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1 **Ecosystem maturity modulates greenhouse gases fluxes from artificial lakes**

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25 **Keywords**

26 Gas fluxes; Ecosystem metabolism; C cycling; Eutrophication; Artificial lakes

27 **Abstract**

28

29 Lentic ecosystems play a major role in the global carbon cycling but the understanding of the  
30 environmental determinants of lake metabolism is still limited, notably in small artificial lakes.  
31 Here the effects of environmental conditions on lake metabolism and CO<sub>2</sub> and CH<sub>4</sub> emissions  
32 were quantified in 11 small artificial gravel pit lakes covering a gradient of ecosystem maturity,  
33 ranging from young oligotrophic to older, hypereutrophic lakes. The diffusive fluxes of CO<sub>2</sub>  
34 and CH<sub>4</sub> ranged from -30.10 to 37.78 mmol m<sup>-2</sup> d<sup>-1</sup> and from 3.05 to 25.45 mmol m<sup>-2</sup> d<sup>-1</sup> across  
35 gravel pit lakes, respectively. Nutrients and chlorophyll *a* concentrations were negatively  
36 correlated with CO<sub>2</sub> concentrations and emissions but positively correlated with CH<sub>4</sub>  
37 concentrations and emissions from lakes. These findings indicate that, as they mature, gravel  
38 pit lakes switch from heterotrophic to autotrophic-based metabolism and hence turn into CO<sub>2</sub>-  
39 sinks. In contrast, the emission of CH<sub>4</sub> increased along the maturity gradient. As a result,  
40 eutrophication occurring during ecosystem maturity increased net emissions in terms of climate  
41 impact (CO<sub>2</sub> equivalent) due to the higher contribution of CH<sub>4</sub> emissions. Overall, mean CO<sub>2</sub>  
42 equivalent emission was 7.9 g m<sup>-2</sup> d<sup>-1</sup>, a value 3.7 and 4.7 times higher than values previously  
43 reported in temperate lakes and reservoirs, respectively. While previous studies reported that  
44 lakes represent emitters of C to the atmosphere, this study highlights that eutrophication may  
45 reverse lake contribution to global C budgets. However, this finding is to be balanced with the  
46 fact that eutrophication also increased CH<sub>4</sub> emissions and hence, enhanced the potential impact  
47 of these ecosystems on climate. Implementing mitigation strategies for maintaining  
48 intermediate levels of maturity is therefore needed to limit the impacts of small artificial  
49 waterbodies on climate. This could be facilitated by their small size and should be planned at  
50 the earliest stages of artificial lake construction.

## 51 1. Introduction

52 Due to the large amounts of terrestrial C that they transport, process and store, inland waters  
53 are an active component of the global carbon (C) cycle thus potentially affect global climate  
54 (Cole et al., 2007; Tranvik et al., 2009). During the last decade, a myriad of studies has focused  
55 on quantifying CO<sub>2</sub> (e.g., Battin et al., 2009; Raymond et al., 2013) and CH<sub>4</sub> (e.g., Bastviken et  
56 al., 2011) emissions from inland waters. In particular, lentic inland ecosystems including lakes  
57 appeared to be globally significant emitters of CO<sub>2</sub> and CH<sub>4</sub> to the atmosphere (e.g., Bastviken  
58 et al., 2011; Deemer et al., 2016; DelSontro et al., 2018; Holgerson and Raymond, 2016)  
59 justifying their inclusion in IPCC guidelines for national and global inventories of greenhouses  
60 gases (GHG) (IPCC, 2019). Nonetheless, current estimates of C emissions from lentic  
61 ecosystems are based on extrapolations of average emission rates to global lake surface area.  
62 These estimates vary considerably, ranging from 0.38 to 0.81 Pg C yr<sup>-1</sup> (e.g., Cole et al., 2007;  
63 DelSontro et al., 2018; Holgerson and Raymond, 2016; Tranvik et al., 2009), highlighting the  
64 existence of large uncertainties for global estimates that was likely driven by a lack of  
65 knowledge of the mechanisms driving emission rate variability (e.g., DelSontro et al., 2018).

66 Eutrophication is a paramount environmental concern worldwide (Carpenter et al.,  
67 1999; Jeppesen et al., 2012), but its role as a driver of the direction and magnitude of GHG flux  
68 from/to lentic ecosystems remains unclear. The role of lakes as sources or sinks of C is highly  
69 dependent on the balance between primary production and ecosystem respiration (e.g., Cole et  
70 al., 2000; Jeppesen et al., 2016; Prairie et al., 2002). Lakes with high loading of organic C tend  
71 to be net heterotrophic ecosystems and act as significant sources of C to the atmosphere through  
72 respiration (e.g., Andersson and Sobek, 2006; Hanson et al., 2003; Laas et al., 2012). However,  
73 the relative importance of organic C load and associated heterotrophic respiration may be offset  
74 by a high CO<sub>2</sub> uptake by primary producers, especially in lakes with high total phosphorus (TP)  
75 concentrations since it is known that TP regulated primary productivity (e.g., Davidson et al.,  
76 2015; Laas et al., 2012; Yang et al., 2019). In some eutrophic lakes, high CO<sub>2</sub> uptake by primary  
77 producers can turn lakes into net autotrophic ecosystems and hence CO<sub>2</sub> sinks (Kosten et al.,  
78 2010; Pacheco et al., 2014; Roland et al., 2010). Yet, this holds for a small proportion of lakes  
79 while most of them are predominant sources of C. This is because the loading of allochthonous  
80 organic C generally greatly exceeds autochthonous primary production (e.g., Bastviken et al.,  
81 2011; Cole et al., 2000, 1994; Deemer et al., 2016). The projected increase in lakes  
82 eutrophication (e.g., Beaulieu et al., 2019) may therefore reverse the global contribution of lakes  
83 to global C budget (Pacheco et al., 2014). While high primary productivity may decrease CO<sub>2</sub>  
84 emissions from lakes, it may act oppositely by promoting CH<sub>4</sub> emissions, providing labile  
85 substrate for methanogenic bacteria and promoting C-rich and oxygen-poor environment (e.g.,  
86 Sepulveda-Jauregui et al., 2018; Vachon et al., 2020; West et al., 2016, 2012; Yang et al., 2019).  
87 This suggests that eutrophication may lead to a decrease in emissions of C from lakes in the  
88 form of CO<sub>2</sub> but, at the same time, to an increase in emissions of C in the form of CH<sub>4</sub>. This is  
89 an important consideration because CH<sub>4</sub> in the atmosphere is up to 34 times more potent as  
90 greenhouse gas (GHG) than CO<sub>2</sub> over a 100-year time scale (IPCC, 2013) and its individual  
91 contribution to total radiative forcing range between 71.8-77.8% when considering aquatic  
92 waterbodies (DelSontro et al., 2018a). This indicates that the effects of eutrophication on CH<sub>4</sub>  
93 emissions can deeply influence lake contribution to global warming.

94 The relationships between eutrophication, CO<sub>2</sub> autotrophy and CH<sub>4</sub> emissions are,  
95 however, highly context-dependant (Deemer et al., 2016; DelSontro et al., 2018a; Ollivier et  
96 al., 2019). For instance, DelSontro et al. (2018) reported that TP enrichment decreased CO<sub>2</sub>  
97 emissions in medium to large-size lakes while it increased emissions in small lakes. In addition,  
98 chlorophyll *a* was positively associated to CH<sub>4</sub> emissions, though the strength of this effect was  
99 stronger for larger ecosystems (DelSontro et al. 2018). This suggests that the extent to which

100 eutrophication influences greenhouse gas emissions may be dependent on lake morphometry  
101 and highlights the high contribution of small lakes to global C emissions from lentic  
102 ecosystems. While they are particularly prone to eutrophication (e.g., Ollivier et al., 2019;  
103 Wetzel, 2001) and extremely abundant worldwide (e.g., Cael et al., 2017; Verpoorter et al.,  
104 2014), small, shallow and artificial lakes remain overlooked compared to natural and large  
105 ecosystems (e.g., Peacock et al., 2019; van Bergen et al., 2019; Webb et al., 2019). For instance,  
106 C fluxes from gravel pit lakes, artificially formed when gravel pits are excavated at or below  
107 the water table (Mollema and Antonellini, 2016), have not been documented yet. Gravel pits  
108 lakes are extremely abundant across the globe (Mollema and Antonellini, 2016), notably  
109 because more than 890 million tons of gravel are produced globally per year (USGS, 2017).  
110 Gravel pit lakes are usually small (1-100 ha), shallow (2-12 m maximal depth) and distributed  
111 in regional clusters experiencing similar climatic conditions. They represent interesting model  
112 ecosystems for disentangling the effects of eutrophication on C emissions in small lentic  
113 ecosystems. In particular, as they age (defined here as the time since the end of excavation),  
114 gravel pit lakes tend to switch from oligotrophic to hypereutrophic waterbodies due to increased  
115 nutrients concentrations, lake productivity and the sedimentation of detritus including  
116 autochthonous material (Kattner et al., 2000; Mollema and Antonellini, 2016; Zhao et al.,  
117 2016). This represents a maturation process (Vitousek and Reiners, 1975) that may be promoted  
118 in gravel pit lakes by their disconnection to the rest of the river network and the importance of  
119 terrestrial inputs in their functioning (Alp et al. 2016). For instance, Zhao et al. (2016) reported  
120 a large range of lake productivity (e.g. chlorophyll-a concentration ranging from 0.9 to 58.8 mg  
121 l<sup>-1</sup>) in gravel pit lakes. Maturation of these artificial ecosystems is multifaceted process that  
122 includes an increased amount of nutrient in the systems (eutrophication), a change in animal  
123 communities, an increased ripisylve along the shore as trees grow and a higher intensity of  
124 human activities as ecosystems become older (Zhao et al. 2016). Because of their high  
125 environmental heterogeneity, gravel pit lakes represent a unique model system to examine the  
126 impact of eutrophication on C cycling, while controlling other factors such as climate variability  
127 given their very restricted geographical distribution.

128 In the present study, the effects of eutrophication on gas metabolism and emissions were  
129 quantified in 11 gravel pit lakes across a large range of ecosystem maturity to identify the  
130 environmental factors driving lakes as C-source or sink. In particular, gravel pit lakes were  
131 predicted to shift from net CO<sub>2</sub> sources to net CO<sub>2</sub> sinks as they mature, mainly due to the  
132 increased CO<sub>2</sub> uptake by primary producers. Nonetheless, increased CH<sub>4</sub> emissions as primary  
133 production and nutrient contents increased were predicted. The impacts of gas fluxes from  
134 gravel pit lakes on climate were also estimated and were predicted to be dependent on the  
135 balance between CO<sub>2</sub> and CH<sub>4</sub> fluxes and hence, between heterotrophic benthic and the  
136 autotrophic water layer metabolism. In addition, and to provide a broader perspective to these  
137 results, the values of gas fluxes measured in the present study were compared to values reported  
138 in the literature in other temperate lentic ecosystems.

