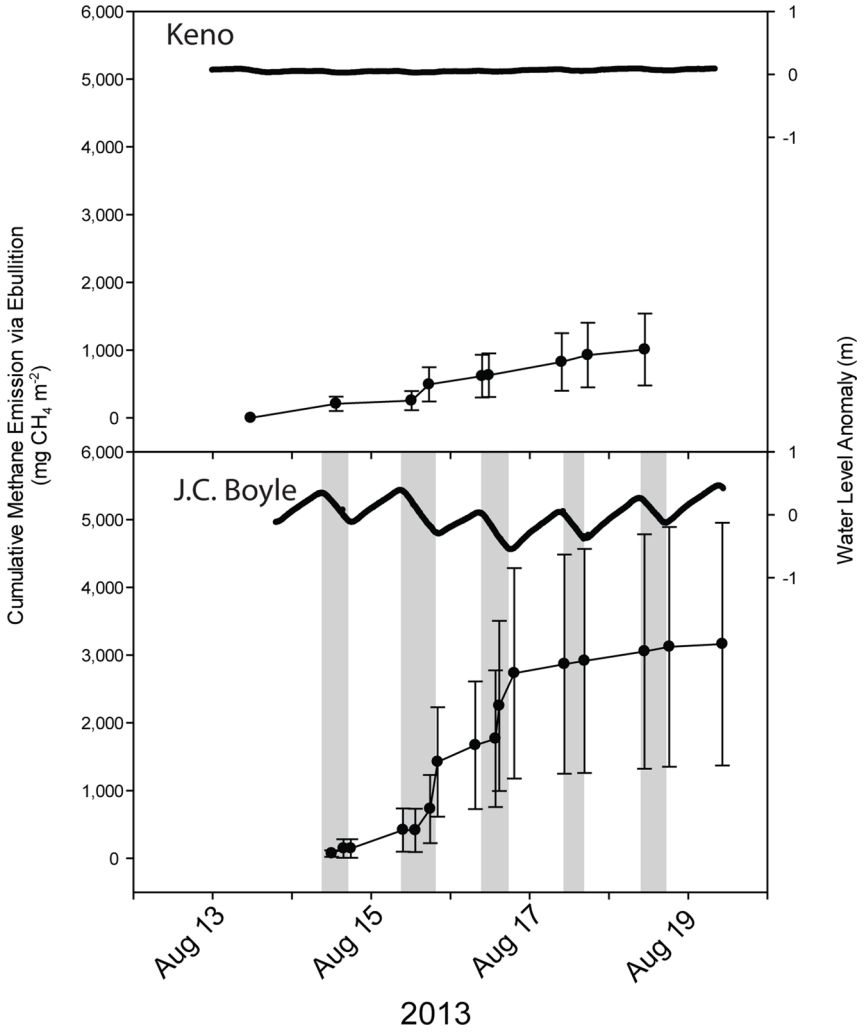

Figure 3. Time series showing average cumulative bubbled mass of CH_4 in traps located at sites in Keno (A) and J. C. Boyle (B) Reservoirs (n = 4 traps in each reservoir). Reservoir drawdown events are represented by shaded gray bars and are also visible as the change in the water surface elevation anomaly (secondary Y-axes and lines). Panels show a contrast between cumulative CH_4 ebullition in two reservoirs positioned in series (i.e., receiving the same water), experiencing different water level management. Error bars represent ±1 SE.

