



## Greenhouse gas budgets of severely polluted urban lakes in India

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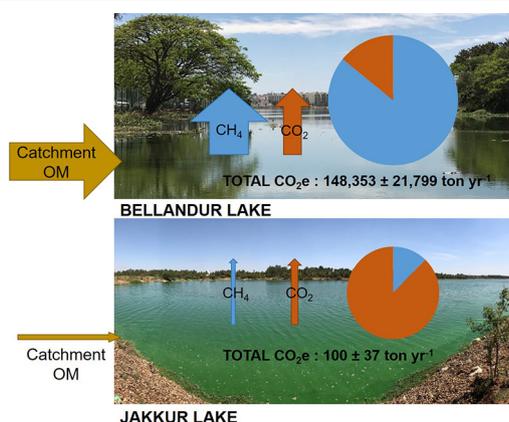
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### HIGHLIGHTS

- Emissions of greenhouse gases from polluted lakes may be an overlooked global source.
- Exceptionally high methane concentrations were measured in Bellandur Lake, India.
- In Jakkur Lake, control of organic matter inputs has reduced methane concentrations.
- Lake restoration can also deliver reductions in greenhouse gas emissions.

### GRAPHICAL ABSTRACT



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### ABSTRACT

Inland waters are important sources of greenhouse gases and emissions from polluted subtropical systems may be contributing to the observed global increase in atmospheric methane concentrations. Here we detail a scoping study where dissolved concentrations of greenhouse gases methane ( $\text{CH}_4$ ), carbon dioxide ( $\text{CO}_2$ ) and nitrous oxide ( $\text{N}_2\text{O}$ ) were measured in two contrasting urban lakes in Bangalore (*Bengaluru*), Karnataka, India, from June 2018 to February 2020. Bellandur Lake is a severely polluted system whilst Jakkur Lake has been subject to partial restoration via treatment of organic matter inputs. Methane concentrations in Bellandur Lake were three orders of magnitude higher than in Jakkur Lake, with a mean concentration of  $3.02 \pm 1.57 \text{ mg CH}_4\text{-C L}^{-1}$  compared to  $1.72 \pm 1.22 \mu\text{g CH}_4\text{-C L}^{-1}$ . At Bellandur Lake, dissolved  $\text{CO}_2$  concentrations were of the same order of magnitude as for  $\text{CH}_4$ , whereas at Jakkur Lake dissolved  $\text{CO}_2$  concentrations were two orders of magnitude greater than for  $\text{CH}_4$ . Concentrations of  $\text{N}_2\text{O}$  were negligible in both lakes. Extrapolating our data to estimate greenhouse gas fluxes, mean daily methane fluxes from Bellandur Lake were consistently in excess of  $1000 \text{ mg CH}_4 \text{ m}^2 \text{ d}^{-1}$ , rendering the lake a source of GHGs to the order of  $148,350 \pm 21,790 \text{ ton yr}^{-1} \text{ CO}_2\text{-e yr}^{-1}$ , compared to  $100 \pm 37 \text{ ton CO}_2\text{-e yr}^{-1}$  from Jakkur Lake, with  $\text{CH}_4$  contributing primarily to this difference. We propose that the contribution of severely polluted urban lakes to global  $\text{CH}_4$  production warrants further investigation, particularly as our evidence suggests that standard secondary wastewater treatment to support restoration of these systems has the potential to significantly reduce  $\text{CH}_4$  emissions.

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## 1. Introduction

Inland waters are active components in global biogeochemical cycles and are known to be important sources of greenhouse gases (GHGs). Global emissions from these systems have been estimated at 2.1 Pg C yr<sup>-1</sup> for carbon dioxide (CO<sub>2</sub>; Raymond et al., 2013), between 40 and 100 Tg yr<sup>-1</sup> for methane (CH<sub>4</sub>; Bastviken et al., 2011; Drake et al., 2018) and between 148 and 277 Gg N yr<sup>-1</sup> for nitrous oxide (N<sub>2</sub>O; Maavara et al., 2019). Uncertainties surrounding total freshwater GHG emissions are considerable; in part due to the global distribution of studies, which are typically biased towards Europe and North America, and by the tendency of aquatic GHG studies to focus on rural environments.

Of the total freshwater CH<sub>4</sub> emissions, more than 40% has been attributed to subtropical and tropical inland waters, where biogenic processes facilitating CH<sub>4</sub> production are exacerbated by temperature (Bastviken et al., 2011). This coincides with the main locus for growth in global CH<sub>4</sub> concentrations, between 30°S and 30°N (Saunois et al., 2016), and it is likely that increased emission of CH<sub>4</sub> from inland waters, including wetlands, reservoirs and lakes, is contributing to the rising trend in atmospheric CH<sub>4</sub> (10 ppb yr<sup>-1</sup>) that has been observed over the past decade (Nisbet et al., 2019; Kirschke et al., 2013; Saunois et al., 2016). A shift in the isotopic signature of CH<sub>4</sub> suggestive of a biogenic source further supports the need to investigate emissions from subtropical and tropical inland waters (Nisbet et al., 2019; Schaefer et al., 2016).

The production of CH<sub>4</sub> in aquatic systems, driven by the supply of organic matter (OM) and the presence of low oxygen conditions, is intensified by excesses of OM and nutrients (Beaulieu et al., 2019; Sepulveda-Jauregui et al., 2018), and at the global scale, an estimated 0.1 Pg C yr<sup>-1</sup> of human sewage is exported to inland waters in the form of readily bioavailable OM (Regnier et al., 2013). In regions where urbanisation outpaces the development of water treatment infrastructure, water quality can be compromised by discharges of substantial quantities of anthropogenic OM to freshwaters. As such, urban freshwaters with poor water quality appear to be a highly plausible, yet largely overlooked, contributing source in the recently observed acceleration of global CH<sub>4</sub> emissions.