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142 **2. Materials and Methods**

143 2.1. Study sites

144 The study area was located in the Garonne floodplain (south-west of Toulouse, France) that  
145 contains more than 1522 ha of gravel pit lakes (Saplaïroles et al., 2007). The study was  
146 conducted in an area with a large quantity of gravel pit lakes with varying environmental  
147 conditions (Alp et al., 2016; Jackson et al., 2017; Zhao et al., 2016). From mid-September to  
148 mid-October 2017, 11 gravel pit lakes were selected to cover a gradient of ecosystem maturity  
149 based on their age that ranged from 12 to 55 years (Fig.1). Several abiotic and biotic  
150 environmental variables were assessed once at three locations within each lake (Table 1).  
151 Specifically, a portable multiparameter probe (EXO2, YSI Incorporated, Ohio, USA) was used  
152 to determine the depth profiles of temperature, pH, turbidity and conductivity. Nine important  
153 physico-chemical variables including variables related to eutrophication (e.g. total phosphorus,  
154 total nitrogen, ammonium, phosphate) were measured according to national standards (NF EN  
155 ISO 10304-1, 1339; AFNOR NF EN ISO 14911) with water sampled at 1 m depth. Sediments  
156 were sampled with an Ekman dredge, sieved, homogenized, and stored in a cooler. Grain size  
157 distribution, ash free dry mass and concentrations in organic carbon, TN, TP were subsequently  
158 measured (LDA26, Valence, France). Water clarity (Secchi depth) was measured using a  
159 Secchi-disk. Chlorophyll *a* (chl<sub>a</sub>) and Cyanobacteria concentrations were measured directly in  
160 the surface water (0.5 m depth) *via* fluorescence using a portable fluorescence photometer  
161 (AlgaeTorch, BBE-Moldaenke, Kiel, Germany). Specifically, the AlgaeTorch measures the  
162 fluorescence of algae cells in the water by exciting the algae pigments by 6 coloured LEDs of  
163 different wavelengths (i.e., 470, 525 and 610 nm) to obtain a fluorescence spectrum. In parallel,  
164 turbidity was determined by measuring the reflection at a wavelength of 700 nm which allows  
165 simultaneous turbidity correction during chlorophyll measurement. A standardised zooplankton  
166 sample was collected at the water surface in all lakes using a 250 µm mesh net. Samples were  
167 sieved and fixed in 100 mL 70 % ethanol. In the laboratory, a 10 mL sub-samples was taken  
168 and debris removed from the sample that was oven-dried (60°C for 72 h) and weighted to the  
169 nearest mg to estimate zooplankton density (mg.L<sup>-1</sup>). The average value of each variable across  
170 the three replicates in each lake was used for subsequent analyses. Additionally, aerial pictures  
171 and geographic information system (GIS) analyses were used to calculate the area, the perimeter  
172 and the percentage of area covered by riparian vegetation (10-metre buffer around each lake),  
173 urbanization and other gravel pit lake dredging (5-km buffer) (Zhao et al., 2016). Lake depth  
174 and volume were determined using bathymetry analyses.

175 2.2. Ecosystem maturity gradient

176 The lakes covered a large range of environmental conditions associated to ecosystem maturity  
177 (Table 1), including productivity (e.g., chl<sub>a</sub> ranging from 2.5 to 48.2 µg L<sup>-1</sup>) and water nutrient  
178 content (e.g., TP ranging from 28.8 to 115.9 µg L<sup>-1</sup>). The lakes were distributed along a gradient  
179 of ecosystem maturity defined using a principal component analyses (PCA) based on  
180 environmental conditions in the lakes. The first two dimensions of the PCA accounted for  
181 74.2% of the total inertia (Fig.2). The main variables positively correlated with PC1 (50.8%)  
182 were the concentrations of chl<sub>a</sub>, of nutrients in water and in sediments, lake age and the level  
183 of urbanization (see details in Supplementary material, Table A1). The content of mineral  
184 matter in sediment, the Secchi depth and the maximal depth were negatively correlated with  
185 PC1. Therefore, PC1 axis was used to represent the multifaceted characteristics of ecosystem  
186 maturity and the distribution of studied lakes along a maturity gradient based on their rank from  
187 the youngest unproductive lake (rank 1; SAB) to the oldest highly productive lake (rank 11,  
188 BVI). The main variables positively correlated with PC2 (23.4%) were related to lake

189 hydromorphology including perimeter, surface and shoreline development. The second  
190 component discriminated smaller and circular lakes (e.g., LIN, BVI) from larger lakes with  
191 more complex shape (e.g. LAM, LAV).

## 192 2.3. Gas concentrations and fluxes

### 194 2.3.1. Dissolved gas concentrations and metabolic balances

195 Depth profiles of dissolved methane (CH<sub>4</sub>), dioxide carbon (CO<sub>2</sub>) and oxygen (O<sub>2</sub>)  
196 concentrations were measured once at three locations within lakes (i.e., at littoral area, center  
197 and deepest area) for capturing the heterogeneity of lakes. In particular, CH<sub>4</sub> and CO<sub>2</sub>  
198 concentrations were measured using submersible gas sensors (Mini CH<sub>4</sub><sup>TM</sup>, Mini CO<sub>2</sub><sup>TM</sup>,  
199 ProOceanus, Nova Scotia, Canada) operating through diffusion of dissolved gases from liquids  
200 through a supported semi-permeable membrane to a non-dispersive infrared detector (NDIR).  
201 Dissolved O<sub>2</sub> concentrations were measured using a portable multiparameter probe (EXO2, YSI  
202 Incorporated, Ohio, USA). Dissolved gas concentrations were measured during 15 minutes  
203 every 0.5 m depth. In addition, temperature, pressure, and humidity of the internal gas were  
204 determined to correct the gas concentration measurement. The solubility coefficients of Weiss  
205 (1974) and Yamamoto et al. (1976) were then used to calculate the dissolved concentrations of  
206 CO<sub>2</sub> and CH<sub>4</sub>, respectively. Because there was no significant variation on gas concentrations  
207 across depth (*P*-value > 0.05), the entire profiles were used to calculate the water column gas  
208 concentration (average of 0.5-m interval profiles values) for each replicate. The mean surface  
209 concentrations were then expressed as a concentration differential ( $\Delta$ gas in  $\mu$ M) from elevation  
210 and temperature corrected saturation values (named hereafter ‘departures from saturation’). Gas  
211 saturation was calculated using atmospheric concentrations of 407.8 ppm for CO<sub>2</sub> and 1869 ppb  
212 for CH<sub>4</sub> (Source: WMO Global Atmosphere Watch). The average value of dissolved gas  
213 concentrations and metabolic balance using the three replicate in each lake was used for  
214 subsequent analyses

215

216

### 217 2.3.2. Atmospheric gas emissions

218 Atmospheric gas emissions were determined for CO<sub>2</sub> and CH<sub>4</sub>. CO<sub>2</sub> flux across air-water  
219 interface were estimated using two floating chambers (FC; volume: 47 L; area: 0.24 m<sup>2</sup>; water  
220 penetration: 6 cm). The FC were covered with reflective alumina tape to minimize internal  
221 heating. The FC were equipped with a vent on the top to equalize the air pressure in the chamber  
222 with the atmospheric pressure before starting the measurements and with a floating structure  
223 (PVC tube  $\varnothing$  90 cm). CO<sub>2</sub> concentrations within the FC were measured continuously using a  
224 non-dispersive infrared (NDIR) spectroscopy logger (ELG CO<sub>2</sub>, SenseAir, Delsbo, Sweden)  
225 (details in Bastviken et al. (2015)). For each lake, the two FC were deployed three times from  
226 the center of lake. The FC were left drifting during measurement to avoid creation of artificial  
227 turbulence. Each deployment was run over about 15 minutes after an equilibration period with  
228 opened vent for 5 minutes. Before starting new deployment, the chamber was lifted, vented for  
229 5 min, and then replaced on the water. Diffusive CO<sub>2</sub> flux at the air-water interface (mmol m<sup>-2</sup>  
230 day<sup>-1</sup>) were calculated according to:

$$231 F_{CO_2} = S \cdot t \cdot V_{FC} (V_m \times A) \quad (1)$$

232 Where *S* is the slope of the linear regression of gas concentration in the chamber *versus* time  
233 (ppm.min<sup>-1</sup>), *t* is a conversion factor of minutes into days (1440 min d<sup>-1</sup>), *V<sub>FC</sub>* is the volume of  
234 the chamber, *V<sub>m</sub>* is the gas molar volume at ambient temperature and pressure and *A* is the area

235 of the chamber. Fluxes derived from chamber deployments were kept only when the linear  
 236 regression exhibited  $R^2 > 0.8$  and a significant slope ( $P$ -value  $< 0.05$ ).  
 237 Then, gas exchange coefficients for  $\text{CO}_2$  ( $k_{\text{CO}_2}$ ) expressed in  $\text{m d}^{-1}$  were derived from  
 238 concomitant measurements of gas flux and partial pressure in air ( $p\text{CO}_{2a}$ ) and in surface water  
 239 ( $p\text{CO}_{2w}$ ) expressed in atmosphere following:

$$240 \quad k_{\text{CO}_2} = \frac{F_{\text{CO}_2}}{K_o \cdot (p\text{CO}_{2w} - p\text{CO}_{2a})} \quad (2)$$

241 where  $K_o$  is the solubility of  $\text{CO}_2$  in water expressed in  $\text{mole l}^{-1} \text{atm}^{-1}$  at surface water  
 242 temperature of given lake (Weiss, 1974).

243 The  $k_{\text{CO}_2}$  was normalized to a Schmidt number of 600 at  $20^\circ\text{C}$  according to Jähne et al. (1987):

$$244 \quad k_{600 \text{ CO}_2} = k_{\text{CO}_2} \left( \frac{600}{Sc_{\text{CO}_2, T}} \right)^{-n} \quad (3)$$

245 Where  $Sc_{\text{CO}_2, T}$  is the Schmidt number for  $\text{CO}_2$  at a given temperature.  $N=2/3$  was used for wind  
 246 speed at  $10 \text{ m} < 3.7 \text{ m s}^{-1}$  (Jähne et al., 1987).

247 Then,  $k_{600 \text{ CO}_2}$  in a given lake was used to extrapolate the gas exchange coefficient for  $\text{CH}_4$  as  
 248 follows:

$$249 \quad k_{\text{CH}_4} = \frac{k_{600 \text{ CO}_2}}{(600 / Sc_{\text{CH}_4, T})^{-n}} \quad (4)$$

250 The diffusive flux of  $\text{CH}_4$  at the air-water interface was then computed following the equation  
 251 proposed by Cole and Caraco (1998):

$$252 \quad F_{\text{CH}_4} = k_{\text{CH}_4} (C_w - C_{eq}) \quad (5)$$

253 Where  $F_{\text{CH}_4}$  is the flux at the air-water interface for  $\text{CH}_4$  ( $\text{mmol m}^{-2} \text{day}^{-1}$ ),  $k_{\text{CH}_4}$  is the gas  
 254 exchange coefficient ( $\text{m day}^{-1}$ ),  $C_w$  is the concentration of  $\text{CH}_4$  in water ( $\text{mmol m}^{-3}$ ), and  $C_{eq}$  is  
 255 the theoretical concentration of  $\text{CH}_4$  in water if the water phase was in equilibrium with the  
 256 atmosphere ( $\text{mmol m}^{-3}$ ).

257 To integrate the contributions of both  $\text{CO}_2$  and  $\text{CH}_4$  fluxes in total GHG emissions from the 11  
 258 gravel pit lakes, the emission terms were expressed in C and  $\text{CO}_2$  equivalent ( $\text{CO}_{2eq}$ ). For this  
 259 purpose, diffusive fluxes were previously converted into  $\text{g m}^{-2} \text{d}^{-1}$  and a global warming  
 260 potential for  $\text{CH}_4$  of 34 was taken assuming a period of 100 years and no carbon feedback loop  
 261 (IPCC, 2013).