between Keno and J. C. Boyle were due to differences in factors other than drawdown. However, the available data suggest that, if anything, CH_4 emission rates should be greater in Keno than in J. C. Boyle, the opposite of what we observed. Turbidity and [Chl a], both of which would be associated with higher rates of CH_4 production, were higher in Keno than J. C. Boyle. During our study, turbidity was more than 3-fold higher in Keno Reservoir (mean: 28 NTU) than in J. C. Boyle Reservoir (mean: 8 NTU), and [Chl a] was roughly 2-fold higher in Keno than in J. C. Boyle (12.03 μg L^{-1} in Keno vs 6.27 μg L^{-1} in J. C. Boyle). Sediment percent organic C at the most active sites for ebullition was comparable between the two reservoirs (6.42% in Keno vs 7.40% in J. C. Boyle). Bottom water temperatures were slightly higher in Keno (21.24 °C) than in J. C. Boyle (19.71 °C), and average bottom water O_2 concentrations were higher in J. C. Boyle (2.41 mg O_2 L^{-1}) than in Keno (0.64 mg O_2 L^{-1}). Hence temperature and O_2 conditions should both favor greater rates of CH_4 production in Keno than in J. C. Boyle, the opposite of what we observed. Although these measurements were carried out over just a short period of time (5 days), they are consistent with longer-term monitoring data for the two reservoirs, which show very similar water chemistry (i.e., no detectable differences in mean annual concentrations of total nitrogen, dissolved organic nitrogen, nitrate, ammonium, total phosphorus, and soluble reactive phosphorus), and a similar difference in [Chl a] between the two reservoirs to that observed during our study.[39] Temperatures in the two reservoirs are generally within 1 °C of each other (http://www.kbmp.net/maps-data/links-data-reports). In addition, a long-term (8-year) record of barometric pressure fluctuations for the region shows that for the 8 years prior to our study, daily barometric pressure fluctuations never exceeded the pressure changes due to 0.5 m daily water level drawdowns in J. C. Boyle, and barometric pressure was quite stable for weeks prior to our experiment. Hence, it is unlikely that barometric pressure fluctuations occurring beyond the end of our experiment in Klamath River reservoirs would cause net CH_4 fluxes from Keno to catch up to those from J. C. Boyle.

Data from Lacamas Lake also suggest that by enhancing ebullition rates, even temporarily, drawdowns might increase CH_4 emissions. This is due to a strong ($P < 0.001$; $R^2 = 0.5$), positive relationship between ebullition rates and bubble CH_4 concentrations in Lacamas Lake, with concentrations peaking during drawdown events (Figure 2b). During drawdowns in

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Lacamas Lake, average bubble CH_4 concentrations were roughly 2-fold higher than those during nondrawdown periods ($P < 0.001$), averaging 61% CH_4 during drawdown periods and 30% during nondrawdown periods (Figure 2b). There was also a significantly ($P < 0.001$) strong ($R^2 = 0.5$) and positive log−linear relationship between ebullition rate and bubble CH_4 concentrations. This relationship may be due simply to bubble aging and the effect of N_2 stripping from sediments that has been observed previously in other systems,[16,17] but it is also consistent with increased sediment-to-atmosphere transfer efficiency for CH_4 in bubbles emitted during drawdown (due to larger bubble sizes and/or higher sediment or water column CH_4 concentrations). Future efforts should aim to characterize differences in bubble size prior to and during drawdown as well as pore water chemistry and groundwater exchange rates during water level drawdown events. ^{13}C may also be a useful tool to help determine the degree to which CH_4 emitted as bubbles during drawdown events is oxidized compared to CH_4 emitted as bubbles during nondrawdown periods, as in Walter et al. (2008).[48]

Although one might argue that the CH_4 emitted during drawdown events would simply build up in sediments and that the same amount of CH_4 would ultimately be released to the atmosphere, there are several reasons this is probably not the case. It is likely that drawdowns short-circuit the CH_4 oxidation that normally is quite an efficient process in lakes and reservoirs, typically converting 50−95+% of CH_4 to CO_2 before it ever reaches the atmosphere.[7] Although some recent work has shown that some methanogenesis can occur under aerobic conditions,[49−51] the great majority of methanogenesis in lentic systems is thought to occur under highly reduced (O_2-poor) conditions whereas methanotrophy requires the presence of oxygen, or at least an electron acceptor.[52] This means that methanogenesis and methanotrophy generally require different chemical environments and mainly occur in different locations. Hence, any process that interrupts the transfer of CH_4 from methanogens (via diffusion or advection) to methanotrophs must decrease the efficiency of CH_4 oxidation, and thus increase CH_4 "leakage" to the atmosphere. It is also possible that bursts of bubbles associated with drawdowns enrich the water column with CH_4 so that the diffusive exchange of gas between bubbles and the water column is not as efficient as at other times of year. Finally, it is possible that by causing an ebullition event, drawdowns bring CH_4 to the surface that would otherwise be oxidized following fall lake turnover, when well-oxygenated water comes into contact with sediments that have been sitting in an anoxic environment during summer stratification.

