# Methane Ebullition from the Bottom Sediment of Loktak Lake and Its Feeder Stream

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#### **Abstract**

Gas ebullition is one of the principal routes of atmospheric methane (CH<sub>4</sub>) emission from freshwater ecosystems. The current study measured the spatial and temporal variation of CH<sub>4</sub> ebullition from Loktak Lake and one of its feeder streams in northeastern India. The mean CH<sub>4</sub> ebullition from the lake ranged between  $53.2 \pm 31.03$  mL m<sup>-2</sup> d<sup>-1</sup> and  $328.5 \pm 88.33$  mL m<sup>-2</sup> d<sup>-1</sup>. Gas ebullition was comparatively higher in the lake periphery compared to the middle of the lake. The mean CH<sub>4</sub> ebullition from the stream ranged between  $45.6 \pm 23.09 \text{ mL m}^{-2} \text{ d}^{-1}$  and  $522.8 \pm 252.5 \text{ mL m}^{-2} \text{ d}^{-1}$ . In the stream, gas ebullition was recorded consistently higher in the section of the stream meandering through the town. The CH<sub>4</sub> yield from the sediments through force ebullition varied from  $194.8 \pm 31.92$  mL m<sup>-2</sup> to  $1193.8 \pm 250.8$  mL m<sup>-2</sup>. The CH<sub>4</sub> content percentages in the gas bubbles ranged between  $48.0 \pm 8.41$  % and  $55.8 \pm 1.50$  %. There was a seasonal variation in ebullition rates, with the peak emission occurring during the summer. A positive correlation between water temperature and the CH<sub>4</sub> flux was observed in the study, indicating a positive feedback effect of CH<sub>4</sub> emission from climatic warming.

**Keywords:** methane, gas ebullition, Loktak lake, feeder stream, greenhouse gas, sediment

#### INTRODUCTION

Methane is an important greenhouse gas, contributing ~33.0% to the total global greenhouse gases (GHGs) emissions and accounts for 15–20% of global warming (Tiwari *et al.*, 2020; Khoiyangbam *et al.*, 2007). CH<sub>4</sub> is accounted for twenty-eight times more global warming potential than CO<sub>2</sub> (Schaefer, 2019). Global average

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atmospheric CH<sub>4</sub> concentration has increased from 722 ppb in 1750 to 1875 ppb in 2019 (NOAA, 2020). Wetlands are the largest natural source of methane (Dean *et al.*, 2018). It is estimated that approx. 30% of the CH<sub>4</sub> emitted to the atmosphere is produced in wetlands, constituting the single largest natural source (Tian *et al.*, 2016). CH<sub>4</sub> is produced primarily in the sediments of wetland ecosystems after a series of stepwise anaerobic processes (Bridgham *et al.*, 2013). Biogenic CH<sub>4</sub> is produced during the terminal step of organic matter degradation in anaerobic sediments (Lofton *et al.*, 2015). CH<sub>4</sub> exchange in wetlands is a multidimensional process (Mitra *et al.*, 2020). It is emitted to the atmosphere by several processes: through diffusive flux, gas ebullition, plant-mediated flux and the seasonal release of CH<sub>4</sub> trapped in bubbles in ice (Bastviken *et al.*, 2008, Kankaala *et al.*, 2005; Yang *et al.*, 2015; Matthew *et al.*, 2020).

Gas ebullition from freshwater sediments is an important source of atmospheric methane (Delwiche and Hemond, 2017). In the shallow portion of reservoirs, rivers and lakes, ebullition may become a dominant pathway of CH<sub>4</sub> emission (Deshmukh et al., 2016; Wu et al., 2019; Xiao et al., 2017). Ebullition is one of the most complex transport pathways for CH<sub>4</sub> release to measure accurately (Villa et al., 2021). Bubbling can be steady or episodic depending on bio-geophysical conditions (Walter et al., 2006), and ebullition flux variability is more significant between seasons than across microsites (Stanley et al., 2019). Net CH<sub>4</sub> fluxes and ebullition were more significant in patches with emergent vegetation than others (Villa et al., 2021). Plant litter plays a vital role in supplying and regulating CH<sub>4</sub> emissions in wetland ecosystems (Gong et al., 2020). There is an immediate need to explore more options to minimise and arrest GHG emissions for mitigating climate change progression. While doing so, a broader database on the temporal and spatial heterogeneity of CH<sub>4</sub> fluxes through different emission routes will be critical in framing the mitigation strategy. The current study attempts to assess the ebullition of CH<sub>4</sub> in Loktak lake and one of its feeder streams. The prime objective is to understand the spatial and temporal variability of CH<sub>4</sub> flux through ebullition from the aquatic ecosystems. The results will lead to a greater understanding of carbon dynamics and help mitigate GHGs from the wetlands.