In this scoping study, we present the GHG budgets of two contrasting urban lakes in Bengaluru, Karnataka, India. Specifically, we compare dissolved GHG concentrations in a lake that is highly polluted by untreated wastewater versus a partially remediated system, receiving treated wastewater, to answer the following questions:

- 1) What is the dominant GHG in the budget for both the polluted and partially remediated lakes?
- 2) How does wastewater treatment affect the GHG budget of receiving water bodies?

## 2. Methods

### 2.1. Site descriptions

Bengaluru – once known as the ‘city of lakes’ – has a network of >200 manmade reservoirs, of varying size and condition. Jakkur Lake is situated in the north of Bengaluru and has a surface area of 0.65 km<sup>2</sup> with a catchment area of 19.2 km<sup>2</sup> (Table 1). The lake has been subject to partial remediation over recent years (Fig. 1) and the Jakkur sewage treatment plant (STP) provides secondary treatment of wastewater before it is discharged into the lake at inlet JK3 (Fig. 2). Sewage received by the STP is treated through an activated sludge process in combination with nitrate reduction. The STP effluent is then diverted through a constructed wetland before entering the main body of the lake. At the start of sampling Jakkur Lake received 10 million litres per day (MLD) of treated water and the STP capacity was upgraded to 15 MLD in

September 2018 (Fig. 1). The lake supports a range of ecosystem services, such as fisheries, recreation and local groundwater recharge.

In contrast, Bellandur Lake is a larger lake situated to the east of Bengaluru, which is considered a severely polluted system. At the time of initial sampling Bellandur Lake was receiving an estimated 268 MLD of raw sewage from the surrounding urban catchment. In November 2019, the construction of a number of diversion channels began, intended to prevent urban inputs entering the main lake body (Fig. 1). A notable difference from Jakkur Lake is the presence of a naturally existing wetland in the south-west of Bellandur, which interfaces directly with the lake (Fig. 2).

Bellandur Lake has made international news in recent years for spontaneously catching fire on several occasions (Bhasti, 2017), with CH<sub>4</sub> emissions implicated, but not formally measured. It receives 40% of Bengaluru's untreated sewage and other industrial waste (around 520 MLD, of which, at present, 250 MLD are treated and diverted for irrigation to the villages upstream), with minimal inputs of fresh water during monsoons. The two lakes in this study, therefore, represent systems under contrasting and changing management regimes and, provide a natural experiment for a study on the effect of pollution on GHG concentrations in urban lakes to illustrate how sewage treatment affects the GHG budget of receiving water bodies.

### 2.2. Headspace sampling

Samples were collected between June 2018 and February 2020 at three primary sites at Bellandur Lake and five sites at Jakkur Lake (Fig. 2). Site BL6 at Bellandur became inaccessible during the sampling period due to construction of a diversion channel (Fig. 1), and was replaced by sampling at site BLN1 then BLN2, which were both deemed representative of BL6. Dissolved gas samples were collected in triplicate at each sampling location following the widely cited headspace method (Billett and Moore, 2008; Kling et al., 1992). Ambient air samples were also collected in triplicate 2 m above the water surface using a 200 mL syringe fitted with a 3-way valve. Approximately 100 mL of air sample was injected into 20 mL glass vials, pre-sealed with a crimp cap and rubber plug. A double-needle system was used to fill the vials, with excess sample escaping through the outlet needle. Both needles were removed simultaneously to prevent any loss or dilution of sample.

Headspace samples were also taken with a 200 mL syringe. 100 mL of water was drawn up from approximately 10 cm below the water surface, with a further 100 mL of air drawn just above the water's surface. The syringe was shaken vigorously for 90 s to fully equilibrate gas between the water and the headspace (Garnett et al., 2016). Approximately 100 mL of headspace gas was then injected into 20 mL glass vials using the same protocol as for the air samples, taking care not to allow any water entering the sample vial.

### 2.3. Gas chromatography analysis

Samples were analysed by standard gas chromatographic techniques, using an Agilent 7890B gas chromatograph (GC) and 7697A headspace autosampler (Agilent, Santa Clara, California). The instrument was fitted with a flame ionisation detector (FID) for the measurement of CH<sub>4</sub> and CO<sub>2</sub>; and a micro electron capture detector (μECD) for the measurement of N<sub>2</sub>O. The limits of detection were 40 ppb, 5000 ppb and 5 ppb for CH<sub>4</sub>, CO<sub>2</sub> and N<sub>2</sub>O respectively (Drewer et al., 2020). Gas samples were stored, in ambient conditions, for up to three months prior to analysis. Laboratory storage tests have confirmed that samples can be stored in ambient conditions for periods exceeding six months, with no observable effect on measured concentrations.

Sample concentrations were determined from the relationship between peak area and standard concentration, derived from calibration with four mixed gas standards. Standard concentrations ranged from below ambient to high: 1.12 to 98.2 ppm for CH<sub>4</sub>; 202 to 5253 ppm for CO<sub>2</sub>; and 0.208 to 1.04 ppm for N<sub>2</sub>O. In some instances, headspace

**Table 1**

Physical and chemical characteristics of Bellandur Lake and Jakkur Lake. Water chemistry data represent ATREE 2019 averages (total sample numbers are indicated by n), where SRP refers to soluble reactive phosphorus, TP refers to total phosphorus and DO refers to dissolved oxygen.