### 262 2.3.3. Benthic gas fluxes

263 An optically clear acrylic benthic chamber mounted on a stainless steel frame was used for  
 264 estimating benthic fluxes of  $\text{O}_2$ ,  $\text{CO}_2$  and  $\text{CH}_4$ . The chamber was configured to have 10 cm of  
 265 chamber walls buried inside the sediment. The chamber was equipped at the top with a data  
 266 logger of dissolved oxygen concentration and temperature (hobo U26, onset), a stirrer, three  
 267 non-return check valves and a PVC tubing (6 mm outer diameter, 4 mm inner diameter) inserted  
 268 through cable glands. The stirrer (5-7 rpm) was mounted at approximately 15 cm above the  
 269 sediment and was used for mixing chamber water during the deployment. The two non-return  
 270 valves placed at the corners of the chamber allowed the water to drain into the chamber during  
 271 the immersion. The small non-return valve in the center allowed water to enter inside the  
 272 chamber during water sampling. Deployments usually started between 08:30 and 09:30 AM by  
 273 slowly lowering the chamber to the sediment on a rope tethered to a float from a small boat.  
 274 The chamber was placed at approximately 4 m from the shoreline. During the incubation,



275 approx. 1 m of water remained over the chamber. The incubation period was 5-6 h during which  
276 oxygen concentrations and temperature were recorded every 5 minutes and water was sampled  
277 every 20 minutes. Water samples were collected using 50 mL syringes with three-way  
278 stopcocks connected to the end of the PVC tubing equipped with a luer-lock syringe valve. 15  
279 mL of water was injected into pre-evacuated 21 mL-serum bottles previously filled with 6 g of  
280 NaCl (final saturation concentration) to inhibit microbial activity before analysis (Bastviken et  
281 al., 2011). Then, a headspace was created at the lab, and CO<sub>2</sub> and CH<sub>4</sub> concentrations in the  
282 headspace were measured by GC-MS (gas chromatograph CompactGC, GASTM, Global  
283 Analyser Solutions equipped with GS-Carbon plot column (Agilent), coupled to mass  
284 spectrometer detector in ionisation mode ISQ, THERMO) after desorption (simultaneous  
285 autosampler-desorber TriPlus 300, Thermo). The benthic flux ( $F_{sed}$ ) for each gas was then  
286 calculated as follows:

$$287 \quad F_{sed,gas} = \frac{S \times V_{BC} \times t}{A_{BC}} \quad (6)$$

288 Where  $S$  is the slope of the linear regression of dissolved gas concentration in the benthic  
289 chamber *versus* time ( $\mu\text{mol m}^{-3} \text{h}^{-1}$ ),  $t$  is a conversion factor of hours into days ( $24 \text{ h d}^{-1}$ ),  $V_{BC}$   
290 is the volume of water in the benthic chamber ( $0.0405 \text{ m}^3$ ) and  $A_{BC}$  is the surface area of the  
291 sediment enclosed in the chamber ( $0.2025 \text{ m}^2$ ). Fluxes derived from chamber deployments were  
292 kept only for linear regression with  $R^2 > 0.8$  and a significant slope.

#### 293 2.4. Comparisons with existing emission data

294 A literature survey of studies quantifying CO<sub>2</sub> and CH<sub>4</sub> diffusive emissions from temperate  
295 lentic ecosystems was performed. A total of 31 references (see details in Supplementary  
296 Material Table A2) were collected and a total of 1026 emissions estimates obtained. Only  
297 summer-autumn measurements were considered to allow comparison with the present study.  
298 For some references, values provided were expressed in CH<sub>4</sub>-C or CO<sub>2</sub>-C. These values were  
299 converted using 1.34 and 3.66 conversion factors, respectively. All CO<sub>2</sub>-equivalent values  
300 (expressed in  $\text{g CO}_2 \text{ m}^{-2} \text{ d}^{-1}$ ) presented in this study were calculated using the global warming  
301 potentials (GWPs) of CH<sub>4</sub> at the 100 year ( $1 \text{ g CH}_4 = 34 \text{ g CO}_2$ ) time-scales (IPCC, 2013).

#### 302 2.5. Statistical analyses

303 The relationships between dissolved gas concentrations, gas emissions and environmental  
304 gradients defined according the coordinates of sites on the PCA axes were examined using  
305 Spearman's rank correlation coefficient. Partial least-squares (PLS) regressions (Abdi, 2003)  
306 were then performed to identify environmental variables significantly associated to gas  
307 concentrations and emissions. Cross-validation was used for selecting the optimal number of  
308 components that minimize the prediction errors (RMSE). The variable importance in projection  
309 (VIP) coefficients were calculated to classify the predictors according to their explanatory  
310 power of the dependent variable. Predictors with VIP larger than 1 are the most relevant for  
311 explaining the dependent variable (Eriksson et al., 1999; Tenenhaus, 1998). Then, specific  
312 relationships between gas concentrations and environmental variables were examined using  
313 linear models. For all parametric analyses, normal distribution and homoscedasticity were  
314 checked. All statistical analyses were performed using R software (R Development Core Team,  
315 2008) using FactoMineR (Lê et al., 2008) and PLS (Mevik and Wehrens, 2007) packages.

316

## 317 **Results**

### 318 2.6. Metabolic balance and gas emissions

319 Dissolved gas concentrations in the water column varied moderately among the studied lakes,  
320 ranging from 259.7 to 403.1  $\mu\text{M}$  for  $\text{O}_2$ , from 5.2 and 39.9  $\mu\text{M}$  for  $\text{CO}_2$  and from 1.97 to 4.42  
321  $\mu\text{M}$  for  $\text{CH}_4$  (Table 2).  $\text{CH}_4:\text{CO}_2$  molar ratio ranged from 0.07 to 0.68 among lakes (mean  $\pm$   
322 SD:  $0.27 \pm 0.18$ ) suggesting predominance of aerobic respiration in the water column and/or  
323 strong methanotrophic activities. Five mature lakes were oversaturated with  $\text{O}_2$  and  
324 undersaturated with  $\text{CO}_2$  suggesting an epilimnetic net autotrophy (Fig. 3A). The least mature  
325 lake was undersaturated with  $\text{O}_2$  and oversaturated with  $\text{CO}_2$  suggesting net heterotrophy of  
326 water column (Fig. 3A). The other lakes were oversaturated with both  $\text{O}_2$  and  $\text{CO}_2$ . The five  
327 lakes exhibiting net autotrophy of water column were  $\text{CO}_2$  sinks (Fig. 3B). The others lakes  
328 were  $\text{CO}_2$  sources. All lakes were  $\text{CH}_4$  sources with a relatively broad range of saturation levels  
329 ( $2.0 - 4.8 \mu\text{M}$ ; Fig. 3B). The least mature lake showed highest departure from saturation for  
330  $\text{CO}_2$  but lowest for  $\text{CH}_4$ .  $\text{CO}_2$  and  $\text{CH}_4$  emission fluxes ranged from  $-30.10$  to  $37.78 \text{ mmol m}^{-2}$   
331  $\text{d}^{-1}$  and from  $3.05$  to  $25.45 \text{ mmol m}^{-2} \text{ d}^{-1}$ , respectively (Table 2). Except for one lake, the fluxes  
332 mirrored the patterns obtained by the departure from saturation (Fig. 3). Five lakes were sinks  
333 of  $\text{CO}_2$  but sources of  $\text{CH}_4$ . The six other lakes were sources of both  $\text{CO}_2$  and  $\text{CH}_4$ . Total net  
334 emissions ranged from  $-0.21$  to  $0.60 \text{ g C m}^{-2} \text{ d}^{-1}$  (Fig.4) and from  $2.6$  to  $12.8 \text{ g CO}_{2\text{eq}} \text{ m}^{-2} \text{ d}^{-1}$ .  
335 The net emissions fluxes were negatively correlated with the gradient of ecosystem maturity  
336 when expressed in C equivalents ( $\rho = -0.85$ ,  $P = 0.002$ ), but positively correlated when  
337 expressed in  $\text{CO}_2$ -equivalents ( $\rho = 0.61$ ,  $P = 0.052$ ).

### 338 2.7. Coupling benthic-pelagic metabolism

339 Benthic gas fluxes were successfully computed on six and eight lakes for  $\text{O}_2$  and  $\text{CO}_2$ ,  
340 respectively (Table 2). They ranged from  $-93.8$  to  $26.4 \text{ mmol m}^{-2} \text{ d}^{-1}$  for  $\text{O}_2$  and from  $-54.4$  to  
341  $228.0 \text{ mmol m}^{-2} \text{ d}^{-1}$  for  $\text{CO}_2$ . Except for two lakes that exhibited benthic primary production  
342 (BVI, SOA), most of lakes exhibited benthic fluxes suggesting sediment respiration ( $\text{O}_2$   
343 consumption,  $\text{CO}_2$  release).  $\text{CH}_4$  fluxes from sediment were successfully computed only for  
344 two lakes exhibiting methanogenesis (BIR:  $140.8 \text{ mmol m}^{-2} \text{ d}^{-1}$  and LAV  $178.7 \text{ mmol m}^{-2} \text{ d}^{-1}$ ),  
345 while in the other lakes models were not significant or  $R^2 < 0.80$ . Six lakes showed benthic-  
346 pelagic metabolism coupling, i.e. atmospheric emissions of  $\text{CO}_2$  by lakes exhibiting sediment  
347 respiration and conversely influx of  $\text{CO}_2$  at the air-water interface by lakes exhibiting benthic  
348 primary production. Uncoupling between benthic and surface metabolism was observed in two  
349 lakes exhibiting a net autotrophy of the water column.  $\sim 15\%$  of  $\text{CO}_2$  and only  $\sim 7\%$  of  $\text{CH}_4$   
350 produced in sediments were emitted to the atmosphere.

### 351 2.8. Environmental drivers of gas concentrations and emission

352  $\text{CO}_2$  concentrations and emissions decreased significantly along the gradient of ecosystem  
353 maturity ( $\rho = -0.83$ ,  $P = 0.003$ ;  $\rho = -0.74$ ,  $P = 0.013$ ; respectively) whereas  $\text{CH}_4$  emissions  
354 and the  $\text{CH}_4:\text{CO}_2$  molar ratio significantly increased ( $\rho = 0.73$ ,  $P = 0.01$ ;  $\rho = 0.78$ ;  $P =$   
355  $0.007$ ; respectively). Variability in  $\text{CO}_2$  and  $\text{CH}_4$  concentrations and fluxes were overall well  
356 explained by the model (Table 3,  $R^2$  of 0.56 for  $\text{CO}_2$ , 0.59 for  $\text{CH}_4$ , 0.61 for  $F_{\text{CO}_2}$ , 0.87 for  $F_{\text{CH}_4}$ ).  
357 Atmospheric gas emissions had overall the same explanatory variables than dissolved gas  
358 concentration. The most influential variables ( $\text{VIP} > 1.0$ ) for  $\text{CO}_2$  concentrations and emissions  
359 were *chl a* and DOC concentrations, lake depth, nutrient concentrations in the water (TP, TN),  
360 urbanization level and TOC contents in sediments (Fig. 5). In particular,  $\text{CO}_2$  concentrations  
361 and emissions decreased with increased values of *chl a*, nutrients, DOC and TOC concentrations  
362 and urbanization. Conversely, deeper lakes exhibited higher  $\text{CO}_2$  concentrations and emissions.  
363 Additionally, positive relationship between  $\text{CO}_2$  concentrations in the water column and  $\text{CO}_2$   
364 fluxes from sediments was detected.  $\text{CH}_4$  concentrations and emissions exhibited opposite  
365 relationships with environmental parameters than those described for  $\text{CO}_2$  concentrations and  
366 emissions. In particular,  $\text{CH}_4$  concentrations and emissions increased with nutrient and TOC

367 contents in sediments and urbanization but decreased with lake depth (Fig. 5). Additionally,  
368 atmospheric CH<sub>4</sub> emissions increased with the concentrations in chl<sub>a</sub>.