#### **MATERIAL AND METHODS**

#### The study site

Loktak Lake is the largest freshwater lake in northeastern India, covering ~286 km² at 768.5 m *a.m.s.l.* It is located between the coordinates of 93° 46′ & 93° 55′ E and 24° 25′ & 24° 42′ N in the valley of the hilly state of Manipur (Fig. 1). Owing to its rich biodiversity and socio-economic importance, Loktak Lake has been listed as a wetland of International Importance under the Ramsar Convention (Ramsar site no. 463, declared on 16<sup>th</sup> June 1993). The lake is fed by many streams that originate from the nearby western hilly landscape. Moirang *Turel* is one such stream that meanders in the valley passing through Moirang town before finally flowing into the lake. The

stream swells during monsoon and gets reduced to narrow water but hardly dries up throughout the year.

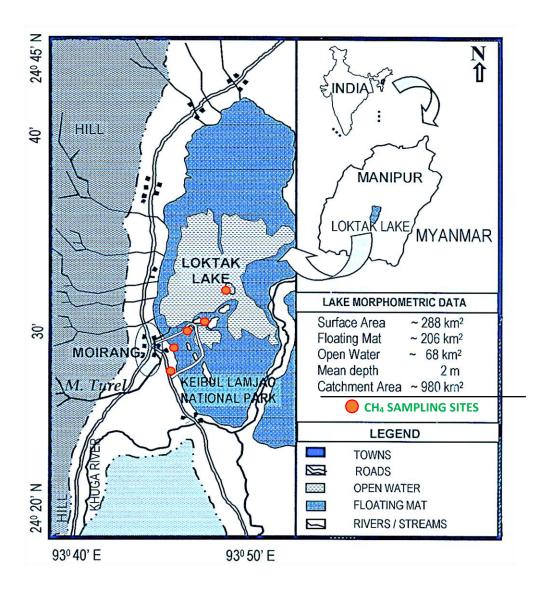


Fig. 1: Map showing the Moirang *Turel* and Loktak Lake

## Gas sample collection from sediments using trap

Gas bubbles released from the lake sediment were intercepted with a gas trap suspended 0.20 m above the sediment surface. The trap consisted of an inverted funnel (0.30 m internal diameter) and a measuring cylinder fixed on an iron tripod stand. The traps were left for several hours until sufficient gas had accumulated and the volume of the trapped gas was measured. Periodically, gas samples were retrieved from the measuring cylinder with an airtight syringe through a septum fixed

at the top of the cylinder and analysed for CH<sub>4</sub> content. Measurements were also made by stirring the sediment to force ebullition until no more gas came out.

#### Gas sample analysis for CH<sub>4</sub>

The gas samples were analysed for CH<sub>4</sub> using a gas chromatograph (Shimadzu, GC-8A) fitted with Flame Ionization Detector (FID) and Porapak N column. The column, injector and detector temperatures were maintained at 70, 130 and 130<sup>0</sup> C, respectively. N<sub>2</sub> was used as the carrier gas, with a flow rate of 20 to 25 mL min<sup>-1</sup>. The flow rates of the hydrogen (the fuel gas) and the zero-air (the supporting gas) was maintained at 25 and 250 mL min<sup>-1</sup>, respectively. The concentration of CH<sub>4</sub> in a gas sample was determined by calculating from the peak area obtained by injecting standard gas mixtures containing known amounts of CH<sub>4</sub> under the same conditions. The primary standard was procured from National Physical Laboratory. The CH<sub>4</sub> flux (F) was calculated using the following equation (Debnath *et al.*, 1996):

$$F = [(C_t - C_0) / t] \times H \times 42.857 \text{ mg m}^{-2} \text{ h}^{-1},$$

Where  $C_0$  is the CH<sub>4</sub> concentration (ppmV) at '0' time,  $C_t$  is the CH<sub>4</sub> concentration (ppmV) after 't' time, t is the time interval, and H is the headspace height (in m).