Drawdown-Associated Ebullition Events Can Dominate a Reservoir's Annual CH_4 Emission. We were able to estimate the impact of drawdown on CH_4 fluxes in Lacamas Lake, the best-characterized of our study systems. During the 2.5 years over which bubble traps were deployed continuously at Lacamas Lake (2013−2016), 83−91% of total annual CH_4 flux to the atmosphere (ebullition plus diffusion) occurred during the period of drawdown and recovery (56−76 days, or 15−21% of the year). A large fraction (46−70%, depending on the year) of total annual hypolimnetic CH_4 accumulation in Lacamas Lake also occurred during reservoir drawdown. Across all reservoirs in our study, average predrawdown rates of ebullition were not especially high (0.0−192.6 mg CH_4 m^{-2} d^{-1}; mean of all reservoirs: 42.0 mg CH_4 m^{-2} d^{-1}), but rates observed during drawdown (0.09−719.0 mg CH_4 m^{-2} d^{-1}; mean: 223.0 mg CH_4 m^{-2} d^{-1}) fell toward the high end of reported CH_4 ebullition rates for reservoirs and lakes. In fact, the mean ebullition rate we observed during drawdowns was higher than 92.5% of reported reservoir CH_4 flux rates,[3] SI Table S1.).

Together, these results and those presented in previous sections constitute the first evidence that (1) water level drawdowns stimulate CH_4 emissions from reservoir sediments across a range of different types of reservoirs experiencing different management regimes and (2) that the magnitude of ebullition events associated with drawdowns can constitute a large fraction of the total annual CH_4 flux from reservoirs. These insights highlight a need to explicitly consider drawdown events in efforts to quantify CH_4 fluxes, both from individual reservoirs and, where possible, in regional and global CH_4 budgeting efforts.

Controls on Drawdown-Related Ebullition Events. We also examined controls on CH_4 fluxes. Of the factors examined, chlorophyll *a* concentrations [chl *a*] most strongly correlated with CH_4 emissions across reservoirs in this study. CH_4 ebullition, CH_4 diffusion, and the change in CH_4 emissions between predrawdown and drawdown periods all scaled strongly and positively with surface water [chl *a*] ($R^2 = 0.88$, 0.95, and 0.84; $P < 0.0005$, 0.0001, and 0.01, respectively; Figure 4). Although some of the observed relationship between

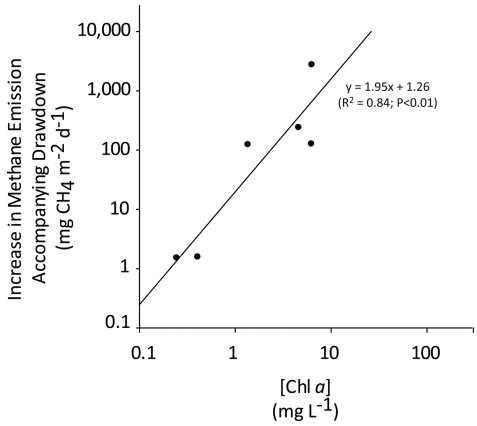

Figure 4. Relationship between average surface (top 3 m) chlorophyll *a* concentrations and increase in CH_4 ebullition accompanying drawdown in each of our study reservoirs. Best fit least-squares regression models and associated statistics are shown.

[chl *a*] and CH_4 emission may be explained by depth ([chl *a*] and depth were inversely correlated in our data set ($r = -0.59$)), the relationship between [chl *a*] and CH_4 ebullition was stronger than the relationship between depth and CH_4 emission ($R^2 = 0.87$ using [chl *a*] as an independent variable and $R^2 = 0.68$ using depth as an independent variable). This is consistent with a recent analysis, showing a relationship between [chl *a*] and CH_4 emission globally.[3]

The J. C. Boyle case suggests that magnitude of drawdown may affect CH_4 emissions within a single reservoir. During a two day experiment in this reservoir, (August 16−17), diel water level drawdowns were extended 0.19 m (first day) and 0.26 additional m (second day) below the standard operating range. CH_4 emissions were larger during these deeper drawdown events (August 16−17, Figure 3) than during standard water level operation (August 15) and during subsequent days (when water levels were not allowed to fall

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as low as they had the previous day, August 17−19). Although depth of drawdown appeared to affect ebullition within J. C. Boyle, there was no detectable relationship between drawdown magnitude and CH_4 emissions when all reservoirs were considered ($P > 0.05$). This suggests that once a critical pressure threshold is crossed (possibly related to recent pressure variation), the magnitude of drawdown ceases to matter, or, alternatively, that other factors (e.g., trophic status) are more important than drawdown magnitude in controlling CH_4 emissions when multiple reservoirs are considered. The magnitude of drawdown that will cause an ebullition event is also likely to vary as a function of CH_4 production rates, reservoir geomorphology, sediment cohesiveness, and reservoir management history. Characterizing these relationships so that predictive models can be developed remains a critical challenge for future research efforts. Similar to drawdown magnitude, there was no detectable relationship between drawdown velocity and any metric of CH_4 production or release, including overall average and during-drawdown ebullitive, diffusive, and total CH_4 fluxes and hypolimnetic CH_4 accumulation ($P > 0.05$ in all cases) across all study reservoirs. Nor was there any significant relationship between drawdown duration and total drawdown-associated CH_4 ebullition, either within a single reservoir across years in the case of Lacamas or across reservoirs ($P > 0.05$ in all cases).