#### 369 2.9. Comparison of emissions with other temperate lentic ecosystems

370 A comparison with values reported in the literature about GHG emissions in other freshwater  
371 (natural and artificial) ecosystems in temperate climate was performed (see details in  
372 Supplementary material, Table A2). This comparison revealed that the studied gravel pit lakes  
373 generally emitted more CO<sub>2</sub>-equivalent (supported by higher CH<sub>4</sub> emission contribution) than  
374 natural ecosystems such as lakes and ponds. In particular, CO<sub>2</sub>-equivalent emissions from the  
375 studied gravel pit lakes were about 3.7 times and 1.5 times higher than emissions from natural  
376 lakes and ponds belonging to the same climatic zone (Fig. 6), respectively. CH<sub>4</sub> and CO<sub>2</sub>-  
377 equivalent emissions from gravel pit lakes were overall similar to emissions reported for other  
378 temperate small artificial-waterbodies such as urban and farm ponds (CH<sub>4</sub>: 195 mg m<sup>-2</sup> d<sup>-1</sup>;  
379 CO<sub>2</sub>eq: 7.8 g m<sup>-2</sup> d<sup>-1</sup>, in average).

### 380 3. Discussion

381

#### 382 3.1. Switching from heterotrophy to autotrophy when increases ecosystem maturity

383 Eutrophication of aquatic ecosystems is key facet of future ecosystem changes (Beaulieu et al.,  
384 2019) and its consequences on C cycling in aquatic ecosystems remains unclear, and this is  
385 particularly true for small, artificial lakes. In the present study, lake productivity (i.e.  
386 eutrophication) increased quickly after artificial lake construction, with some lakes being  
387 hypereutrophic after 26 years (Carlson, 1977; Wetzel, 2001). This gradient of ecosystem  
388 maturity was accelerated by human activities as suggested by the positive correlation between  
389 variables related to lake productivity (e.g., TP concentrations), lake age and urbanization level.  
390 Consistently with previous studies, increased chlorophyll *a* concentration (likely due to  
391 increased nutrient loadings) was negatively correlated with CO<sub>2</sub> concentrations and emissions  
392 from lakes (e.g., Li et al., 2015; Pacheco et al., 2014). As they mature, lakes shifted from  
393 heterotrophic to autotrophic-based metabolism due to the CO<sub>2</sub> uptake by primary producers. As  
394 they turned into a CO<sub>2</sub> sink, more C buried in lake sediments, as suggested by the negative  
395 relationships between the total organic C contents in sediments and both CO<sub>2</sub> concentrations  
396 and efflux. As observed previously, eutrophication may reverse lake contribution to global C  
397 budget (Pacheco et al., 2014), in particular in the small ecosystems. Nonetheless, this finding  
398 contrasts with some studies reporting that small lakes globally strong emitters of CO<sub>2</sub> (e.g.,  
399 Holgerson and Raymond, 2016), especially when eutrophic (DeSontro et al., 2018a; Ollivier  
400 et al., 2019). The extent to which eutrophication affects the direction and magnitude of CO<sub>2</sub>  
401 emissions from lakes is likely dependent on the balance between the rates of primary  
402 productivity and the input of terrestrial C, substrate of heterotrophic respiration. Here, the young  
403 gravel pit lakes exhibited high CO<sub>2</sub> concentrations and emissions probably because, even in  
404 small quantities (average DOC concentrations < 3 mg L<sup>-1</sup>), allochthonous C subsidies exceeded  
405 autochthonous C-CO<sub>2</sub> consumption limited by low nutrient concentrations (as already  
406 described for many oligotrophic lakes, e.g., Biddanda et al., 2001; Del Giorgio et al., 1997;  
407 Depew et al., 2006). Furthermore, a coupling between sediment respiration and CO<sub>2</sub>  
408 concentrations in water column of young lakes was observed, likely promoted by the shallow  
409 depth of the lakes. This suggests that sediment respiration may be a major driver of CO<sub>2</sub>  
410 oversaturation in young gravel pit lakes (Kortelainen et al., 2006). Conversely, eutrophication  
411 led to an uncoupling between benthic heterotrophic metabolism and water column autotrophy  
412 suggesting that CO<sub>2</sub> uptake by primary producers offset the CO<sub>2</sub> efflux from sediments. The  
413 overweight of primary producers' CO<sub>2</sub> uptake over ecosystem respiration is likely favored by  
414 low inputs of terrestrial materials to gravel pit lakes. Indeed, even if DOC concentrations  
415 increased in concert with primary productivity along the maturity gradient, they remained  
416 overall low among gravel pit lakes (< 6 mg L<sup>-1</sup>, except for one lake, LAM, with DOC = 17.5  
417 mg L<sup>-1</sup>). Several studies reported that switching from net autotrophy to net heterotrophy  
418 occurred at DOC concentrations higher than 6-10 mg L<sup>-1</sup> (e.g., Andersson and Sobek, 2006;  
419 Hanson et al., 2003; Prairie et al., 2002). Consequently, it is likely that here, increase in DOC  
420 concentrations was not enough for maintaining heterotrophy along the maturity gradient.

#### 421 3.2. Eutrophication increases CH<sub>4</sub> emissions

422 In contrast with CO<sub>2</sub> fluxes, atmospheric CH<sub>4</sub> fluxes exhibited positive relationships with  
423 nutrients (TP, TN) and organic material contents in sediments, and DOC and chlorophyll *a*  
424 concentrations in water, suggesting that eutrophication promoted CH<sub>4</sub> emissions. Conversely,  
425 CH<sub>4</sub> fluxes were negatively correlated with lake depth. This could reflect that higher depth  
426 leads to higher potential oxidation capacity of CH<sub>4</sub> produced in the sediments before reaching  
427 the atmosphere (e.g., Bastviken et al., 2004). Yet, this could also be explained by the

428 accumulation of organic matter in the water and in the sediments through sinking, resulting in  
429 a reduced water depth, along the ecosystem maturation (e.g., Staehr et al., 2010). In particular,  
430 highly productive autotrophic systems produce substantial organic detritus since about 15-35%  
431 of dead phytoplankton settles to sediments (Baines and Pace, 1994). This accumulation of easily  
432 degradable phytoplankton-derived organic substrate (e.g., West et al., 2016, 2012) tends to  
433 promote C-rich and oxygen-depleted environment favouring CH<sub>4</sub> production (e.g., Bastviken  
434 et al., 2004; DelSontro et al., 2018; Borrel et al., 2011; Sepulveda-Jauregui et al., 2018; West et  
435 al., 2012). Additionally, a positive correlation between the CH<sub>4</sub> concentration and nutrient  
436 contents (notably TP) in sediments of gravel pit lakes was found. The link between CH<sub>4</sub>  
437 production and nutrient content in sediments has been paid less attention. Here, the positive  
438 relationship could merely reflect enhanced primary production in pelagic waters, since TP  
439 content in sediments increased with lake productivity. Enhanced TP content in sediments may  
440 in turn fuel methanogenesis of accumulated organic C by methanogenic archae (e.g., Adhya et  
441 al., 1998; Alphenaar et al., 1993). CH<sub>4</sub> benthic fluxes could not be successfully measured in most  
442 lakes, even those exhibiting high CH<sub>4</sub> concentrations in the water column. That is likely due to  
443 methodological biases associated with the benthic chambers (e.g., short incubation time, CH<sub>4</sub>  
444 oxidation occurring at the sediment water-interface inside the benthic chamber). Nonetheless  
445 and consistently with several studies, most of the CH<sub>4</sub> produced in sediments (~93% based on  
446 the two lakes with successful measurement of benthic methanogenesis) was oxidized before  
447 reaching the atmosphere (e.g., Bastviken et al., 2003; Yang et al., 2019). This oxidation was  
448 likely promoted by well-oxygenated water of the gravel pit lakes. Despite this, the small part  
449 of CH<sub>4</sub> escaping oxidation outweighed the primary producers' CO<sub>2</sub> uptake along the maturation  
450 gradient. This stronger contribution of CH<sub>4</sub> emissions is particularly since CH<sub>4</sub> has a global  
451 warming potential (GWP) approximately 34 times higher than that of CO<sub>2</sub> (IPCC, 2013). As a  
452 result, while the lakes turned into a CO<sub>2</sub>-sink as they matured, the increased CH<sub>4</sub> emissions due  
453 to eutrophication led to the increase of atmospheric C flux when expressed in CO<sub>2</sub> equivalents.  
454 Furthermore, these measurements of CH<sub>4</sub> emissions are probably underestimated because  
455 ebullition was not measured, despite being an important CH<sub>4</sub> efflux mechanism in shallow and  
456 eutrophic lakes (Bastviken et al., 2004; DelSontro et al., 2016; Huttunen et al., 2003; McGinnis  
457 et al., 2006). Moreover, lateral CH<sub>4</sub> transport (DelSontro et al., 2018b; Fernández et al., 2016)  
458 or CH<sub>4</sub> production in the oxic water layers by cyanobacteria (Bižić et al., 2020; Günthel et al.,  
459 2020), or within anaerobic microniches associated to suspended particles (Bianchi et al., 1992;  
460 Grossart et al., 2011) or through methylphosphonate metabolism by planktonic microbes  
461 (Khatun et al., 2019; Wang et al., 2017) cannot be excluded (Peeters et al., 2019). Yet, the  
462 balance between autotrophy (mainly in water column) and respiration (mainly in sediment),  
463 both influenced by maturity, seems clearly determine the total flux of GHG from gravel pit  
464 lakes.

### 465 3.3.Perspectives

466 This study provides first insights on autumn emissions in gravel pit lakes and highlights the  
467 importance of CH<sub>4</sub> emissions from mature ecosystems. In particular, gravel pit lakes studied  
468 here, similarly to others man-made small ecosystems (e.g., farm and urban ponds), generally  
469 emitted more CO<sub>2</sub>-equivalent (supported by higher CH<sub>4</sub> emission contribution) than lakes,  
470 ponds and reservoirs ecosystems. This is probably because these small and shallow artificial  
471 ecosystems tend to be highly productive (e.g., Ortiz-Llorente and Alvarez-Cobelas, 2012; Webb  
472 et al., 2019) and hence, exhibit frequent phytoplankton and cyanobacteria blooms that may fuels  
473 CH<sub>4</sub> production rates (e.g., Günthel et al., 2020; West et al., 2016). Since gravel pit lakes are  
474 made in terrestrial ecosystems (considered as C sinks), GHG emissions, in particular the CH<sub>4</sub>  
475 ones, can be considered as a direct consequence of the construction of the waterbody (concept  
476 of net emissions, Prairie et al., 2018). Considering their potential high number in the landscape

477 of many countries, small artificial waterbodies should be integrated in the future estimates of  
478 regional and global C emissions from inland waters. The present study included a limited  
479 number (n = 11) of gravel pit lakes and a more complete survey is still needed to fully estimate  
480 their contribution. This is particularly important to quantify annual emissions of gravel pit lakes  
481 since it is likely that benthic and epilimnetic metabolisms as well as resulting gas flux vary  
482 between seasons and years (e.g., Jeppesen et al., 2016; Laas et al., 2012). In particular,  
483 considering that the magnitude of sediment respiration and the epilimnetic metabolism are  
484 strongly regulated by primary production, one might expect a switch from net autotrophy to net  
485 heterotrophy during the winter season due to a reduced phytoplankton growth (Marshall and  
486 Peters, 1989). A long-term survey is therefore needed to fully characterize the dynamics of  
487 GHG fluxes and the extent to which gravel pit lakes are C sink or sources over time. It would  
488 be also interesting to assess the organic C stored in sediments that can potentially be considered  
489 as an offset to net C emissions (Prairie et al., 2018). Finally, a crucial avenue of research is to  
490 identify the extent to which the switch from heterotrophy to autotrophy is regulated by biotic  
491 interactions and the structure of food webs (e.g., Atwood et al., 2013; Cole et al., 2000;  
492 Davidson et al., 2015). A previous study on the same gravel pit lakes showed changes in fish  
493 communities from an initial biomass of pioneer and predator species (e.g., European perch) in  
494 young lakes to higher biomass of cyprinids species in mature and eutrophic lake (Zhao et al.,  
495 2016), probably driven by fishery management. Changes in fish community composition  
496 (including planktivorous fishes) along the maturation gradient may strongly control the  
497 zooplankton community, which, as primary consumers of phytoplankton, might have an effect  
498 on primary productivity (e.g., Carpenter et al., 1999). Here, zooplankton biomass exhibited no  
499 significant changes along the maturity gradient. However, a lack of complete survey cannot  
500 rule out changes in species composition or abundance along the maturity gradient. It is likely  
501 that both controls (i.e., by food web and eutrophication) exert here in concert and that the  
502 increased primary production due to eutrophication is accelerated in lakes where the biomass  
503 of predatory fish is reduced. Further researches will be needed to disentangle the relative  
504 importance of both controls.