The gases emission rates were calculated only from the data showing a linear increase of concentration with time.

## RESULTS AND DISCUSSION

#### Gas ebullition from the sediment

The current study was conducted in Loktak lake and one of its feeder streams (Moirang *Turel*), covering three seasons: the pre-monsoon, monsoon and winter. In the feeder stream, three sites each in the upstream water, the section of stream within the town, and the downstream water was selected. In Loktak Lake, the investigation was carried out on the peripheral sediments and in the middle water. Measurements were carried out in two selected months of each season: Pre-monsoon (Period–II: April–May), Monsoon (Period–II: August–September) and Winter (Period–III: November–December). Higher sediment gas generation is well expected in the sections of Moirang *Turel*, meandering through the town, with excessive organic loading. The generated gas accumulates, forming numerous sediments embedded in tiny gasbags. With time, the gasbags grow in size until it reaches a volume and buoyancy that the bags could no longer be retained in the sediment, but get released through ebullition as gas bubbles.

Gas ebullition from the lake sediment occurred throughout the year, with varying rates ranging between 4.0 and 405 mL m<sup>-2</sup> d<sup>-1</sup>, with a mean of  $214 \pm 131$  mL m<sup>-2</sup> d<sup>-1</sup>. The gas ebullition was observed more pronounced during the pre-monsoon (Lake periphery,  $328.5 \pm 88.33$  mL m<sup>-2</sup> d<sup>-1</sup>; Middle water,  $301.8\pm89.50$  mL m<sup>-2</sup> d<sup>-1</sup>) and

tapered down during the monsoon (Lake periphery,  $279.0 \pm 82.25$  mL m<sup>-2</sup> d<sup>-1</sup>; Middle water  $264.0 \pm 65.91$ mL m<sup>-2</sup> d<sup>-1</sup>). The gas generation was minimal during the winter months (Lake periphery,  $59.8 \pm 37.21$  mL m<sup>-2</sup> d<sup>-1</sup>; Middle water,  $53.5 \pm 31.03$  mL m<sup>-2</sup> d<sup>-1</sup>). The mean CH<sub>4</sub> percentage in gas bubbles during the monsoon in the Lake periphery and middle was 51.5 % and 53.3 %, respectively (Table 1). The data on the CH<sub>4</sub> in the gas bubbles indicates that the percentages of CH<sub>4</sub> decreased slightly during the winter season (Period-III) in both the lake zones (periphery and middle water).