	Bellandur	Jakkur
Latitude, longitude	12.935638, 77.672224	13.087504, 77.610769
Catchment area (km <sup>2</sup> )	279	19.2
Lake surface area (km <sup>2</sup> )	3.7	0.65
No. of inlets	5	3
Mean depth (m)	2.6	3.19
Maximum depth (m)	4.6	3.3
Mean annual SRP (mg L <sup>-1</sup> )	7.0 ± 2.76 (n = 7)	3.0 ± 2.04 (n = 8)
Mean annual TP (mg L <sup>-1</sup> )	5.14 ± 1.33 (n = 7)	3.4 ± 1.95 (n = 8)
Mean annual DO (mg L <sup>-1</sup> )	0.12 ± 0.14 (n = 6)	17.6 ± 5.76 (n = 10)

CH<sub>4</sub> concentrations were exceptionally high and fell outside the range of the calibration standards. These samples were diluted 100-fold with inert nitrogen gas to levels which could be measured reliably by the instrument. Dilutions of this magnitude were found to overestimate CH<sub>4</sub> and CO<sub>2</sub> concentrations; with errors of 5 to 19% for CH<sub>4</sub>, and 19 to 22% for CO<sub>2</sub>. N<sub>2</sub>O concentrations in diluted samples could not be reliably measured as concentrations were already low; dilution resulted in concentrations dropping below the detection limit.

#### 2.4. Mass budget calculations

Two sites were chosen for detailed mass budget and GHG evasion investigation: Bellandur Outlet (BL1), and Jakkur Observation Deck (JK1). These two sites were the most frequently sampled (n = 6 at both sites). BL1, the Varthur outlet in Bellandur Lake, demonstrated relatively still water before the more turbulent outlet and was hence deemed comparable to JK1, the main lake sampling site in Jakkur Lake. The outlet sampling point at Jakkur (JK7) was turbulent and therefore not appropriate for use in scaling GHG observation data to the whole lake.

Total dissolved gas was calculated from the measured headspace and ambient air concentrations following Henry's Law and using the Bunsen solubility method to adjust gas solubility for temperature (Wiesenburg and Guinasso, 1979). Dissolved gas concentrations in mg C m<sup>-3</sup> ( $C_{dissolved}$ ) were multiplied by the lake volume in m<sup>3</sup> ( $V_{lake}$ ) to derive approximations of the whole lake GHG mass budget in tonnes ( $mass_{GHG}$ ), as given by Eq. (1):

$$mass_{GHG} = (C_{dissolved} \cdot V_{lake}) \cdot 10^{-9} \quad (1)$$

#### 2.5. Evasion flux calculations

Evasion fluxes ( $F$ ) of CH<sub>4</sub> and CO<sub>2</sub> were calculated for each sampling time point using Eq. (2):

$$F_G = k_G \cdot \Delta C_G \quad (2)$$

In Eq. (2),  $k_G$  is the transfer velocity for gas  $G$  (in m·s<sup>-1</sup>), and  $\Delta C_G$  is the difference between the measured concentrations of gas  $G$  in water and air (in g·m<sup>-3</sup>) (Cole and Caraco, 1998, Bastviken et al., 2004).

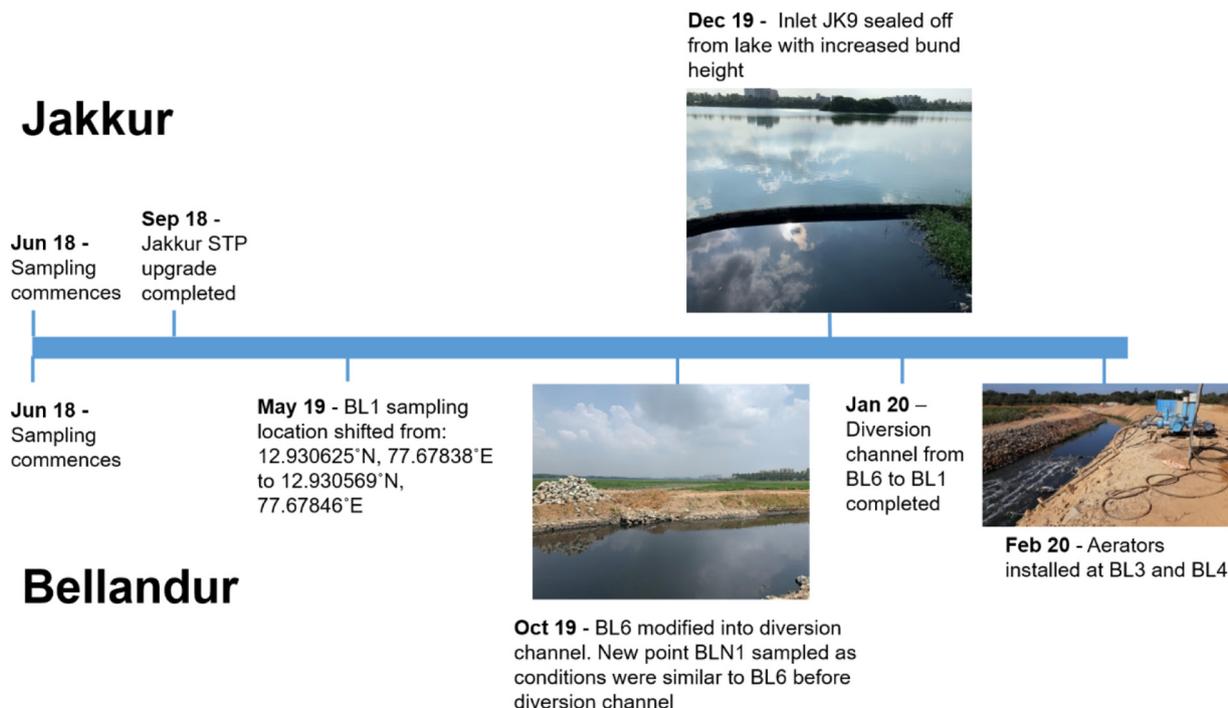
The transfer coefficient  $k_G$  was calculated for each sampling time point as (Prairie and del Giorgio, 2013):