We also tested for correlations between ebullition and alternative potential controlling factors such as near-sediment water temperature, wind speeds (and gusts), wind direction, and changes in atmospheric pressure. To accomplish this, we used high temporal frequency data (one pressure measurement every 5 min) from nondrawdown periods in 2013 at deep water and shallow inlet sites in Lacamas Lake and at a shallow inlet site in Keno Reservoir. Each site was analyzed separately, using ∼4 months of data for Lacamas sites and 3 days of data for the Keno site. At all three sites, all correlations between CH_4 ebullition rates and potential physical drivers of CH_4 ebullition were weak ($r < 0.3$) or statistically insignificant.

Although drawdowns consistently resulted in elevated CH_4 emission across our study reservoirs, there was considerable spatial variability in CH_4 emission both between traps within a single reservoir (e.g., Figures 2 and 3) and between reservoirs (Figure 1). Reservoir mean fluxes spanned 3 orders of magnitude (0.5−465 mg CH_4 m^{-2} d^{-1}), and time-averaged within-reservoir fluxes varied substantially between traps (e.g., 0−952 mg CH_4 m^{-2} d^{-1} in J. C. Boyle, the reservoir with the greatest variation between sampling sites). Bubble dissolution models predict that the importance of ebullition declines as water depth increases,[8] with little ebullition occurring from waters deeper than ∼8 m[5], but others have seen substantial CH_4 bubbling from depths greater than 8 m.[53] Depth was significantly correlated with CH_4 ebullition across all traps in all study reservoirs, with higher fluxes generally occurring at shallower sites ($P < 0.001$, adj. $R^2 = 0.66$). However, we observed some of the highest rates of ebullition at 11 m depth and substantial time-averaged CH_4 ebullition fluxes (3.7 mg CH_4 m^{-2} d^{-1}) in water up to 42 m deep. It has also been suggested that reservoir inlets are especially active sites for CH_4 production and emission, perhaps due to elevated rates of carbon input to these regions.[54,55,43] In our study, inlet sites had the highest average CH_4 fluxes in five out of six reservoirs and the most pronounced response to drawdown in four of the five reservoirs experiencing drawdowns. In J. C. Boyle, the reservoir where the inlet site(s) was not the most active site for CH_4 ebullition, the highest average CH_4 fluxes occurred at a site located near the reservoir's outlet.

Implications and Future Directions. The strong response of eutrophic reservoirs to water level drawdown forcing we report here adds a twist to a recently posited conceptual framework[35] wherein higher CH_4 production rates push systems from a "forcing controlled regime" where forcing mechanisms such as barometric or hydrostatic pressure control CH_4 emissions to a "CH_4 production controlled regime", where CH_4 production rates overwhelm the sediment's capacity to modulate CH_4 emissions. In our study, higher CH_4 emission rates appear to correlate with *greater* sensitivity to physical forcing events, not less (Figure 4), even when drawdown events occur daily, as in J. C. Boyle. This may be due to the fact that sediments can store a large amount of CH_4 relative to what is released to the atmosphere. Of course, if sediment CH_4 storage capacity is far exceeded by CH_4 accumulation, then drawdowns are likely to exert a small relative effect on the timing of CH_4 release because they are likely bubbling at near maximum rates. However, if sediments are capable of holding a large proportion of the CH_4 produced by reservoir sediments in a given year, then drawdowns could increase CH_4 emissions to the atmosphere substantially. This is particularly likely in cases where periodic mixing events "reset" the sediments by bringing terminal electron acceptors (e.g., O_2, NO_3^-, etc.) down to the sediment−water interface, thereby stimulating rapid periodic methane oxidation. In Lacamas Lake, we used the ratios of CH_4 released during drawdowns to CH_4 released during predrawdown periods (Figure 2) to calculate that Lacamas sediments can (conservatively) hold 50−700% of the annual nondrawdown ebullition flux. Lacamas was one of most active emitters of CH_4 in this study, and CH_4 emission rates in Lacamas are high compared to many other lakes and reservoirs globally (higher than 83% of reservoirs in a recent synthesis).[1,3] Hence, Lacamas produces a lot of CH_4 relative to other reservoirs, yet its sediments can hold as much or more CH_4 than it emits to the atmosphere on an annual basis. Although the size of the CH_4 pool held in sediments is likely vary substantially between reservoirs, our results from Lacamas suggest that there could be many systems where within sediment accumulation of CH_4 is substantial relative to annual CH_4 fluxes. In such systems, reservoir water level management could play an important role in controlling CH_4 emissions. The sensitivity of reservoirs to drawdowns is likely to be a function of sediment characteristics (e.g., sediment cohesiveness and, especially, organic matter content[13]), reservoir average depth, the frequency of drawdowns, rates of methanogenesis, and CH_4 oxidation, and is a topic meriting further attention in future work.