#### 505 **4. Conclusion**

506 This study demonstrated that eutrophication increased CO<sub>2</sub> sequestration in small and shallow  
507 lakes by increasing the relative importance of primary productivity compared to ecosystem  
508 respiration. Nevertheless, eutrophication also increased net GHG emissions in terms of climate  
509 impact by providing labile organic matter for enhanced CH<sub>4</sub> production. Consequently, gravel  
510 pit lakes turned into CO<sub>2</sub>-sinks along the ecosystem maturity gradient but also into hotspots of  
511 CH<sub>4</sub> emissions. Additionally, since CH<sub>4</sub> emissions increased as gravel pit lake mature,  
512 mitigation strategies are urgently needed to maintain intermediate levels of eutrophication by  
513 limiting nutrient inputs, using biomanipulation or removing sediments. By providing a first  
514 insight into the biogeochemical functioning of gravel pit lakes, this study confirms recent  
515 evidences on the important role of small artificial waterbodies on global C cycling. Further  
516 studies are therefore needed to assess the GHG emissions and their environmental drivers in  
517 small artificial ecosystems in order to accurately estimate their global contribution to C budgets.

518

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527

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Fig.1: Map of the 11 gravel pit lakes in the Garonne floodplain (south-west of Toulouse, France).

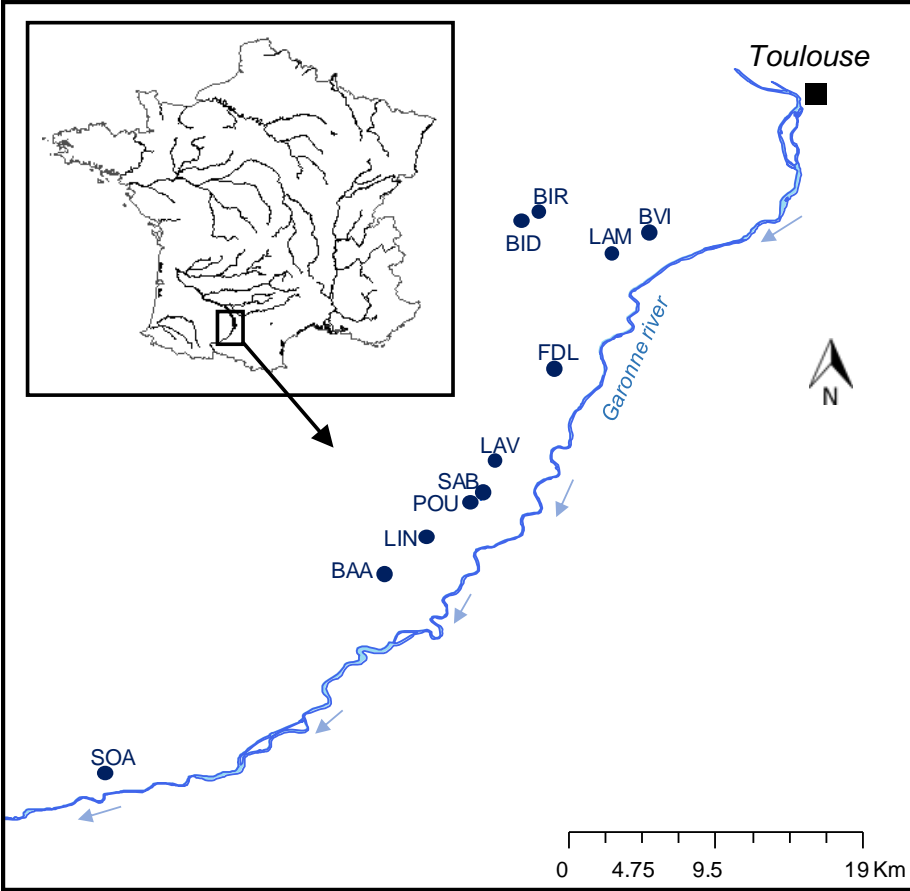


Fig. 2: Environmental characteristics of the 11 studied lakes: (A) correlation circle for the environmental variables (only significant variables were included,  $P$ -value < 0.05), and (B) factorial plane of the lakes on the PCA.

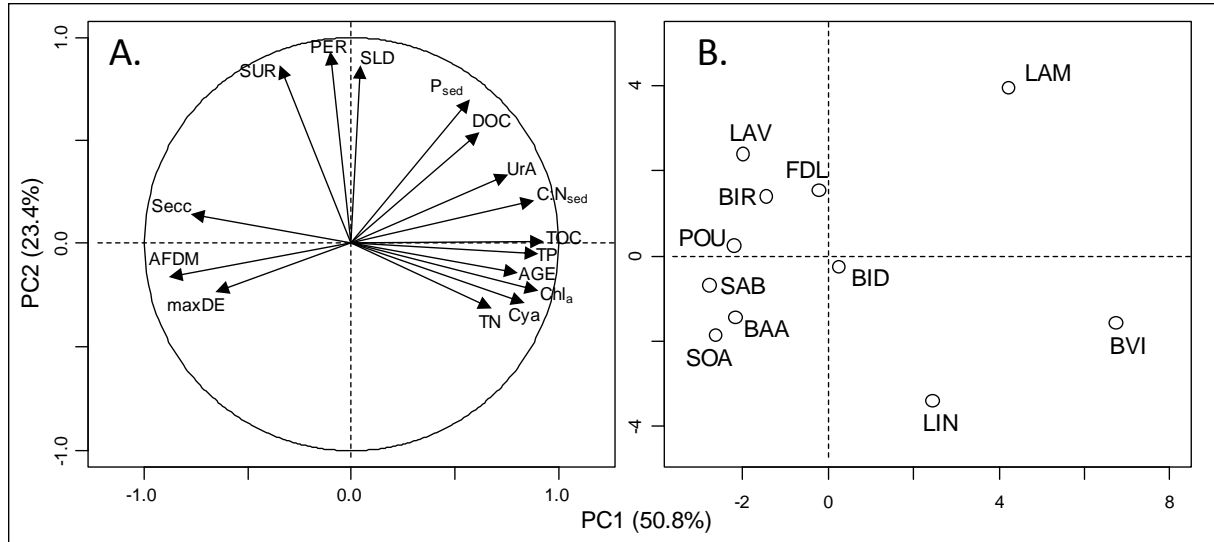


Fig. 3: Relationship between (A) CO<sub>2</sub> and O<sub>2</sub> departure from saturation (mean ± SD) and (B) CO<sub>2</sub> and CH<sub>4</sub> departure from saturation (μM) in the studied gravel pit lakes. Dots are filled based on the level of ecosystem maturity

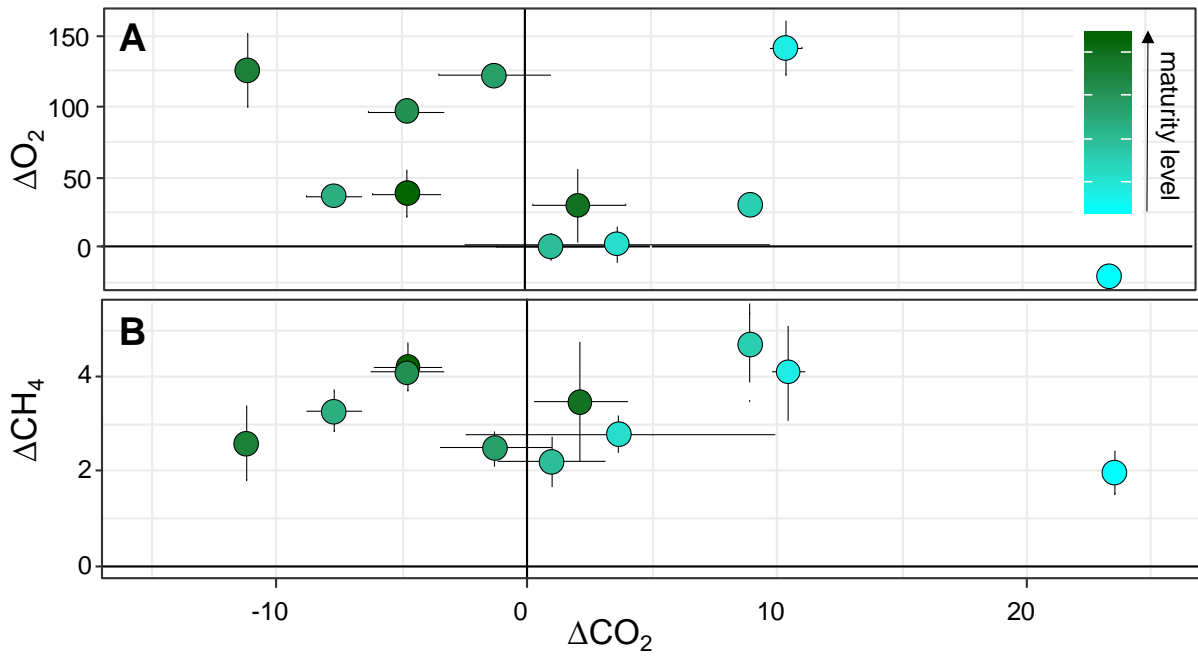


Fig.4: Total emission fluxes expressed in  $\text{g C m}^{-2} \text{d}^{-1}$  (mean  $\pm$  SD) in the 11 studied gravel pit lakes. Dots represent the mean total emission fluxes and are filled based on the level of ecosystem maturity. Bar plots represent the relative contribution of  $\text{CH}_4$  and  $\text{CO}_2$  fluxes into net total emission fluxes.

