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Influence des processus externes et internes sur les émissions de gaz à effet de serre dans les lacs de petite taille

The influence of external and internal processes in regulating greenhouse gas emissions in small lakes

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Résumé

La majorité des études sur les gaz à effet de serre (GES) se concentrent sur les lacs profonds de moyenne à grande tailles et possédant un régime de mélange bien défini, tandis que les lacs de petite taille et peu profonds ont reçu moins d’attention malgré leur abondance et leur ubiquité. Dans ces lacs, les processus allochtones et autochtones qui contrôlent les émissions de GES et leur stockage dans la colonne d’eau ne sont pas encore bien compris. Les caractéristiques physiques, biochimiques et biologiques des petits lacs changent rapidement suivant les conditions météorologiques, affectant les taux, le moment, le lieu et la voie de libération des GES. De plus, la méthode sélectionnée pour estimer le coefficient de transfert gazeux (k) peut affecter notre capacité à estimer avec précision les flux de GES provenant des petits lacs et leur contribution au budget global des émissions de GES. Dans cette thèse, l'interaction entre les caractéristiques limnologiques et les émissions de GES, ainsi que leur stockage dans le milieu pélagique et les zones littorales ont été étudiés pendant deux à trois saisons d’eau libre dans deux lacs de petite taille. Ces lacs se contrastaient par la présence d’efflorescences phytoplanctoniques récurrentes et l'abondance de macrophytes. Les flux diffusifs de GES ont été calculés avec trois méthodes standards, et comparés pour des années montrant différentes conditions météorologiques.

Pendant l’été le plus chaud (2012), le lac Jacques (moins de 2 m de profondeur) a montré une stratification persistante malgré sa faible profondeur, son hypolimnion est devenu anoxique, et le lac est devenu un puits de CO_2 pendant les périodes sèches, tandis qu’après les périodes orageuses, le CO_2 et le CH_4 se sont accumulés dans l’hypolimnion. Les flux diffusifs du CH_4 à l’été 2012 étaient similaires à ceux mesurés au milieu de l'été précédent, et quatre fois plus élevés lors du retournement automnal. La comparaison entre les méthodes de calcul des flux a révélé que les estimations du modèle de renouvellement de surface étaient plus près des valeurs obtenues par les chambres flottantes (max 39% de différence) par rapport aux estimations basées uniquement sur la vitesse du vent (max 52% de différence). Néanmoins, en l’absence de données détaillées sur les échanges de chaleur, les modèles basés sur le vent peuvent tout de même fournir une approximation raisonnable des flux de gaz.

Le potentiel de réchauffement climatique (PRC) du lac (resultant de l’ébullition et de la diffusion des GES) était significativement plus élevé pendant l’été le plus frais, avec une plus grande contribution de l’ébullition par comparaison avec l’été le plus chaud. La zone littorale (< 1 m de profondeur, 0.07 km^2) avait un potentiel de réchauffement plus faible que la zone pélagique (> 1 m de profondeur, 0.11 km^2), mais seulement en 2011. En fait une fois normalisée par unité de

surface, la zone littorale avait un potentiel plus élevé de réchauffement que la zone pélagique en 2012.

Les analyses révèlent que la température de l'eau et la Chl-*a* expliquent respectivement 27% et 20% de la variance du CO_2 pour les deux années, montrant ainsi l'effet négatif de la croissance du phytoplancton sur le CO_2 dissous. La variance du CH_4 pendant l'année pluvieuse a été principalement expliquée par le phosphore total et les nitrates (respectivement 25% et 19%), tandis que les nitrates et la température expliquent 35% et 29% de la variance en N_2O. Pendant l'année sèche, la variance de CH_4 est plutôt expliquée par la Chl-a et les nitrates (respectivement 47% et 36%), tandis que pour le N_2O, aucune des variables testées n'était un prédicteur significatif de sa variance. Ces résultats suggèrent qu'au-dessus d'un certain seuil d'énergie solaire incidente au système, les processus autochtones (stabilité de la colonne d'eau, croissance du phytoplancton, niveaux d'oxygène) peuvent dominer les processus allochtones (pluie, vent, apports externes en nutriments) dans le contrôle des émissions en GES.