**Table 1:** CH<sub>4</sub> ebullition rates in the lake and stream

| Gas ebullition   |                   | Study period       |                   |
|--|-------------------|--------------------|-------------------|
|  | Period – I        | Period – II        | Period – III      |
| A. Undisturbed sediment  |                   |                    |                   |
| I. Lake periphery  |                   |                    |                   |
| Gas ebullition (mL m <sup>-2</sup> d <sup>-1</sup> )           | $328.5 \pm 88.33$ | $279.0 \pm 82.25$  | $59.8 \pm 37.21$  |
| CH <sub>4</sub> content (%)                                    | $51.3 \pm 2.22$   | $51.5 \pm 6.61$    | $51.0 \pm 7.53$   |
| CH <sub>4</sub> released (mL m <sup>-2</sup> d <sup>-1</sup> ) | $168.3 \pm 46.55$ | $147.6 \pm 60.23$  | $32.2 \pm 20.67$  |
| II. Lake Middle water  |                   |                    |                   |
| Gas ebullition (mL m <sup>-2</sup> d <sup>-1</sup> )           | $301.8 \pm 89.50$ | $264.0 \pm 65.91$  | $53.5 \pm 31.03$  |
| CH <sub>4</sub> content (%)                                    | $53.0 \pm 1.41$   | $53.3 \pm 4.62$    | $52.5 \pm 5.45$   |
| CH <sub>4</sub> released (mL m <sup>-2</sup> d <sup>-1</sup> ) | $160.6 \pm 50.08$ | $142.4 \pm 44.90$  | $28.6 \pm 16.73$  |
| III. Upstream (Moirang <i>Turel</i> )                          |                   |                    |                   |
| Gas ebullition (mL m <sup>-2</sup> d <sup>-1</sup> )           | $290.8 \pm 64.54$ | $285.3 \pm 116.3$  | $55.0 \pm 31.73$  |
| CH <sub>4</sub> content (%)                                    | $55.3 \pm 4.5$    | $53.3 \pm 3.10$    | $50.2 \pm 3.6$    |
| CH <sub>4</sub> released (mL m <sup>-2</sup> d <sup>-1</sup> ) | $161.4 \pm 39.64$ | $151.7 \pm 62.98$  | $27.2 \pm 15.07$  |
| IV. Within town (Moirang <i>Turel</i> )                        |                   |                    |                   |
| Gas ebullition (mL m <sup>-2</sup> d <sup>-1</sup> )           | $522.8 \pm 252.5$ | $436.25 \pm 205.6$ | $47.75 \pm 35.98$ |
| CH <sub>4</sub> content (%)                                    | $55.8 \pm 1.50$   | $52.3 \pm 4.11$    | $53.0 \pm 6.82$   |
| CH <sub>4</sub> released (mL m <sup>-2</sup> d <sup>-1</sup> ) | $291.8 \pm 140.2$ | $223.8 \pm 100.7$  | $26.1 \pm 20.32$  |
| V. Downstream (Moirang <i>Turel</i> )                          |                   |                    |                   |
| Gas ebullition (mL m <sup>-2</sup> d <sup>-1</sup> )           | $478.0 \pm 201.3$ | $338.2 \pm 118.2$  | $45.6 \pm 23.09$  |
| CH <sub>4</sub> content (%)                                    | $53.33 \pm 3.39$  | $51.56 \pm 4.88$   | $50.33 \pm 3.16$  |
| CH <sub>4</sub> released (mL m <sup>-2</sup> d <sup>-1</sup> ) | $261.8 \pm 113.9$ | $167.2 \pm 63.66$  | $22.6 \pm 11.48$  |

| Gas ebullition                                 | Study period      |                    |                    |
|--|-------------------|--------------------|--------------------|
|  | Period – I        | Period – II        | Period – III       |
|  |                   |                    |                    |
| B. Force ebullition                            |                   |                    |                    |
| I. Lake periphery                              |                   |                    |                    |
| Gas ebullition (mL m <sup>-2</sup> )           | $825.6 \pm 58.58$ | $818.3 \pm 69.64$  | $209.3 \pm 44.56$  |
| CH <sub>4</sub> content (%)                    | $55.6 \pm 4.72$   | $53.3 \pm 6.24$    | $49.3 \pm 4.57$    |
| CH <sub>4</sub> released (mL m <sup>-2</sup> ) | $457.5 \pm 67.96$ | $438.9 \pm 86.55$  | $104.2 \pm 28.33$  |
| II. Lake Middle water                          |                   |                    |                    |
| Gas ebullition (mL m <sup>-2</sup> )           | $830.3 \pm 72.85$ | $831.5 \pm 61.90$  | $295.5 \pm 30.14$  |
| CH <sub>4</sub> content (%)                    | $54.8 \pm 5.06$   | $55.6 \pm 5.91$    | $48.0 \pm 8.41$    |
| CH <sub>4</sub> released (mL m <sup>-2</sup> ) | $457.1 \pm 80.25$ | $458.7 \pm 52.97$  | $141.8 \pm 38.10$  |
| III. Upstream (Moirang <i>Turel</i> )          |                   |                    |                    |
| Gas ebullition (mL m <sup>-2</sup> )           | $791.8 \pm 36.16$ | $711.0 \pm 80.56$  | $194.8 \pm 31.92$  |
| CH <sub>4</sub> content (%)                    | $53.8 \pm 7.46$   | $54.8 \pm 9.81$    | $50.6 \pm 10.2$    |
| CH <sub>4</sub> released (mL m <sup>-2</sup> ) | $425.2 \pm 59.09$ | $393.9 \pm 115.1$  | $95.5 \pm 4.73$    |
| IV. Within town (Moirang <i>Turel</i> )        |                   |                    |                    |
| Gas ebullition (mL m <sup>-2</sup> )           | 1193.8 ± 250.8    | $772.8 \pm 68.46$  | $242.0 \pm 110.8$  |
| CH <sub>4</sub> content (%)                    | $54.3 \pm 7.89$   | $55.3 \pm 7.93$    | $51.6 \pm 11.03$   |
| CH <sub>4</sub> released (mL m <sup>-2</sup> ) | $649.3 \pm 187.8$ | $423.1 \pm 25.76$  | $120.9 \pm 54.24$  |
| V. Downstream (Moirang <i>Turel</i> )          |                   |                    |                    |
| Gas ebullition (mL m <sup>-2</sup> )           | $845.2 \pm 92.64$ | $811.4 \pm 105.86$ | $226.60 \pm 58.04$ |
| CH <sub>4</sub> content (%)                    | $54.2 \pm 4.55$   | $53.2 \pm 6.42$    | $50.4 \pm 6.62$    |
| CH <sub>4</sub> released (mL m <sup>-2</sup> ) | $457.3 \pm 54.78$ | $432.3 \pm 77.99$  | $116.0 \pm 41.26$  |