$$k_G = \frac{k_{600}}{(Sc_G/600)^n} \quad (3)$$

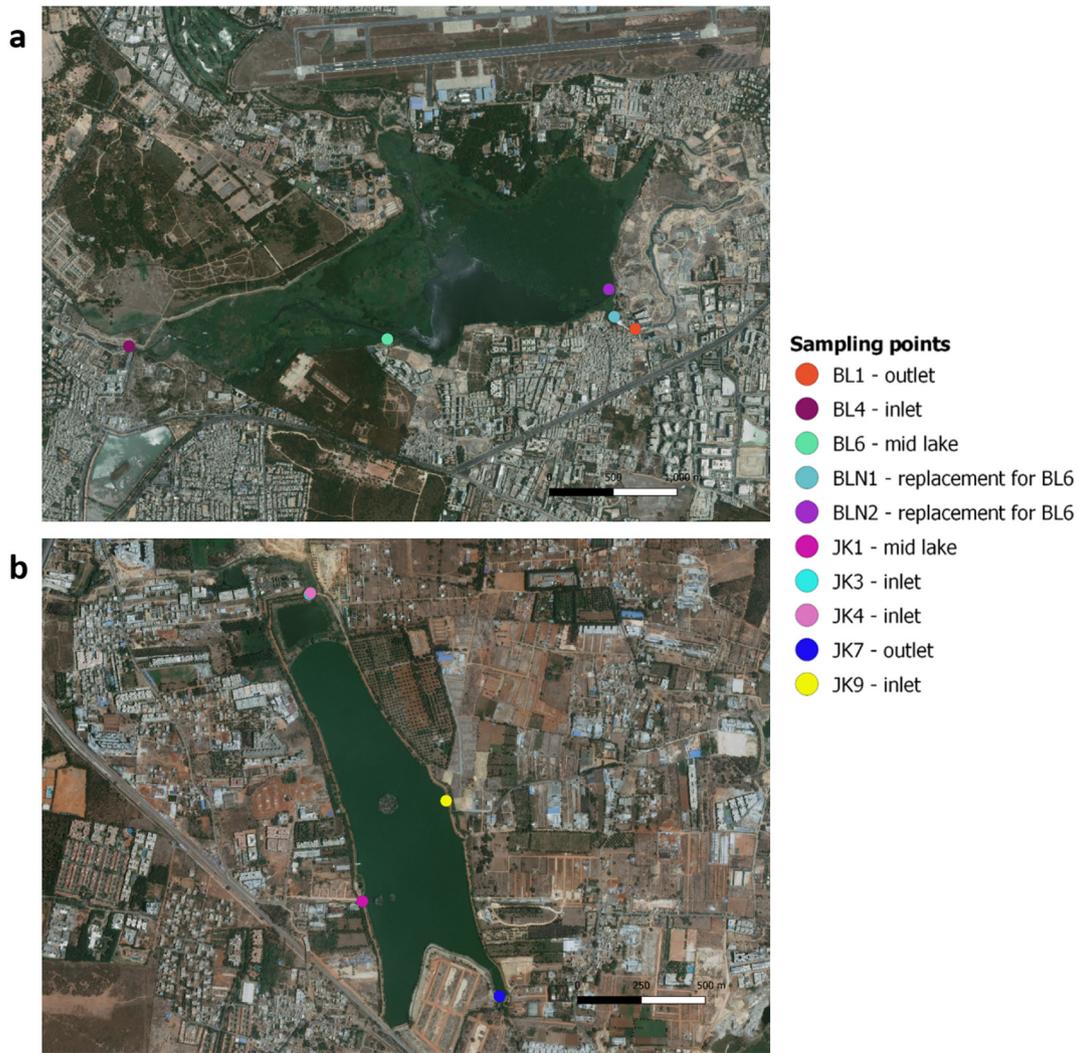
In Eq. (3),  $k_{600}$  is the transfer velocity of gas  $G$  normalised to a Schmidt number of 600,  $Sc_G$  is the Schmidt number for gas  $G$  at temperature  $T$ , and  $n$  is a wind speed-related constant with value 2/3 and 1/2 for wind speed < 3.7 m s<sup>-1</sup> and >3.7 m s<sup>-1</sup> respectively (Guérin et al., 2007).

The Schmidt number ( $Sc_G$ , Eq. (4)) for each gas was calculated using the third-degree polynomial relation with temperature proposed by Wanninkhof (1992):

$$Sc_G = A - BT + CT^2 - DT^3 \quad (4)$$



**Fig. 1.** Chronology of in-lake management at Jakkur and Bellandur lakes during the sampling period.



**Fig. 2.** Aerial image of Bellandur Lake with gas and headspace sampling locations marked (a). Aerial image of Jakkur Lake with gas and headspace sampling locations marked (b). Satellite imagery obtained from Bing Maps™.

The values of the non-linear fit coefficients (A, B, C and D) in Eq. (4) for CH<sub>4</sub> and CO<sub>2</sub> dissolved in fresh water are summarised in Table 2.

The normalised transfer velocity  $k_{600}$  (Eq. (5)) was calculated using the parametrisation on  $U_{10}$  (wind speed at 10 m above the surface) proposed by Cole and Caraco (1998):

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

Finally,  $U_{10}$  (Eq. (6)) was calculated from wind speed measurements taken at height  $z = 5$  m, assuming a logarithmic wind profile with displacement height  $d = 0.001$  m and roughness length  $z_0 = 0.0002$  m:

$$U_{10} = u(z) \frac{\ln\{(10-d)/z_0\}}{\ln\{(z-d)/z_0\}} \quad (6)$$

**Table 2**

Values of the third-order polynomial coefficients used in the calculation of the temperature-dependent Schmidt number (Eq.) for CO<sub>2</sub> and CH<sub>4</sub> in fresh water (reproduced from Table A1 in Wanninkhof (1992)).