Le lac St-Augustin (moins de 6 m de profondeur) montrait des efflorescences de cyanobactéries en 2012 (été chaud et sec) et des fleurs d'eau de dinoflagellés en 2011 (été moyennement pluvieux). La période de canicule a généré un déficit en oxygène dans l'hypolimnion, et le lac était continuellement supersaturé en CH_4 et en N_2O (jusqu'à 2.7 µM et 15.2 nM respectivement), et périodiquement sous-saturé en CO_2 (jusqu'à -8.5 µM). Pendant l'année la plus pluvieuse, le lac était également supersaturé en CH_4, mais les concentrations étaient plus basses (en moyenne 0.27 µM) par rapport à 2012 (0.36 µM respectivement). Les concentrations en Chl-*a* et la biomasse des dinoflagellés et des cyanobactéries expliquaient relativement bien la variabilité des niveaux de saturation du CO_2 et de CH_4 ($R^2 > 0.4$, $p < 0.01$) dans la zone pélagique, mais pas dans la zone littorale du lac ($R^2 < 0.3$). L'abondance totale des cellules picoplanctoniques dans les eaux de surfaces était un bon prédicteur pour le niveau de saturation en CH_4 dans les zones littorales et pélagiques du lac ($R^2 = 0.55$ et $R^2 = 0.47$, respectivement). La biomasse du zooplancton était corrélée au niveau de saturation du CO_2 ($R^2 = 0.21$) dans la zone pélagique du lac.

Les résultats de cette thèse donnent un aperçu de l'interaction entre les facteurs de contrôles physiques et biologiques sur les émissions en GES provenant des petits lacs. Le réchauffement climatique entrainera une plus forte stratification thermique, ce qui pourra mener vers : i) une forte variation temporelle des concentrations en GES, avec des moments chauds et/ou des points chauds d'émission, ii) une augmentation de l'importance de la diffusion du CH_4 et du N_2O

relativement au CO_2. L’extrapolation et la modélisation de la contribution des petits lacs au budget global des GES requièrent plus de données sur comment les processus autochtones et allochtones qui contrôlent les niveaux de saturation en GES répondront aux forçages météorologiques. Les estimations basées sur quelques mesures discrètes sans évaluation de la variabilité saisonnière et interannuelle des émissions en GES, peuvent être largement biaisées, en particulier sous des conditions climatiques plus chaudes.

Abstract

The majority of greenhouse gas (GHG) studies focus on large and deep lakes with well-defined mixing regimes, while small lakes have received less attention despite their abundance and ubiquity. In these lakes, allochtonous and autochthonous processes controlling GHG emissions and water column storage are not yet well understood. Physical, biochemical and biological characteristics of small lakes change rapidly with weather conditions, affecting the rates, timings, locations and pathways of GHG releases. In addition, the method which is selected to estimate the gas transfer coefficient (k) may affect the capacity to accurately estimate the GHG flux from small lakes and their contribution to the global GHG budget. In this thesis, the interactions among the limnological characteristics and the rates, pathways and timings of GHG emissions and storage in the pelagial and littoral zones of two contrasting small-sized lakes with and without recurrent phytoplankton blooms was investigated during two or three open water seasons. Diffusive GHG fluxes were calculated with three standard methods and compared between years with different weather conditions.