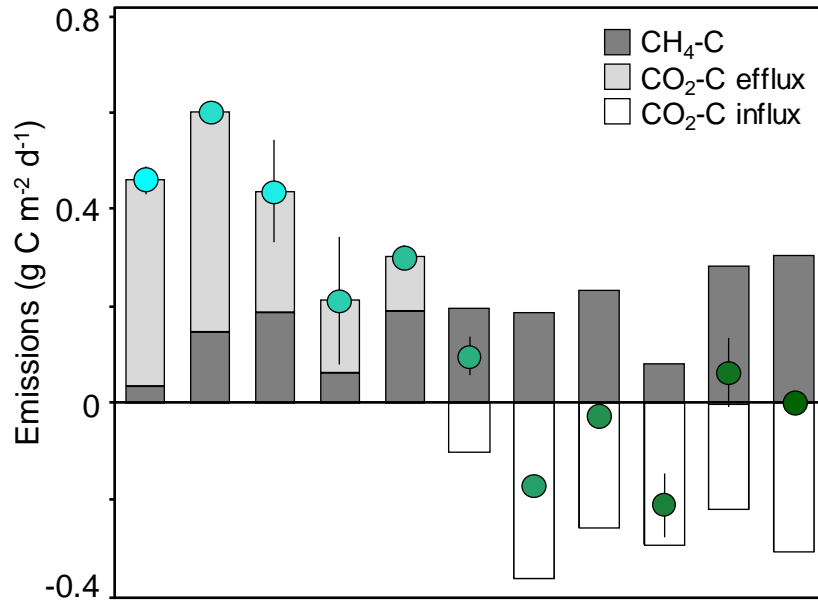




Fig. 5: Relationship between CO<sub>2</sub> or CH<sub>4</sub> dissolved concentrations in the water column and emissions with the most influential environmental variables according the PLS analyses. (A) CO<sub>2</sub> concentrations (μM) and concentrations in chl *a* (μg L<sup>-1</sup>); (B) CO<sub>2</sub> emissions (mmol m<sup>-2</sup> d<sup>-1</sup>) and DOC concentrations (mg C L<sup>-1</sup>); (C) CH<sub>4</sub> concentrations (μM) and P content in sediments (mg P Kg<sup>-1</sup>); (D) CH<sub>4</sub> emissions (mmol m<sup>-2</sup> d<sup>-1</sup>) and TOC content in sediments (g C Kg<sup>-1</sup>). Dots are filled based on the level of ecosystem maturity.

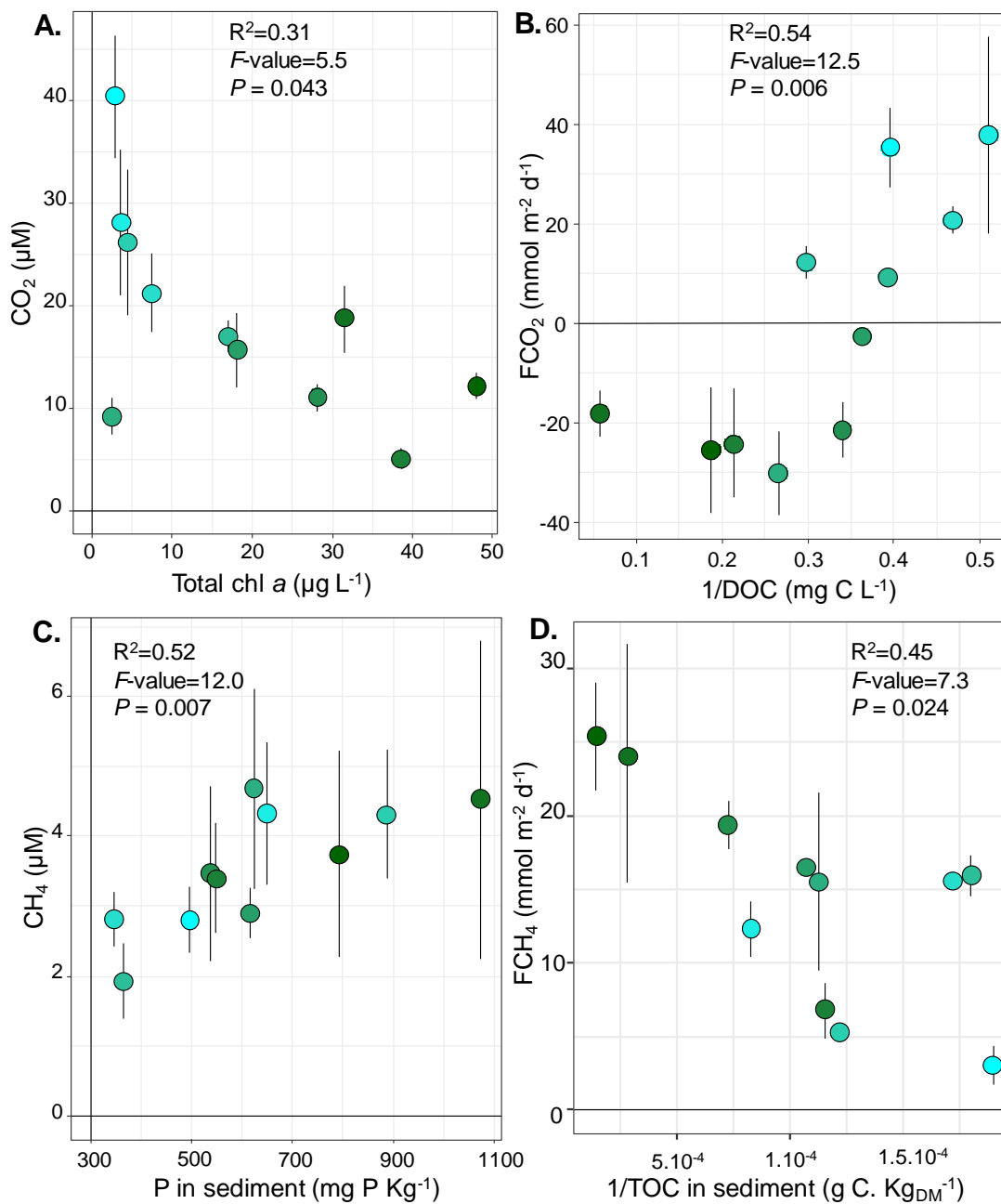
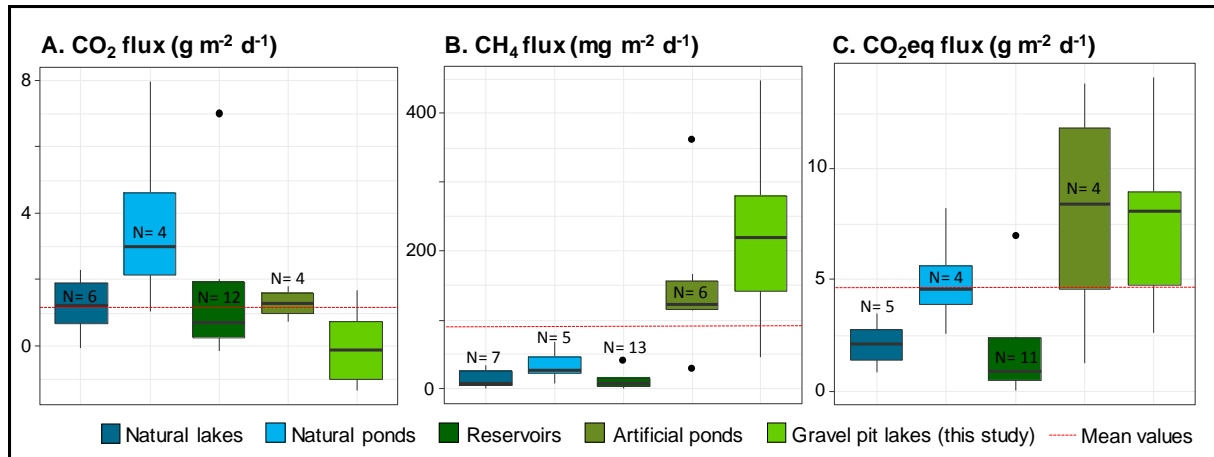


Fig. 6: Estimated flux of CO<sub>2</sub>, CH<sub>4</sub> and CO<sub>2</sub>-eq across various temperate lentic ecosystems. Values are mean flux extracted from different studies (see Supplementary material, Table A2) and from the present study (gravel pit lakes). The lines within the boxes represent median fluxes. Artificial ponds include farm, urban and storm water ponds. Mean values (red dotted line) were 1.16 g m<sup>-2</sup> d<sup>-1</sup>, 89.75 mg m<sup>-2</sup> d<sup>-1</sup> and 4.67 g m<sup>-2</sup> d<sup>-1</sup> for CO<sub>2</sub>, CH<sub>4</sub> and CO<sub>2</sub>-eq fluxes, respectively.



# Ecosystem maturity modulates greenhouse gases fluxes from artificial lakes

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Table A1: Pearson correlation coefficients of the environmental variables on the first (PC1) and second (PC2) axis. PC1 and PC2 explained 50.8% and 23.4% of the total variance, respectively. Only significant variables were displayed ( $P$ -value < 0.05)

| Variable           | PC1 (50.8%) | PC2 (23.4%) |
|--------------------|-------------|-------------|
| AGE                | 0.803       |             |
| SUR*               |             | 0.865       |
| PER                |             | 0.931       |
| maxDE              | -0.660      |             |
| SLD*               |             | 0.863       |
| UrA                | 0.754       |             |
| Temp               |             | 0.737       |
| Secc               | -0.774      |             |
| TP                 | 0.897       |             |
| TN                 | 0.681       |             |
| DOC                | 0.614       |             |
| TOC                | 0.929       |             |
| Chl <sub>a</sub>   | 0.901       |             |
| Cya                | 0.833       |             |
| AFDM               | -0.881      |             |
| TOC <sub>sed</sub> | 0.871       |             |
| N <sub>sed</sub>   | 0.880       |             |
| P <sub>sed</sub>   |             | 0.696       |

Table A2: Literature synthesis of CO<sub>2</sub> and CH<sub>4</sub> diffusive emissions from temperate lentic ecosystems. Values presented are mean (ranges) and were extracted from different studies. Only summer-autumn or dry season measurements were considered. \* For these references, values provided were expressed in CH<sub>4</sub>-C or CO<sub>2</sub>-C and converted using 1.34 and 3.66 conversion factors, respectively. All CO<sub>2</sub>- equivalent values (expressed in g CO<sub>2</sub> m<sup>-2</sup> d<sup>-1</sup>) presented in this study were calculated using the global warming potentials (GWPs) of CH<sub>4</sub> at the 100 year (1 g CH<sub>4</sub> = 34 g CO<sub>2</sub>) time- scales (IPCC, 2013). ‘FC’ refers to floating chambers and ‘TBL’ to thin boundary layer. <sup>a</sup> median values instead of means. Bold values represents mean and range values for each ecosystem type.