Given the diversity of reservoirs and the current lack of information regarding reservoir bathymetry and biogeochemistry at large spatial scales, it is not currently possible to credibly extrapolate from our observations to continental or global scales. However, reservoir drawdowns are quite common, suggesting that this CH_4 release mechanism has potential to affect timing and magnitude of CH_4 emissions significantly at landscape or larger scales. For example, stage height data from 157 U.S. reservoirs showed 95% of reservoirs experienced at least one annual drawdown ≥0.5 m, and 70% experienced multiple drawdown events of this magnitude (median: two events per reservoir[56]). Thus the potential role of reservoir drawdowns as a control on large scale estimates of CH_4 emission from reservoirs deserves further attention. In addition, the potential impact of drawdown-related CH_4 releases on past

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efforts to quantify CH_4 fluxes should be evaluated. Short-term CH_4 ebullition measurements in reservoirs (typical deployments lasting only hours to days) have almost certainly missed pulsed CH_4 bubbling events associated with water-level drawdowns, resulting in substantial underestimates of reservoir CH_4 emissions. The strong effect we observed across all of the reservoirs in this study, the ubiquity of reservoir drawdown events, and the absence of drawdown-associated CH_4 flux estimates from other reservoir CH_4 emissions studies all suggest that CH_4 emission from reservoirs has been substantially underestimated. In addition, as a major increase in the number and cumulative surface area of reservoirs is anticipated globally,[57] the importance of these issues is likely to increase.

Finally, given the importance of water level manipulation in controlling CH_4 fluxes, it is possible that altered reservoir water level management could reduce reservoir greenhouse gas emissions. Data from Keno and J. C. Boyle reservoirs suggest that decreasing the number (and possibly the magnitude) of drawdowns could reduce CH_4 emissions, highlighting a potential trade-off between power generation and greenhouse gas fluxes. It is also possible that altering timing of reservoir drawdown (e.g., by delaying drawdown from the end of the stratified summer period to a period when lake waters are better mixed) might decrease the effect of drawdowns on CH_4 emissions by stimulating rapid methanotrophy at the sediment−water interface, but this hypothesis requires further testing. Furthermore, the observed relationship between eutrophication status and the effect of drawdown on CH_4 emissions (Figure 4) indicates that, in addition to the well-characterized benefits of reducing nutrient loading to aquatic ecosystems,[58,59] this strategy may also mitigate greenhouse gas emissions. This possibility certainly merits further investigation. More specifically, researchers should investigate how reservoir greenhouse gas emissions respond to increases and decreases in nutrient loading and how reservoir characteristics, including management, affect the relationship between eutrophication and greenhouse gas emission. In addition, there is a need to better understand (1) rates and controls of methanogenesis and methanotrophy in reservoir sediments, and particularly the availability of "free" CH_4 (CH_4 in bubbles) to methanotrophs (e.g., through the use of process rate measurements or isotopic indicators of oxidation such as the ^{13}C signature of CH_4), (2) the role of reservoir and sediment characteristics such as geomorphology, temperature, sediment organic matter content, and sediment texture, in determining a sediment's tendency to retain (or release) CH_4 via ebullition, and (3) the physics governing the migration of bubbles upward through sediments as a function of water level fluctuations (i.e., whether upward migration and eruption simply result from increased buoyancy or whether groundwater flushing resulting from a decrease in pressure head or more complex sediment physics (e.g., fracture dynamics) are at play[11,13,60]). Addressing these unknowns would both help address important knowledge gaps and provide management-relevant information that could help reservoir managers to mitigate greenhouse gas emissions.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.6b03185.

Nine figures and one table (PDF)

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