Gas	A	B	C	D
CH <sub>4</sub>	1897.8	114.28	3.2902	0.039061
CO <sub>2</sub>	1911.1	118.11	3.4527	0.04132

### 2.5.1. Uncertainties on evasion fluxes

Following error propagation rules, the total uncertainty (expressed as a variance,  $\sigma_F^2$ ) on the evasion flux (Eq. (2)) is given by:

$$\sigma_F^2 = \sigma_k^2 \left( \frac{\partial F}{\partial k} \right)^2 + \sigma_{\Delta C}^2 \left( \frac{\partial F}{\partial \Delta C} \right)^2 \quad (7)$$

Evaluating the partial derivatives (denoted by  $\partial$ ), Eq. (7) becomes:

$$\sigma_F^2 = \sigma_k^2 (\Delta C)^2 + \sigma_{\Delta C}^2 k^2 \quad (8)$$

Using the definitions of  $k$  and  $k_{600}$  (Eqs. (3) and (5)), the variance on  $k$  can be expressed as:

$$\sigma_k^2 = \sigma_{U_{10}}^2 \left( \frac{\partial k}{\partial U_{10}} \right)^2 + \sigma_{Sc}^2 \left( \frac{\partial k}{\partial Sc} \right)^2 \quad (9)$$

Evaluating the partial derivatives, Eq. (9) can be re-written as:

$$\sigma_k^2 = \frac{1}{(Sc/600)^{2n}} \left[ \sigma_{U_{10}}^2 \left( 0.357 \times U_{10}^{0.7} \right)^2 + \sigma_{Sc}^2 \left\{ \frac{n}{Sc} \left( 2.07 + 0.21U_{10}^{1.7} \right) \right\}^2 \right] \quad (10)$$

Using the parametrisation of the Schmidt number ( $Sc$ ) on temperature ( $T$ ) given in Eq. (4), the variance term for  $Sc$  is:

$$\sigma_{Sc}^2 = \sigma_T^2 (-B + 2CT - 3DT^2) \tag{11}$$

Assuming that the uncertainties on measurement height ( $z$ ), displacement height ( $d$ ) and roughness length ( $z_0$ ) are negligible compared to the uncertainty on wind speed ( $u$ ), it follows from the definition of  $U_{10}$  (Eq. (6)) that the uncertainty on  $U_{10}$  is:

$$\sigma_{U_{10}}^2 \approx \sigma_u^2 \left[ \frac{\ln \left\{ \frac{(10-d)/z_0}{(z-d)/z_0} \right\}}{\ln \left\{ (z-d)/z_0 \right\}} \right]^2 \tag{12}$$

Finally, the variance on  $k$  can be evaluated by inserting Eqs. (11) and (12) into Eq. (10).

The uncertainties on concentrations ( $\sigma_{\Delta C}^2$ ) were set to 19% for  $CH_4$  and 22% for  $CO_2$  (maximum values of dilution-related errors estimated from gas chromatography analysis).

For derivation of a global warming potential (GWP) weighted GHG budget for the two lakes, a GWP of 28 was used for  $CH_4$  (Myhre et al., 2013).