| Ecosystem type | n   | CO <sub>2</sub> flux<br>(g m <sup>-2</sup> d <sup>-1</sup> ) | CH <sub>4</sub> flux<br>(mg m <sup>-2</sup> d <sup>-1</sup> ) | CO <sub>2eq</sub> flux<br>(g m <sup>-2</sup> d <sup>-1</sup> ) | Method | References                       |
|----------------|-----|--|---|--|--------|----------------------------------|
| Natural        |     |  |   |  |        |                                  |
| Lake           | 11  |  | 7.9 (0.3 – 36.6)  |  | FC     | Bastviken et al., 2004*          |
| Lake           | 1   | 1.94   | 6.06  | 2.14   | TBL    | Casper et al., 2000              |
| Lake           | 9   | -0.09 (-0.4 – 0.03)  |   |  | TBL    | Kelly et al., 2001*              |
| Lake           | 1   | 0.84   | 15.6  | 1.37   | FC     | Bartosiewicz et al., 2015        |
| Lake           | 310 | 1.64   | 35.6  | 2.74   | TBL    | Li et al., 2018*                 |
| Lake           | 15  |  | 0.46 (0.19 – 0.87)  |  | FC     | West et al., 2016*               |
| Lake           | 3   | 2.29   | 35.3  | 3.49   | FC     | DelSontro et al., 2016           |
| Lake           | 121 | 0.62   | 5.76  | 0.81   | TBL    | Whitfield et al., 2011           |
| <b>Lake</b>    |     | <b>1.21 (-0.1 – 2.3)</b>                                     | <b>16.5 (0.5 – 35.6)</b>                                      | <b>2.11 (0.8 – 3.5)</b>  |        |                                  |
|                |     |  |   |  |        |                                  |
| Pond           | 1   |  | 22.6  |  | FC     | Duc et al., 2013                 |
| Pond           | 2   | 7.96   | 8.02  | 8.23   | FC     | Glaz et al., 2016 <sup>a</sup>   |
| Pond           | 1   | 3.48   | 26.7  | 4.39   | FC     | Bergen et al., 2019*             |
| Pond           | 10  | 2.51   | 67.4  | 4.8  | FC     | DelSontro et al., 2016           |
| Pond           | 1   | 1.01   | 46.2  | 2.58   | FC     | Burger et al., 2016 <sup>a</sup> |
| <b>Pond</b>    |     | <b>3.7 (1.0 – 8.0)</b>                                       | <b>34.2 (8.0 – 67.4)</b>                                      | <b>5.0 (2.6 – 8.2)</b>   |        |                                  |
| Artificial     |     |  |   |  |        |                                  |
| Reservoir      | 10  | 0.71 (-1.0 – 3.1)  | 17.04 (0.1 – 70.7)  | 1.29 (0.1 – 3.4)   | FC     | Tremblay, 2005*                  |
| Reservoir      | 5   | 0.66 (0.2 – 1.3)   | 6.8 (3.0 – 11.0)  | 0.90 (0.6 – 1.4)   | FC     | St. Louis et al., 2000           |
| Reservoir      | 6   | 1.89 (0.7 – 6.4)   | 14.5 (5.8 – 30.5)   | 2.38 (0.8 – 7.5)   | FC     | Bevelhimer et al., 2016          |
| Reservoir      | 6   | -0.15 (-1.2 – 1.2)   | 6.23 (3.2 – 9.5)  | 0.06 (-1.0 – 1.6)  | FC     | Soumis et al., 2004              |
| Reservoir      | 1   | 2.01   | 10.49   | 2.37   | TBL    | Jacinthe et al., 2012            |

|                         |           |                              |                              |                           |     |                        |
|-------------------------|-----------|------------------------------|------------------------------|---------------------------|-----|------------------------|
| Reservoir               | 1         | 0.66                         | 0.50                         | 0.68                      | FC  | Demarty et al., 2009   |
| Reservoir               | 10        | 0.1 (-0.002 – 0.31)          | 2.9 (0.2 – 10.6)             | 0.20 (0.05 – 0.38)        | FC  | Bastien et al., 2009   |
| Reservoir               | 4         | 0.28 (0.1 – 0.69)            | 0.28 (0.1 – 0.7)             | 0.29 (0.1 – 0.69)         | TBL | Diem et al., 2012      |
| Reservoir               | 1         | 0.12                         | 17.3                         | 0.71                      | FC  | Fearnside, 2002        |
| Reservoir               | 1         |                              | 43.3                         |                           | TBL | Descloux et al., 2017  |
| Reservoir               | 2         | 0.75 (0.3 – 1.2)             |                              |                           | TBL | Morales-Pineda et al., |
| Reservoir               | 1         | 7.07                         | 0.34                         | 7.01                      | TBL | Mosher et al., 2015    |
| Reservoir               | 5         |                              | 4.1 (2.9 – 5.8)              |                           | TBL | Maeck et al., 2013     |
| Reservoir               | 153       | 1.95                         | 13.7                         | 2.41                      | TBL | Li et al., 2018*       |
| <b>Reservoir</b>        |           | <b>1.34 (-0.2 – 7.1)</b>     | <b>10.6 (0.3 – 43.3)</b>     | <b>1.66 (0.06 – 7.01)</b> |     |                        |
| Farm Pond               | 101       | 1.82                         | 114.2                        | 5.70                      | TBL | Webb et al., 2019      |
| Farm Pond               | 77        | 1.07                         | 115.5                        | 11.12                     | FC  | Ollivier et al., 2019  |
| <b>Farm Pond</b>        |           | <b>1.5 (1.1 – 1.8)</b>       | <b>114.9 (114.2 – 115.5)</b> | <b>8.4 (5.7 – 11.1)</b>   |     |                        |
| Urban Pond              | 40        | 0.75                         | 30.3                         | 1.29                      | TBL | Peacock et al., 2019   |
| Urban Pond              | 5         |                              | 129                          |                           | FC  | Grinham et al., 2018   |
| Urban Pond              | 7         |                              | 165.9                        |                           | FC  | Ortega et al., 2019    |
| <b>Urban Pond</b>       |           | <b>0.8 (-)</b>               | <b>108.4 (30.3 – 165.9)</b>  | <b>1.3 (-)</b>            |     |                        |
| <b>Stormwater pond</b>  | <b>15</b> | <b>1.50</b>                  | <b>362.4</b>                 | <b>13.82</b>              | FC  | Gorsky et al., 2019    |
| <b>Gravel pit lakes</b> | <b>11</b> | <b>-0.049 (-1.32 – 1.66)</b> | <b>233.0 (48.9 – 408.2)</b>  | <b>7.9 (2.6 – 12.8)</b>   | FC  | Present study          |

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Table 1: Environmental characteristics (minimum, mean and maximum values) of the 11 studied gravel pit lakes.

|   | Full names  | Short names                       | Min                 | Mean                | Max                 |
|---|---|-----------------------------------|---------------------|---------------------|---------------------|
| <i>Hydromorphology</i>                    | Age (years)   | AGE* <sup>δ</sup>                 | 12.0                | 26.9                | 55.0                |
|   | Surface (ha)  | SUR*                              | 1.8                 | 13.7                | 21.2                |
|   | Perimeter (km)  | PER                               | 0.57                | 2.27                | 4.55                |
|   | Mean depth (m)  | mDE                               | 1.20                | 2.38                | 3.60                |
|   | Maximal depth (m)   | maxDE*                            | 1.90                | 4.03                | 6.90                |
|   | Volume (m <sup>3</sup> )  | VOL                               | 2.5x10 <sup>4</sup> | 2.7x10 <sup>5</sup> | 7.4x10 <sup>5</sup> |
|   | Slope (%) <sup>1</sup>  | SLO                               | 26.7                | 46.9                | 93.3                |
|   | Shoreline development <sup>2</sup>                              | SLD*                              | 2.06                | 3.05                | 5.04                |
|   | Urban area (%)  | UrA* <sup>δ</sup>                 | 5.3                 | 17.3                | 47.3                |
|   | Gravel pit area (%)   | GrA                               | 0.30                | 1.91                | 3.54                |
|   | Riparian vegetation area (%)                                    | RiA                               | 8.5                 | 47.8                | 87.3                |
| Forest area (%)                           | FoA   | 0.5                               | 28.1                | 56.6                |                     |
| <i>Water quality</i>                      | Specific conductance (μs.cm <sup>-1</sup> )                     | Cond                              | 247.9               | 367.1               | 619.3               |
|   | Temperature (°C)  | Temp*                             | 18.0                | 19.0                | 20.3                |
|   | pH  | pH                                | 7.95                | 8.28                | 8.78                |
|   | Secchi depth (m)  | Secc                              | 0.25                | 0.84                | 1.60                |
|   | Turbidity (FTU)   | Turb                              | 4.0                 | 11.6                | 25.4                |
|   | Total phosphorus (μg.L <sup>-1</sup> )                          | TP* <sup>δ</sup>                  | 28.8                | 59.6                | 115.9               |
|   | Phosphate (P-PO <sub>4</sub> <sup>2-</sup> μg.L <sup>-1</sup> ) | PPO4                              | 4.7                 | 16.0                | 76.6                |
|   | Total nitrogen (mg.L <sup>-1</sup> )                            | TN* <sup>δ</sup>                  | 0.48                | 1.28                | 3.11                |
|   | Nitrates (N-NO <sub>3</sub> <sup>-</sup> mg.L <sup>-1</sup> )   | NNO3                              | <0.11               | 0.20                | 0.98                |
|   | Ammonium (N-NH <sub>4</sub> <sup>+</sup> mg.L <sup>-1</sup> )   | NNH4                              | 0.08                | 0.10                | 0.17                |
|   | Nitrite (N- NO <sub>2</sub> <sup>-</sup> μg.L <sup>-1</sup> )   | NNO2                              | <3.0                | 3.5                 | 16.0                |
|   | Sulphate (mg.L <sup>-1</sup> )                                  | SO4                               | 25.0                | 52.3                | 139.0               |
|   | Dissolved organic carbon (mg C.L <sup>-1</sup> )                | DOC                               | 2.0                 | 4.3                 | 17.5                |
|   | Total organic carbon (mg C.L <sup>-1</sup> )                    | TOC                               | 1.70                | 4.28                | 9.50                |
|   | Dissolved oxygen (mg.L <sup>-1</sup> )                          | DO                                | 8.1                 | 10.1                | 12.9                |
|   | Dissolved oxygen at saturation (%)                              | DOsat                             | 86.3                | 108.8               | 140.3               |
|   | Total chlorophyll (μg chl-a L <sup>-1</sup> )                   | Chl <sub>a</sub> * <sup>δ</sup>   | 2.5                 | 17.3                | 48.2                |
| Cyanobacteria (μg chl-a L <sup>-1</sup> ) | Cya   | 2.1                               | 8.8                 | 248.2               |                     |
| Zooplankton biomass (mg/m <sup>3</sup> )  | Zoo   | 48.2                              | 34.3                | 764.1               |                     |
| <i>Sediment</i>                           | Grain size distribution (%)                                     |                                   |                     |                     |                     |
|   | Clay (<2 μm)  | Cl <sub>a</sub>                   | 0.3                 | 2.8                 | 5.5                 |
|   | Silt (2-50 μm)  | Sil                               | 2.4                 | 52.1                | 83.3                |
|   | Sand (>50 μm)   | San                               | 13.6                | 45.1                | 97.3                |
|   | Ash free dry mass (%)   | AFDM                              | 85.4                | 94.6                | 98.6                |
|   | Total organic carbon (g C. Kg <sub>DM</sub> <sup>-1</sup> )     | TOC <sub>sed</sub> * <sup>ξ</sup> | 1.1                 | 15.7                | 73.3                |
|   | Kjeldahl nitrogen (g N.Kg <sub>DM</sub> <sup>-1</sup> )         | N <sub>sed</sub> * <sup>δ</sup>   | 0.10                | 1.72                | 5.66                |
|   | Phosphorus (mg P.Kg <sub>DM</sub> <sup>-1</sup> )               | P <sub>sed</sub> *                | 0.35                | 0.62                | 1.07                |
| C:N                                       | C:N <sub>sed</sub>  | 4.1                               | 8.8                 | 14.4                |                     |

<sup>1</sup> SLO = (maxDE / Distance from banks) x 100

<sup>2</sup> SLD = PER / (2√πSUR) according to Hutchinson (1957)

\*variables included in partial least squares models. <sup>δ</sup> log-transformed variables. <sup>ξ</sup> inverse transformation.