During the hotter summer (2012), Lake Jacques (less than 2 m deep) had a persistent layered structure despite its shallowness, its hypolimnion became anoxic, and the lake was a sink for CO_2 during dry periods, while CO_2 and CH_4 accumulated in the hypolimnion after rainstorms. Diffusive fluxes of CH_4 in the summer of 2012 (1.8 mmol m^{-2} d^{-1}) were similar to those measured in mid-summer of the previous year (1.1 mmol m^{-2} d^{-1}), and over four times higher during the autumnal overturn. The comparison between methods revealed that surface renewal model estimations were more similar to the floating chamber measurements (max 39% difference) than estimations based solely on wind speed (max 52% difference). Nevertheless, in the absence of detailed data on heat exchange, wind based models can still provide reasonable approximations of gas fluxes. The global warming potential (GWP) of the lake (ebullition and diffusion) was significantly higher during the cooler summer (28.1×10^3 mol CO_2 d^{-1}) with a greater contribution from ebullition (17.5×10^3 mol CO_2 d^{-1}), than in the hotter summer (14.4×10^3 mol CO_2 d^{-1}) with a smaller contribution from ebullition (3.3×10^3 mol CO_2 d^{-1}). The littoral zone (< 1 m deep, 0.07 km^2) had a smaller warming potential than the pelagial zone (> 1 m deep, 0.11 km^2), particularly in 2011 (8.9 and 19.1×10^3 mol CO_2 d^{-1}, respectively).

Linear regression analysis revealed that water temperature and Chl-a explained respectively 27% and 21% of the CO_2 variance for both years, showing the negative effect of phytoplankton growth on dissolved CO_2. Most of the variance in CH_4 during the rainy year could be explained by TP and NO_3^- (respectively 25% and 19%), while NO_3^- and temperature explained respectively

35% and 29% of the variance in N_2O. Conversely, in the dry year, the variance in CH_4 was best explained by Chl-a and NO_3^- (respectively 47% and 36%), while none of the tested variables could significantly explain the variance in N_2O. These results suggest that above a certain threshold of incoming solar energy, autochthonous processes (water column stability, phytoplankton growth, oxygen levels) may dominate over allochthonous processes (rain, wind, external inputs of nutrients) in controlling GHG.

Lake St Augustin (less than 6 m deep) had noxious cyanobacterial blooms in 2012 (hot, dry summer) and a dinoflagellate bloom in 2011 (cool, rainy summer). The hotter weather generated hypolimnetic oxygen depletion, and the lake was continuously supersaturated in CH_4 and N_2O (up to 2.7 µM and 15.2 nM respectively), and periodically under-saturated in CO_2 (down to -8.5 µM). In the rainier year, the lake was also supersaturated in CH_4, but the concentrations were significantly lower (in average 0.27 µM) than in 2012 (0.36 µM respectively). The Chl-a concentrations, dinoflagellates and cyanobacteria biomass could explain the variability in CO_2 and CH_4 saturation levels ($R^2 > 0.4$, $p < 0.01$) in the pelagial, but not in the littoral zone of the lake ($R^2 < 0.3$). The total number of bacterioplankton in surface waters was a good predictor of the level of CH_4 saturation in both the littoral and pelagial zones of the lake ($R^2 = 0.55$ and $R^2 = 0.47$, respectively). Zooplankton biomass was only correlated with the level of CO_2 saturation ($R^2 = 0.21$) in the pelagial zone of the lake.

The results of this thesis provide insights into the interplay between physical and biological controls on GHG emissions from small lakes. The autochthonous and allochtonous effects on GHG emissions in lakes may differ from year to year depending on the main processes responsible for changes in GHG saturation levels. Stronger lake stratification, as a result of global warming, will lead to: i) high temporal variability in their GHG emissions with pronounced hot moments and/or hot spots, ii) higher CH_4 and N_2O diffusive fluxes relative to CO_2. The up-scaling and modelling of the contributions of small lakes to the global GHG budget require more data on how autochthonous and allochthonous processes, controlling GHG saturation levels, will respond to meteorological forcing. Estimations based on a few discrete measurements without assessing seasonal and interannual variability in GHG emissions may be largely biased, particularly under a warmer climate.

AVANT-PROPOS

Les articles de cette thèse présentent des recherches originales sur les émissions et le stockage des gaz à effet de serre dans les lacs de petite taille, et l'influence des processus allochtones et autochtones sur ce. La thèse est composée de quatre parties. La première partie est une synthèse sur le contexte de recherche, les objectifs, les résultats principaux et une conclusion résumée. La deuxième partie est le coeur de la thèse et présente trois articles scientifiques décrits ci-dessous. La troisième partie présente la liste des références citées et la quatrième est une annexe.