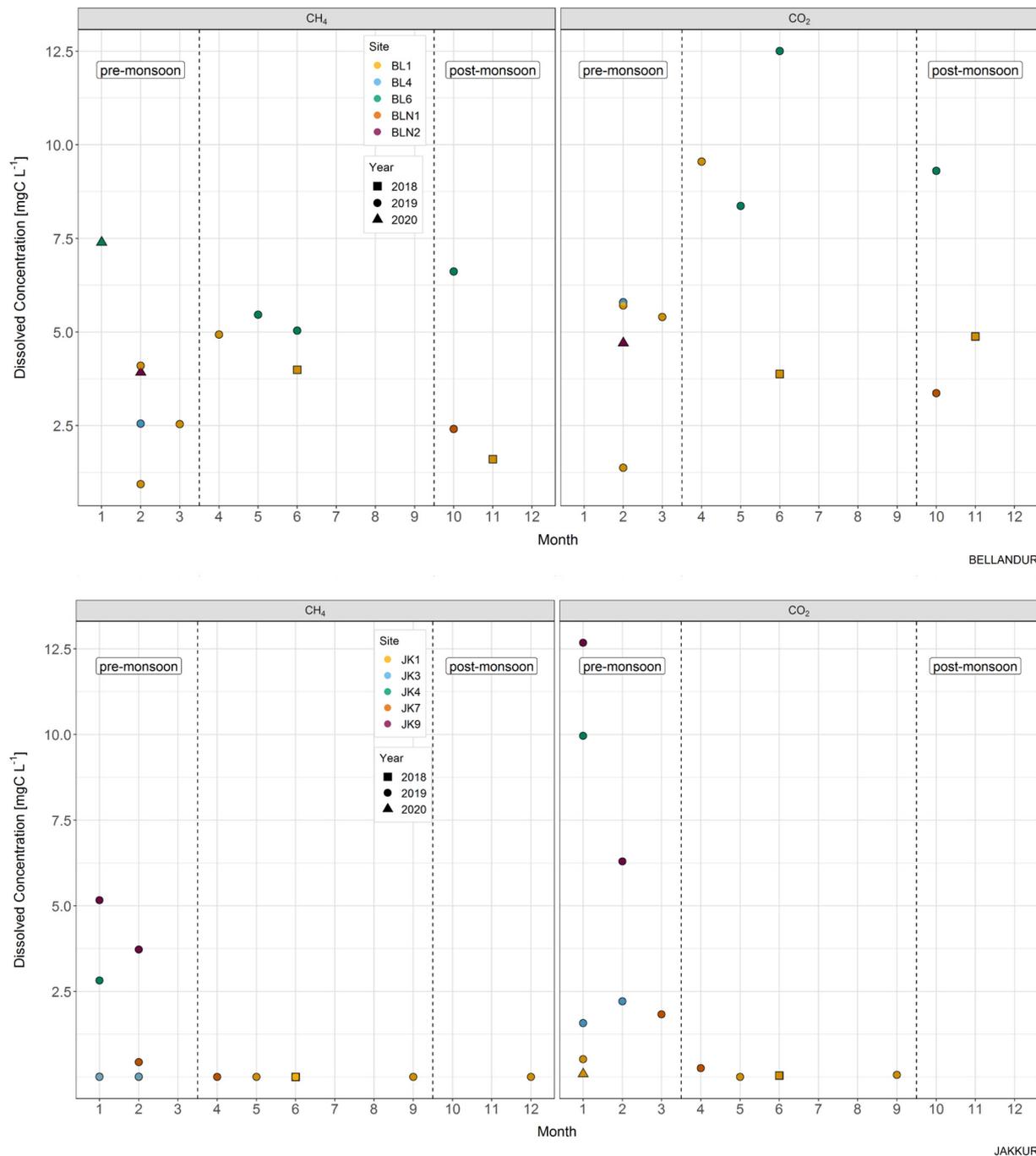


Fig. 3. Time series of dissolved  $CH_4$  and  $CO_2$  concentrations ( $mg\ C\ L^{-1}$ ) at all Bellandur Lake sampling points (top panel) and Jakkur Lake sampling points (bottom panel) between June 2018 and February 2020. Pre and post-monsoon periods are marked on the plots for reference.

### 3. Results

#### 3.1. Concentrations and evasion at all measurement sites

Dissolved CH<sub>4</sub> and CO<sub>2</sub> concentrations at Bellandur Lake were highly variable between sampling points and across sampling occasions (Fig. 3). On average, concentrations of both CH<sub>4</sub> and CO<sub>2</sub> were highest at site BL6 (6.13 ± 1.08 mg CH<sub>4</sub>-C L<sup>-1</sup> and 10.1 ± 2.17 mg CO<sub>2</sub>-C L<sup>-1</sup>), the mid lake sampling point. At Jakkur Lake, concentrations at the mid lake sampling point (JK1) were consistently low, and highest mean concentrations for both CO<sub>2</sub> and CH<sub>4</sub> were observed in lake inlet JK9 (4.44 ± 1.02 mg CH<sub>4</sub>-C L<sup>-1</sup> and 9.49 ± 4.52 mg CO<sub>2</sub>-C L<sup>-1</sup>). Highest dissolved concentrations for CO<sub>2</sub> and CH<sub>4</sub> were measured in January and February at Jakkur Lake, coinciding with the pre-monsoon period, although no clear seasonal patterns are evident otherwise.

Daily evasion fluxes of CH<sub>4</sub> at Bellandur Lake were typically three orders of magnitude larger than at Jakkur, but substantial fluxes were observed at inlets JK9 and JK4 (Fig. 4). The highest mean CH<sub>4</sub> flux (8048 mg m<sup>-2</sup>·d<sup>-1</sup>) occurred in October at site BL6 at Bellandur Lake. Daily evasion fluxes of CO<sub>2</sub> were also highest at site BL6, reaching in excess of 54,000 mg·m<sup>-2</sup>·d<sup>-1</sup> for June. At sites JK9 and JK4 at Jakkur Lake daily evasion fluxes were within the same order of magnitude as for Bellandur Lake. Fluxes were low at sites JK3 and JK7, and very low to negligible at site JK1.

#### 3.2. Lake scale GHG evasion

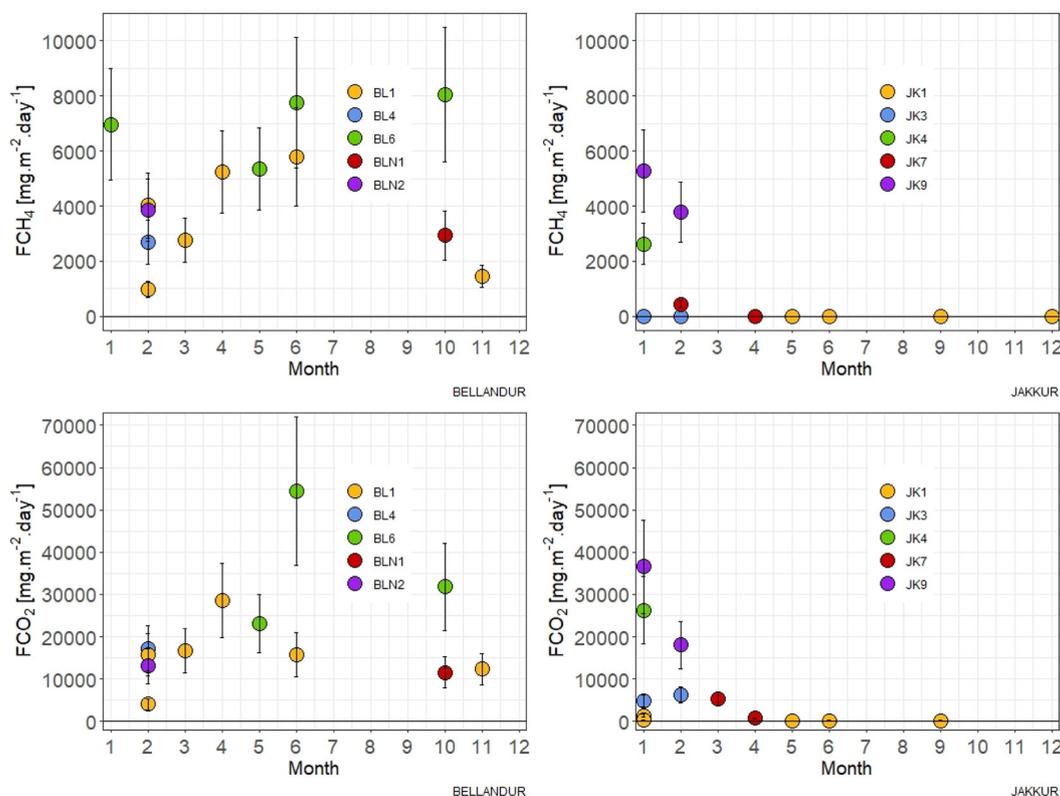
To determine lake-scale GHG evasion potential, data from two sites deemed representative of lake conditions were used. CH<sub>4</sub> concentrations measured at site BL1 ranged from 0.935 to 4.93 mg CH<sub>4</sub>-C L<sup>-1</sup>, with a mean of 3.02 ± 1.57 mg CH<sub>4</sub>-C L<sup>-1</sup> (Table 3). Mean concentrations at BL1 were more than a thousand times higher than mean concentrations measured at JK1, 1.72 ± 1.22 µg CH<sub>4</sub>-C L<sup>-1</sup> (range 0.331 to