Table 2: Gas concentrations (mean  $\pm$  SD) and fluxes at the interfaces surface water/air (mean  $\pm$  SD) and sediment/water for the 11 gravel pit lakes. Lakes are ordered according to level of maturity from the youngest to the oldest lake. For  $k_{\text{CO}_2}$  the number in brackets indicates the number of measurements with floating chambers used for calculating  $k$ . n.s. indicates invalidated data because models were not significant or  $R^2 < 0.80$ .  $\Delta_{\text{gas}}$  indicates the departures from saturation.

| Lakes  |                  |                  |                  |                  |                   |                  |                  |                  |                  |                  |                  |
|--|------------------|------------------|------------------|------------------|-------------------|------------------|------------------|------------------|------------------|------------------|------------------|
|  | SAB              | POU              | BAA              | LAV              | SOA               | BID              | BIR              | FDL              | LIN              | LAM              | BVI              |
| <i>Gas concentrations (<math>\mu\text{M}</math>)</i>                         |                  |                  |                  |                  |                   |                  |                  |                  |                  |                  |                  |
| CO <sub>2</sub>  | 39.9 $\pm$ 6.0   | 26.9 $\pm$ 7.1   | 20.1 $\pm$ 3.8   | 25.3 $\pm$ 7.1   | 17.4 $\pm$ 1.6    | 15.2 $\pm$ 3.6   | 8.7 $\pm$ 1.8    | 11.6 $\pm$ 0.9   | 5.2 $\pm$ 0.8    | 18.6 $\pm$ 3.3   | 11.7 $\pm$ 1.2   |
| $\Delta\text{CO}_2$  | 23.5 $\pm$ 5.3   | 10.4 $\pm$ 0.7   | 3.7 $\pm$ 6.1    | 8.9 $\pm$ 0.1    | 0.9 $\pm$ 2.2     | -1.3 $\pm$ 2.3   | -7.7 $\pm$ 1.1   | -4.8 $\pm$ 1.5   | -11.2 $\pm$ 0.2  | 2.1 $\pm$ 1.9    | -4.8 $\pm$ 1.4   |
| CH <sub>4</sub>  | 2.6 $\pm$ 0.6    | 4.2 $\pm$ 1.0    | 2.8 $\pm$ 0.6    | 4.4 $\pm$ 0.3    | 2.0 $\pm$ 0.2     | 2.9 $\pm$ 0.5    | 4.3 $\pm$ 1.3    | 3.6 $\pm$ 0.4    | 3.2 $\pm$ 0.7    | 4.0 $\pm$ 1.2    | 4.0 $\pm$ 0.7    |
| $\Delta\text{CH}_4$  | 2.0 $\pm$ 0.5    | 4.1 $\pm$ 1.0    | 3.1 $\pm$ 0.4    | 4.8 $\pm$ 0.9    | 2.2 $\pm$ 0.5     | 2.5 $\pm$ 0.4    | 3.3 $\pm$ 1.4    | 4.1 $\pm$ 1.2    | 2.6 $\pm$ 0.8    | 3.5 $\pm$ 2.3    | 4.2 $\pm$ 1.5    |
| CH <sub>4</sub> :CO <sub>2</sub>   | 0.07 $\pm$ 0.005 | 0.18 $\pm$ 0.12  | 0.14 $\pm$ 0.05  | 0.18 $\pm$ 0.05  | 0.11 $\pm$ 0.003  | 0.20 $\pm$ 0.07  | 0.51 $\pm$ 0.03  | 0.31 $\pm$ 0.007 | 0.68 $\pm$ 0.16  | 0.25 $\pm$ 0.08  | 0.31 $\pm$ 0.04  |
| O <sub>2</sub>   | 259.7 $\pm$ 8.6  | 325.6 $\pm$ 50.2 | 275.2 $\pm$ 13.0 | 359.4 $\pm$ 80.9 | 323.5 $\pm$ 61.5  | 395.0 $\pm$ 7.1  | 309.0 $\pm$ 3.0  | 327.7 $\pm$ 61.6 | 403.1 $\pm$ 33.9 | 312.2 $\pm$ 12.1 | 310.2 $\pm$ 16.7 |
| $\Delta\text{O}_2$   | -19.9 $\pm$ 1.7  | 141.2 $\pm$ 19.1 | 2.9 $\pm$ 12.9   | 30.6 $\pm$ 1.0   | 0.6 $\pm$ 10.1    | 122.7 $\pm$ 7.1  | 36.7 $\pm$ 3.0   | 96.8 $\pm$ 3.0   | 125.5 $\pm$ 26.5 | 30.1 $\pm$ 25.8  | 37.8 $\pm$ 16.7  |
| <i>Gas exchange coefficient (<math>\text{m d}^{-1}</math>)</i>               |                  |                  |                  |                  |                   |                  |                  |                  |                  |                  |                  |
| $k_{\text{CO}_2}$  | 1.5 $\pm$ 0.3(4) | 3.4 $\pm$ 1.7(5) | 6.4 $\pm$ 0.6(3) | 1.3 $\pm$ 0.3(4) | 10.4 $\pm$ 0.8(3) | 5.6 $\pm$ 1.2(3) | 4.0 $\pm$ 1.2(4) | 4.5 $\pm$ 1.5(5) | 2.1 $\pm$ 1.0(6) | 8.6 $\pm$ 1.5(5) | 5.0 $\pm$ 3.0(5) |
| $k_{\text{CH}_4}$  | 1.6 $\pm$ 0.3    | 3.4 $\pm$ 1.6    | 6.3 $\pm$ 0.6    | 1.2 $\pm$ 0.3    | 10.3 $\pm$ 0.8    | 5.5 $\pm$ 1.1    | 3.9 $\pm$ 1.3    | 4.5 $\pm$ 1.5    | 2.1 $\pm$ 1.0    | 8.6 $\pm$ 1.5    | 5.1 $\pm$ 3.1    |
| <i>Atmospheric emissions (<math>\text{mmol m}^{-2} \text{d}^{-1}</math>)</i> |                  |                  |                  |                  |                   |                  |                  |                  |                  |                  |                  |
| $F_{\text{CO}_2}$  | 37.8 $\pm$ 8.0   | 35.3 $\pm$ 19.8  | 20.8 $\pm$ 2.7   | 12.2 $\pm$ 3.2   | 9.2 $\pm$ 1.5     | -8.1 $\pm$ 1.8   | -30.1 $\pm$ 8.4  | -21.5 $\pm$ 5.7  | -24.2 $\pm$ 10.9 | -18.2 $\pm$ 4.7  | -25.5 $\pm$ 12.6 |
| $F_{\text{CH}_4}$  | 3.0 $\pm$ 1.3    | 12.4 $\pm$ 1.9   | 15.7 $\pm$ 0.5   | 5.4 $\pm$ 0.1    | 16.0 $\pm$ 1.4    | 16.5 $\pm$ 0.5   | 15.6 $\pm$ 4.1   | 19.4 $\pm$ 4.6   | 6.8 $\pm$ 1.4    | 23.6 $\pm$ 8.1   | 25.4 $\pm$ 3.6   |
| <i>Benthic fluxes (<math>\text{mmol m}^{-2} \text{d}^{-1}</math>)</i>        |                  |                  |                  |                  |                   |                  |                  |                  |                  |                  |                  |
| $F_{\text{sed,O}_2}$   | -28.4            | n.s              | n.s              | -93.8            | 21.2              | n.s              | n.s              | -29.8            | n.s              | -47.5            | 26.4             |
| $F_{\text{sed,CO}_2}$  | 154.3            | 122.9            | 228.0            | 144.1            | n.s               | n.s              | -33.6            | 22.1             | 33.7             | n.s              | -54.4            |
| $F_{\text{sed,CH}_4}$  | n.s              | n.s              | n.s              | 178.7            | n.s               | n.s              | 140.8            | n.s              | n.s              | n.s              | n.s              |

Table 3: Loadings of the PLS analyses explaining the variability in gas concentrations (CO<sub>2</sub>, CH<sub>4</sub>) and emissions ( $F_{CO_2}$ ,  $F_{CH_4}$ ) among gravel pit lakes. Coefficients (Coef.) and loadings (Load.) are provided only for parameters with VIP > 0.7. Highly influential variables (VIP > 1.0) and loadings values > 0.3 are displayed in bold. n.a. represents ‘not analyzed’. Parameter names are provided in the Table 1.

| Y                   | CO <sub>2</sub> |        |              | $F_{CO_2}$  |        |              | CH <sub>4</sub> (μM) |        |              | $F_{CH_4}$  |        |              |
|---------------------|-----------------|--------|--------------|-------------|--------|--------------|----------------------|--------|--------------|-------------|--------|--------------|
| Components          | 1               |        |              | 1           |        |              | 1                    |        |              | 2           |        |              |
| r <sup>2</sup> Y    | 0.56            |        |              | 0.61        |        |              | 0.59                 |        |              | 0.87        |        |              |
| Parameter           | VIP             | Coef.  | Load.        | VIP         | Coef.  | Load.        | VIP                  | Coef.  | Load.        | VIP         | Coef.  | Load.        |
| Chla                | <b>1.55</b>     | -1.378 | <b>-0.42</b> | <b>1.36</b> | -3.320 | <b>-0.37</b> | <0.7                 |        |              | <b>1.21</b> | 0.779  | <b>-0.51</b> |
| CH <sub>4</sub>     | <0.7            |        |              | <0.7        |        |              | n.a.                 |        |              | n.a.        |        |              |
| DOC <sup>ξ</sup>    | <b>1.52</b>     | 1.345  | <b>0.41</b>  | <b>1.74</b> | 4.231  | <b>0.47</b>  | 0.81                 | -0.067 | -0.23        | 0.74        | -0.476 | -0.23        |
| F <sub>sedCO2</sub> | <b>1.04</b>     | 0.923  | 0.28         | <0.7        |        |              | n.a.                 |        |              | n.a.        |        |              |
| maxDE               | <b>1.34</b>     | 1.184  | <b>0.36</b>  | <b>1.40</b> | 3.406  | <b>0.37</b>  | <b>1.11</b>          | -0.092 | <b>-0.32</b> | <b>1.29</b> | -0.835 | <b>-0.32</b> |
| Nsed <sup>δ</sup>   | <0.7            |        |              | <0.7        |        |              | <b>1.04</b>          | 0.086  | <b>0.30</b>  | <b>1.05</b> | 0.678  | <b>0.30</b>  |
| Psed                | <0.7            |        |              | <0.7        |        |              | <b>1.58</b>          | 0.130  | <b>0.46</b>  | 0.73        | 0.440  | 0.25         |
| SLD                 | <0.7            |        |              | <0.7        |        |              | 0.99                 | 0.083  | 0.29         | <0.7        |        |              |
| SUR                 | 0.72            | 0.635  | 0.19         | <0.7        |        |              | 0.74                 | 0.061  | 0.21         | <0.7        |        |              |
| Temp                | <0.7            |        |              | <0.7        |        |              | <b>1.26</b>          | 0.104  | <b>0.36</b>  | <0.7        |        |              |
| TN <sup>δ</sup>     | <b>1.10</b>     | -0.975 | -0.29        | <b>1.08</b> | -2.627 | -0.29        | <0.7                 |        |              | <0.7        |        |              |
| TOCsed <sup>ξ</sup> | <b>1.18</b>     | 1.045  | <b>0.32</b>  | <b>1.13</b> | 2.757  | <b>0.30</b>  | <b>1.49</b>          | -0.123 | <b>-0.43</b> | <b>1.60</b> | -1.028 | <b>-0.43</b> |
| TP <sup>δ</sup>     | <b>1.26</b>     | -1.115 | <b>-0.34</b> | <b>1.26</b> | -3.069 | <b>-0.34</b> | <0.7                 |        |              | 0.75        | 0.481  | <b>-0.51</b> |
| UrA <sup>δ</sup>    | <b>1.17</b>     | -1.034 | <b>-0.31</b> | <b>1.44</b> | -3.495 | <b>-0.38</b> | <b>1.02</b>          | 0.084  | 0.29         | <b>1.79</b> | 1.156  | 0.29         |

<sup>δ</sup> log-transformed variables. <sup>ξ</sup> inverse transformation.