Articles de la thèse

1. **Bartosiewicz M.**, Laurion I. and S. MacIntyre. 2015. GHG emissions and storage in a small shallow lake. Hydrobiologia (Springer), DOI: 10.1007/s10750-015-2240-2.

2. **Bartosiewicz M.**, Laurion I., MacIntyre S. and R. Maranger. 2015. Impact of summer heat wave on the oxygen, nutrients and global warming potential of a small lake. *Soumis* à Limnology and Oceanography.

3. **Bartosiewicz M.**, Laurion I., Przytulska A. and R. Maranger. 2015. Effects of planktonic food web structure and algal blooms on greenhouse gases in a hypereutrophic lake. *En preparation* pour Freshwater Biology (Wiley).

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Le poème de James Joyce m’a accompagné pendant l’écriture de cette thèse.

All day I hear the noise of waters	*Toute la journée j’entends le bruit des eaux*
Making moan,	*Faisant gémir,*
Sad as the sea-bird is when, going	*Triste comme l’oiseau de mer est quand, allant*
Forth alone,	*En avant seul,*
He hears the winds cry to the water's	*Il entend les vents pleurer vers la monotone*
Monotone.	*Des eaux.*
The grey winds, the cold winds are blowing	*Les vents gris, les vents froids soufflent*
Where I go.	*Où je vais.*
I hear the noise of many waters	*J’entends le son de tant d’eaux*
Far below.	*Loin ci-dessous.*
All day, all night, I hear them flowing	*Toute la journée, toute la nuit, je les entends s’écouler*
To and fro.	*Çà et là.*

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Glossaire des Abreviations

GES (GHG) - Gaz à Effet de Serre
PRC (GWP) - Potentiel de Réchauffement Climatique

CH_4 - Methane

N_2O - Protoxyde d'azote

MO - Matière organique

MOD (DOM) - Matière organique dissoute

UV - Rayonnement ultraviolet

MOP (POM) - Matière organique particulaire

COP (POC) - Carbone organique particulaire

COD (DOC) - Carbone organique dissous

Chl-*a* – Chlorophylle *a*

LOI - La perte au feu (loss on ignition)

DO - oxygène dissous

TP - Phosphore total

TN - Azote total

SRP - Phosphore réactif soluble

CV - Coefficient de variation

APC (PCA) - Analyse en composantes principales

MOB - Bactéries oxydant le méthane

K – Le coefficient de transfert de gaz

LW - Le rayonnement solaire à ondes longues

SW -Le rayonnement solaire à ondes courtes

MLD - Profondeur de la couche de mélange

PP - production primaire

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PARTIE 1 – SYNTHÈSE

1.1. Introduction

1.1.1. L'importance mondiale des petits lacs

Les petits lacs ont longtemps été sous-estimés pour leur rôle dans de nombreux processus écologiques et biochimiques importants à l'échelle mondiale, jusqu'à récemment, la majorité des études limnologiques se sont concentrées sur le fonctionnement et les caractéristiques de grands écosystèmes distincts (Downing et al., 2006; Downing, 2010). L'un des premiers rapports sur la distribution des lacs (Thienemann, 1925) suggère qu'environ 1.8% de la surface terrestre est couverte de lacs et que la superficie globale des lacs est dominée par de grands lacs. Ceci mène à une sous-estimation constante de la superficie globale des lacs, qui est probablement presque deux fois plus étendue que celle présentée dans les livres de limnologie (Kalff, 2001), et plus précisément sur l'importance des petits lacs. Avec des outils de télédétection plus performants, des études récentes ont montré que les petites zones d'eaux continentales contribuent largement à la superficie mondiale des lacs (les lacs plus petits qui <0,01 km^2 constituent 31% de la superficie totale des lacs, Tableau 1.1).