 $<sup>\</sup>pm$  denotes SD of the means

The mean gas ebullition rates in the upstream section of Moirang *Turel* was 290.8  $\pm$  64.54 mL m<sup>-2</sup> d<sup>-1</sup> (Period-I); 285.3  $\pm$  116.3 mL m<sup>-2</sup> d<sup>-1</sup> (Period-II); and 55.0  $\pm$  31.73 mL m<sup>-2</sup> d<sup>-1</sup> (Period-III), respectively (Table 1). Corresponding mean gas ebullition rates during the three seasons in the downstream water were 522.8  $\pm$  252.5 mL m<sup>-2</sup> d<sup>-1</sup> (Period-I); 436.25  $\pm$  205.6 mL m<sup>-2</sup> d<sup>-1</sup> (Period-II); 47.75  $\pm$  35.98 mL m<sup>-2</sup> d<sup>-1</sup> (Period-III). The mean gas ebullition rates in the section of the stream, flow passing the town

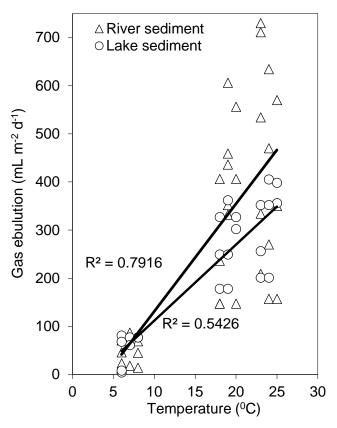
were  $478.0 \pm 201.3$  mL m<sup>-2</sup> d<sup>-1</sup> (Period-I);  $338.2 \pm 118.2$  mL m<sup>-2</sup> d<sup>-1</sup> (Period-II);  $45.6 \pm 118.2$  mL m<sup>-2</sup> d<sup>-1</sup> (Period-III);  $45.6 \pm 118.2$  mL m<sup>-2</sup> d<sup>-1</sup> (Period-IIII);  $45.6 \pm 118.2$  mL m<sup>-2</sup> d<sup>-1</sup> (Period-III);  $45.6 \pm 118.2$  mL m<sup>-2</sup> d<sup>-1</sup> (Period-IIII);  $45.6 \pm 118.2$  mL 23.09 mLm<sup>-2</sup> d<sup>-1</sup> (Period-III). Season wise the gas ebullition rates were higher during the pre-monsoon (Period-I), followed subsequently by the monsoon (Period-II) and winter (Period-III). Variation in the ebullition rates is primarily affected by the difference in water temperature during the different seasons. It can be observed that there was a positive correlation between lake water temperature and gas ebullition from the stream water ( $R^2 = 0.792$ ) (Fig. 2). In a study in aquaculture ponds in China (Yang et al., 2020), the authors established an evident seasonal variation of CH<sub>4</sub> ebullition from the aquatic ecosystems. Sediment temperature was found to influence the seasonal variation in the CH<sub>4</sub> ebullition. A rise in temperature of the water leads to an increase in the rate of chemical reaction in water besides reducing the solubility of gases (Haroon, et al., 2010). Spatially, gas ebullition was observed highest in the section of the stream, flow passing the town and lowest in the upstream water. The gas ebullition rates in the downstream water in between the two. The spatial variation in the ebullition rates was primarily attributed to the availability of carbon substrates for gas generation. The mean CH<sub>4</sub> percentages in the gas bubble collected from stream water ranged between were 50.2 and 55.8 % during these periods.