**Table 3**

Mean concentrations for CH<sub>4</sub> and CO<sub>2</sub> at all sampling sites. When number of sampling occasions is greater than one, standard deviation and range of concentrations are also provided.

	Dissolved CH <sub>4</sub> -C (mg L)	Dissolved CO <sub>2</sub> -C (mg L)
Bellandur		
BL1	3.02 ± 1.57 (0.93–4.93) n = 6	5.13 ± 2.67 (1.37–9.55) n = 6
BL4	2.55	5.80
BL6	6.12 ± 1.08 (5.04–7.39) n = 4	10.06 (8.37–12.5) n = 3
BLN1	2.41	3.37
BLN2	3.92	4.70
Jakkur		
JK1	0.002 ± 0.001 (0.0–0.003) n = 6	0.14 ± 0.21 (0.03–0.52) n = 5
JK3	0.007 ± 0.005 (0.003–0.01) n = 2	1.89 ± 0.44 (1.57–2.21) n = 2
JK4	2.82	9.96
JK7	0.11 ± 0.21 (0.0–0.44) n = 4	1.04 ± 1.11 (0.26–1.82) n = 2
JK9	4.44 ± 1.02 (3.72–5.16) n = 2	9.48 ± 4.52 (6.29–12.7) n = 2

3.46 µg CH<sub>4</sub>-C L<sup>-1</sup>). Mean CO<sub>2</sub> concentrations at BL1 were also higher than at JK1, 5.13 ± 2.67 mg CO<sub>2</sub>-C L<sup>-1</sup> compared to 0.142 ± 0.212 mg CO<sub>2</sub>-C L<sup>-1</sup>, though CO<sub>2</sub> concentrations at JK1 were particularly variable (Table 3). Concentrations of N<sub>2</sub>O were small (around ambient) or not detectable due to dilution and are hence considered negligible for GHG evasion calculations.



**Fig. 4.** Daily mean evasion fluxes of carbon dioxide and methane estimated from headspace gas samples taken at multiple locations at Bellandur and Jakkur lakes over the period June 2018 to February 2020.

**Table 4**

Lake scale potential evasion for CH<sub>4</sub> and CO<sub>2</sub> from Bellandur Lake (site BL1) and Jakkur Lake (site JK1). Associated standard deviations are reported. Note that GWPs have been applied to determine the total CO<sub>2</sub>e evasion value.

	BL1	JK1
Potential CH <sub>4</sub> -C evasion t yr <sup>-1</sup>	3413 ± 578	0.33 ± 0.06
Potential CO <sub>2</sub> -C evasion t yr <sup>-1</sup>	5711 ± 844	24 ± 10
TOTAL CO <sub>2</sub> e evasion t yr <sup>-1</sup>	148,353 ± 21,799	100 ± 37

The annual evasion estimate for CH<sub>4</sub>-C from Bellandur Lake was 3413 ± 578 ton yr<sup>-1</sup>, four orders of magnitude greater than for Jakkur Lake (0.33 ± 0.06 ton yr<sup>-1</sup>), and for CO<sub>2</sub>-C was two orders of magnitude greater (Table 4). When adjusted for GWPs, potential losses to the atmosphere of 148,353 ± 21,799 ton CO<sub>2</sub>e yr<sup>-1</sup> were estimated for Bellandur Lake, and 100 ± 37 ton CO<sub>2</sub>e yr<sup>-1</sup> for Jakkur Lake.

#### 4. Discussion

Mean surface water concentrations of dissolved CH<sub>4</sub> measured at Bellandur Lake (BL1) were three orders of magnitude higher than the mean reported for Jakkur Lake (Table 3) and considerably higher than the mean concentration of 0.05 mg CH<sub>4</sub>-C L<sup>-1</sup> ± 0.17 mg CH<sub>4</sub>-C L<sup>-1</sup>, reported by Panneer Selvam et al. (2014) in a previous sampling campaign of freshwaters in southern India. Comparisons between these two studies indicate the unprecedented scale of CH<sub>4</sub> concentrations in Bellandur Lake, with outlet concentrations ranging from 0.93 mg CH<sub>4</sub>-C L<sup>-1</sup> to 4.93 mg CH<sub>4</sub>-C L<sup>-1</sup>, resulting in an extremely high evasion potential from this heavily polluted system. In a study of methane in reservoirs in India, dissolved concentrations of up to 1.8 mg CH<sub>4</sub>-C L<sup>-1</sup> were detected, but only at considerable water depth (~40 m) and methane oxidation rendered surface water concentrations significantly lower than observed in this study (Narvenkar et al., 2013). Ebullition was frequently observed during sampling, suggesting that the water was super-saturated with CH<sub>4</sub>. Very low dissolved oxygen (Table 1) likely prevents the destruction of CH<sub>4</sub> by oxidation, rendering evasion to the atmosphere the most plausible end fate.