Superficie (km^2)	Nombre de lacs	Superficie totale des lacs (km^2)	Source
0.001-0.01	277,400,000	692,600	Downing et al., 2006
0.01-0.1	24,120,000	602,100	Downing et al., 2006
0.1-1	2,097,000	523,400	Downing et al., 2006
1-10	182,300	455,100	Downing et al., 2006
10-100	15,905	392,362	Lehnet et Döll, 2004
100-1,000	1,330	329,816	Lehnet et Döll, 2004
1000-10,000	105	257,856	Lehnet et Döll, 2004
>10,000	17	985,769	Lehnet et Döll, 2004
All lakes	304,000,000	4,200,000	Downing et al., 2006

Tableau 1.1.1 Le nombre et la superficie des lacs mondiaux. Les valeurs sont inclusives pour les bornes inférieures mais exclusives pour les bornes supérieures. Inspiré de Downing et al. 2006.

Compte tenu de leur forte abondance dans le paysage, les petits lacs jouent un rôle important en biométéorologie, la dispersion de la pollution de l'air, la formation de nuages, la circulation de l'eau et de la chaleur, et les loisirs (Segal et al., 1997). Les petits lacs et les bassins sont aussi importants pour leur rôle dans la préservation de la biodiversité. Par exemple, Scheffer et al. (2006) a mis en évidence que les petits lacs contiennent souvent plus d'espèces relativement à leur petite surface par rapport à des lacs plus grands et plus profonds, représentant ainsi des îles de la biodiversité régionale. Plusieurs mécanismes peuvent contribuer à phénomène. Tout d'abord, les petits lacs peuvent fournir des conditions environnementales hautement localisées (Williams et al., 2004). En un deuxième temps, ces conditions environnementales peuvent varier considérablement sur des échelles de temps journalières et saisonnières, offrant ainsi des possibilités dans l'établissement d'une communauté plus diversifiée.

Souvent sur un court laps de temps (<1000 ans), les petits lacs stockent, traitent et émettent de grandes quantités de nutriments et de carbone. Ces systèmes, longtemps considérés comme de simples conduits entre les environnements terrestre, océanique et atmosphérique, sont en réalité des réacteurs d'importance mondiale où la matière organique peut être transformée et ré-émise vers l'atmosphère sous forme de GES (Cole et al., 2007; Tranvik et al., 2009). Les études effectuées par Bastviken et al. (2004) et Kortelainen et al. (2006) ont révélé que les quantités de carbone à la surface d'un lac diminuent drastiquement avec l'augmentation de la taille du lac. Similairement, la séquestration de la matière organique dans les sédiments lacustres a été suggérée etre un ordre de grandeur plus élevé dans les petits lacs que dans lesgrands (Downing et al., 2006). Globalement, en dépit de leur faible surface, le taux de traitement et le stockage de la matière organique dans les petits lacs peu profonds est si élevé que ces eaux pourraient bientôt émerger comme les écosystèmes les plus importants à l'échelle mondiale.

Bien que l'abondance des petits lacs puisse diminuer à certains endroits autour du globe a cause des changements climatiques et de l'augmentation de l'évaporation (Le Viol et al., 2009; Carroll et al., 2011), au niveau régional, le nombre de petits lacs pourrait augmenter en raison d'une fréquence plus élevé de pluies soudaines et de la rétention de l'eau dans le paysage. Dans les régions tropicales, malgré une tendance générale à la désertification, le niveau de l'eau a augmenté en raison de l'augmentation du ruissellement (Leduc et al., 1997; 2001) créant de nombreu petits lacs permanents ou éphémères (e.g. dans Le Sahel; Desconnets et al., 1997). Dans les régions subtropicales et tempérées, le nombre d'étangs d'origine agricole a augmenté (Leon et al., 2010), ainsi que le nombre de bassins de loisirs, de sécurité et de rétention de la pollution. Dans certaines régions localisées à de hautes altitudes ou latitudes, le nombre de petits lacs augmente en réponse au reculement des glaciers (Salerno et al., 2014) ou à leur disparition (Huggel et al., 2002), aux inondations des tourbières alpines (S. Glatzel commun. pers.) ou du dégel du pergélisol (Bouchard et al., 2014). Dans les régions où les petits lacs étaient rares dans le passé, leur récent développement pourrait entraîner un transport disproportionné de fortes quantités de carbone et de nutriments provenant des sols préalablement congelés vers l'atmosphère (Walter et al., 2006; Negandhi et al., 2013).