## Force ebullition of sediment gas

The seasonal variation in the amount of gas released between the pre-monsoon and monsoon through force ebullition in the lake was relatively minimal. This variation was observed in both the peripheral sediment (Period-I,  $825.6 \pm 58.58$  mL m<sup>-2</sup>; Period-II,  $818.3 \pm 69.64$  mL m<sup>-2</sup>) and middle sediment (Period-I,  $830.3 \pm 72.85$  mL m<sup>-2</sup>; Period-II,  $831.5 \pm 61.90$  mL m<sup>-2</sup>). However, a marked decrease in gas yield was observed during the winter season (Lake periphery,  $209.3 \pm 44.56$  mL m<sup>-2</sup>; Middle water,  $295.5 \pm 30.14$  mL m<sup>-2</sup>). The mean CH<sub>4</sub> percentages in the gas extracted through force ebullition during the three seasons ranged between 48.0 and 55.6 % (Table 1).

In contrast, the amount of gas released through force ebullition in the stream closely followed the ebullition trend observed in the undisturbed measurements. The amount of gas yield was highest during the pre-monsoon (Upstream,  $791.8 \pm 36.61$  mL m<sup>-2</sup>; within town,  $1193.8 \pm 250.8$  mL m<sup>-2</sup>; downstream,  $845.2 \pm 92.64$  mL m<sup>-2</sup>) followed by the monsoon (Upstream,  $711.0 \pm 80.56$  mL m<sup>-2</sup>; Within the town,  $772.8 \pm 68.46$  mL m<sup>-2</sup>; downstream,  $811.4 \pm 105.9$  mL m<sup>-2</sup>). The amount of gas extracted through force ebullition was minimal during the winter season (upstream,  $194.8 \pm 31.92$  mL m<sup>-2</sup>; within the town,  $242.0 \pm 110.8$  mL m<sup>-2</sup>; downstream,  $226.60 \pm 58.04$  mL m<sup>-2</sup>). The overall mean CH<sub>4</sub> percentages in the gas extracted through force ebullition of the stream ranged between were 48.0 to 55.6% (Table 1). The aforementioned observed values of bubble gas ebullition from the stream and lake sediment were more prominent than those reported for acidic bog lake by Casper *et al.* (2003), with a total gas release of 76-77 mL m<sup>-2</sup> d<sup>-1</sup>. In some portions of Loktak lake, there has been continuous ebullition of gas. Such sites may be designated to capture CH<sub>4</sub>. Methane

capture has certain associated advantages as the captured CH<sub>4</sub> can be appropriately utilised as an energy source.



**Fig. 2:** Mean gas ebullition rates, in the stream  $[\Delta]$  and lake [O], plotted against water temperature

### **CONCLUSION**

Methane is a greenhouse gas twenty-eight times more potent than CO<sub>2</sub> on a 100-year time horizon. Wetlands play an essential role in the global carbon cycle and act as sources and sinks of carbon. The majority of the existing literature on CH<sub>4</sub> emission studies from wetlands focuses on diffusive emission. However, ebullition has been well established as a significant route for CH<sub>4</sub> release from these ecosystems. Despite these underlying facts, CH<sub>4</sub> ebullition from aquatic ecosystems remains poorly understood. This study evaluates the spatial and temporal variation in the CH<sub>4</sub> ebullition in a freshwater lake and its feeder stream. The results revealed that CH<sub>4</sub> ebullition from the lake and feeder streams showed both spatial and temporal variation. In the stream, gas ebullition was recorded consistently higher in the section of the stream meandering through the town. The results established a positive correlation between lake water temperature and fluxes of CH<sub>4</sub>. It is noteworthy that wetland ecosystems like Loktak lake, if restored and appropriately conserved, may serve as a carbon sink, reducing the emission of greenhouse gases.

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