The CH<sub>4</sub> fluxes from Bellandur Lake were at the high end of emissions from aquatic systems globally (Rosentreter et al., 2021) and other tropical freshwater systems: for example, Gondwe and Masamba (2014) reported maximum CH<sub>4</sub> emissions of 2161 mg·m<sup>-2</sup>·d<sup>-1</sup> measured over open water in the Okavango Delta, Botswana, whilst CH<sub>4</sub> emissions measured in a hydro-electric reservoir in French Guyana were in the range 193–850 mg·m<sup>-2</sup>·d<sup>-1</sup> (Abril et al., 2005). Such high fluxes at Bellandur Lake are consistent with the observed ebullition fluxes from the water surface. In contrast, CH<sub>4</sub> fluxes from Jakkur lake (Fig. 4) were considerably lower than in other tropical or sub-tropical systems (e.g. 35 mg·m<sup>-2</sup>·d<sup>-1</sup> in the Okavango Delta (Gondwe and Masamba, 2014) and 9 mg·m<sup>-2</sup>·d<sup>-1</sup> in coastal marshes in Louisiana, USA (DeLaune and Pezeshki, 2003)).

The substantially lower concentrations and fluxes of CH<sub>4</sub> measured at Jakkur Lake compared to Bellandur Lake are indicative that secondary treatment of wastewater has reduced the OM source to the lake. In this lake, CO<sub>2</sub> was dominant in terms of both dissolved GHG concentrations and total annual CO<sub>2</sub>e evasion (Table 4), in contrast to Bellandur Lake. It should be noted, however, that concentrations of both CO<sub>2</sub> and CH<sub>4</sub> in two polluted inlets to Jakkur Lake (Fig. 3) were of the same order of magnitude as at Bellandur Lake. Whilst high concentrations did not propagate to the mid lake sampling point used for scaling up to potential lake scale GHG evasion, these data provide evidence that hotspots of emissions continue to occur within Jakkur's system. One of the in-lake management measures implemented during our sampling timeframe was the raising of the bund height to seal off inlet JK9 (Fig. 1), which was associated with higher GHG concentrations. Although this measure has a number of other potential benefits, such as improved lake water quality, in terms of GHG release it is likely the

measure has simply pushed evasion further upstream, thus highlighting the importance of determining catchment scale GHG budgets.

The fires on Bellandur Lake, which have received national and international news coverage, could indicate that severely polluted lakes act as massive OM reactors, producing vast quantities of CH<sub>4</sub> that are released to the atmosphere through diffusion (measured here) and most likely at an even greater magnitude via ebullition which is not directly quantified here. In a previous city scale assessment of CH<sub>4</sub> emissions from freshwaters, fluxes via ebullition contributed to ~60% of total emissions (Herrero Ortega et al., 2019), suggesting that our data do not provide a complete CH<sub>4</sub> budget assessment for Bellandur and Jakkur lakes. Although the role of CH<sub>4</sub> in the lake fires has not been demonstrated, the link is highly plausible as our measurements have shown consistent supersaturation accompanied by substantial ebullition. Controlling the OM entering the lake should help to reduce the risk of fires, although existing sources of OM in the lake sediments may need in-situ management measures, such as oxidation, in order to reduce emissions in the short- to medium term.

The headspace sampling approach provided a convenient tool to gauge the magnitude of the GHGs present in the lakes in this scoping study, but we acknowledge the significant uncertainties associated with this measurement method, particularly the need for sample dilution prior to GC analysis which rendered N<sub>2</sub>O undetectable. Uncertainties in the annual greenhouse gas emissions are also generated by the relatively low number of sampling points around each lake and the low sampling frequency. A focus for future work in these highly dynamic lakes should be to include higher resolution measurements to quantify the evasion fluxes from water to atmosphere. Eddy-covariance would be particularly useful because diffusive and convective fluxes would be captured at short time scales (typically half-hourly averaging intervals) and the continuous, automated nature of the measurements would enable the study of seasonal fluctuations, and of the potential effects of remediation on the GHG budgets. In combination with other techniques, including the headspace method employed here, resultant data could also allow for identification of potential GHG hotspots, at the lake scale and beyond.

Our initial results suggest that managing OM inputs to severely polluted lakes could have implications not only for water quality and ecosystem health, but for GHG budgets too. Whilst further data on GHG emissions from restored versus polluted systems is required to corroborate this finding, we propose that lake restoration can facilitate management of GHG emissions, and therefore may present an important climate change mitigation strategy. Recognising that polluted urban waters are a major global source of GHGs is the first step in managing these sources. The recent guidance by IPCC to include artificial waters in national GHG inventories (Lovelock et al., 2019), as many urban lakes and reservoirs are, highlights the need to understand these sources much better and provides political imperative to develop measures to control these sources. This scoping study indicates that greater provision of wastewater treatment, particularly in rapidly growing developing countries, is a relatively practical approach to reduce GHG emissions from inland waters. Controlled CH<sub>4</sub> capture in wastewater treatment, in relation to anaerobic digestion technologies producing biogas, makes this an even more attractive economic prospect. In summary, the management of OM inputs into polluted lakes presents opportunities not only for GHG emissions reductions, but also for biodiversity improvement and economic opportunities via alternative energy sources.

#### CRediT authorship contribution statement

LC, CH, PJ and JD developed the initial idea for the scoping study. PJ and SB coordinated the sample collection and shipment, and provided input on lake chemistry data and management regimes. SW analysed the samples. SW, AP and CH performed the data analysis and AP led the paper writing, with contributions from all authors.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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