Compte tenu de leur nature transitoire, le fonctionnement des petits lacs est gravement affecté par les effets de l'eutrophisation et des changements climatiques. Moss et al. (2013) ont souligné que les changements dans le fonctionnement de petits écosystèmes lacustres peuvent être liés: i) aux effets directs du réchauffement climatique tels que des régimes de précipitations anormaux ou la survenance de vagues de chaleur estivales renforçant la stratification et conduisant à l'hypoxie, ii) aux effets indirects liés aux changements dans les interactions trophiques entre les poissons, le zoo- et le phytoplancton. Il peut exister une relation considérablement forte entre les changements climatiques et l'eutrophisation, issue de la hausse des apports en nutriments et de l'augmentation des températures qui tendent à intensifier le déséquilibre entre le phosphore et l'azote conduisant à des fleurs d'eau de cyanobactéries (Paerl et Huisman, 2009). Ces fleurs d'eau sont souvent suivies par la désoxygénation de la colonne d'eau et de la mort de la faune pélagique qui affectent le cycle des nutriments et leur rétention dans les sédiments. Dans le sens inverse, il a été montré que les petits lacs eutrophes plus chauds et désoxygénés émettent plus de GES, y compris de fortes quantités de CH_4 et N_2O qui ont potentiel de réchauffement climatique (PRC) 28 et 298 fois plus élevé que le CO_2 (Myhre et al., 2013).

1.1.2 Production, consommation et voies d'émission de gaz à effet de serre

L'étude des émissions de GES provenant des lacs a commencé bien longtemps avant que notre point de vue standard en limnologie soit développé (Forel, 1892). Dans les premières lettres à son ami le père Carlo Campi, Allessandro Volta qui a observé de mystérieux bouillonnements sur un lac après une période de dégel lui écrivit : *« Alors le troisième de ce mois (Nov. 1776)… en étant sur un petit bâteau sur le Lac Magiorre et passant à proximité d'une zone couverte de roseaux, j'ai commencé à pousser et remuer le fond de l'eau avec ma canne. Tellement 'd'air' a émergé que j'ai décidé d'en recueillir une certaine quantité dans un grand récipient en verre… ».*

Comme beaucoup de scientifiques dans leurs premières expériences, Volta a décidé de mettre le feu à son sujet expérimental (« L'air » recueilli). Cet essai a conduit au résultat le plus inattendu lorsque les gaz collectés dans le fond du lac présentaient une flamme bleu claire durant leur incinération. Les expériences suivantes et les séries de lettres publiées dans des revues françaises et allemandes ont posé les fondations de l'étude de la production de GES dans les écosystèmes aquatiques. Récemment, au cours de ces dernières décennies, l'intérêt porté à l'étude des voies de production et d'émission des GES par les lacs, du fait de leur potentiel et du réchauffement climatique, n'a cessé d'augmenter (Cole et al., 2007). Les trois GES les plus émis par l'eau continentale du fait de la décomposition de la matière organique et des phénomènes de respiration sont le CO_2, le CH_4 et le N_2O (IPPC 2013).

La matière organique (MO) des lacs peut être d'origine terrestre ou aquatique. Dans la littérature la majorité des études considèrent que la MO d'origine terrestre constitue la réserve principale de MO décomposée par les bactéries dans les lacs, et posent le postulat que la MO allochtone est la première source du CO_2 produit (Cole et al., 1994). Cependant de récents travaux de Meberly et al. (2013) montrent que la MO terrestre peut aussi être décomposée in situ (dans les sols) et être ensuite exportée sous forme de CO_2 dans les lacs. Indépendamment de l'origine du CO_2 des lacs, qu'il soit exporté directement des terres ou qu'il soit produit à l'intérieur du lac à partir de substrats terrestres, la majorité des petits lacs reçoivent plus de MO qu'ils ne peuvent en stocker dans leur sédiments ce qui conduit à un relargage de carbone et d'azote dans l'atmosphère, les lacs devenant net hétérotrophiques (del Giorgio et Peters, 1993).

La décomposition microbienne et l'oxydation photochimique de la MO, se produisant soit dans la colonne d'eau ou dans les sédiments, sont les principales réactions à l'origine des GES contenues dans l'eau des lacs. L'acétate et le CO_2 sont les deux premiers produits de la fermentation de la MO dans les sédiments. Près de la moitié de la MO est dégradée de manière incomplète dans les sédiments (en acétate) et fournie en conséquence les substrats pour des réactions de méthanogénèse anaérobie (Conrad et al., 2010). Dans les lacs polymictiques de faible envergure où l'hypolimnion est rarement entièrement anoxique, le CH_4 produit par les sédiments est oxydé à l'interface sédiment-eau, fournissant du CO_2 aux niveaux supérieurs de la colonne d'eau. La respiration dans la colonne d'eau (lié aux poissons, au plancton ou aux macrophytes) peut être une autre source significative de CO_2 dans les lacs. Del Giorgio et Peters (1993), ont observé que la respiration du plancton excède la photosynthèse algale lorsque les concentrations en chlorophylle a sont supérieures à 17 $mg \cdot m^{-3.}$ Ces analyses, réalisées sur un jeu de données de 118 lacs extrait de 15 publications, ont prouvé que l'augmentation de la respiration avec le statut trophique du lac est beaucoup plus importante pour le phytoplancton seul (collecté avec un filet présentant des mailles de 60 µm de diamètre) que pour n'importe quels autres composants de la respiration du lacs (bactéries et zooplancton). Cela signifie que lorsque le statut trophique augmente (et la biomasse phytoplanctonique), la respiration phytoplanctonique peut être responsable de l'augmentation des concentrations en CO_2. Les bactéries hétérotrophes contribuent de 65 à 40 % aux concentrations en CO_2 dans les lacs oligotrophes et eutrophes respectivement (del Giorgio et Peters, 1993). L'importance relative du zooplancton à la respiration totale du lac est dans la majorité des cas en dessous de 20%, avec quelques exceptions dans les lacs ultraoligotrophes (Cole et al., 1989).

Le dioxyde de carbone est aussi produit dans les lacs à partir de la décomposition photochimique de la matière organique. Durant cette réaction, les photons en dessous de 500 nm entraînent le clivage d'une fraction des molécules de la MOD (Strome et Miller, 1978), qui en plus du CO_2 relargue des nutriments qui stimulent la croissance du phytoplancton. Dans certains petits lacs humiques, des teneurs importantes en MOD (représentant parfois jusqu'à 80 % du carbone organique total), conduisent à des taux élevés de minéralisation photochimique (Cooper et al., 1989). Par exemple, 10% du

CO_2 dissous d'un lac humique en Suède provient de la photolyse de la MOD alors que 40 % est issu de la respiration benthique, et 50 % de la respiration pélagique (Jonsson et al., 2001). Les molécules larges de la MOD (tels que les acides humiques) sont, durant la photolyse, réduit en de plus petite molécules qui peuvent être immédiatement exploitées par les bactéries (Backlund, 1992; Salonen et Vahatalo, 1994).

Le CO_2 des lacs est consommé par le phytoplancton durant la photosynthèse (Talling 1976). Une photosynthèse intense mène à une élévation du pH avec potentiellement une précipitation du CO_2 sous forme de bicarbonate. Dans des cas extrêmes lorsque le pH excède 10.5-11 (Jeppesen et al. 1998), l'eau du lac peut assimiler le CO_2 atmosphérique par une réaction chimique directe (Kuss and Schneider, 2004). Les macrophytes peuvent aussi consommer le CO_2 de la colonne d'eau (Westlake, 1975). L'efficacité locale de cette consommation est, cependant, plus fortement contrôlée par la vélocité du courant et la morphologie des plantes que dans le cas du phytoplancton (Nielsen et al., 2006). La méthanogénèse peut aussi être considérée comme une réaction génératrice de CO_2 dans les sédiments (appelée réduction de carbonate hydrogénotrophe; Whiticar et al., 1986). Dans les hypolimnions (Rudd et Taylor, 1980; Casper, 1992) ou dans les microzones anoxiques présentes dans la partie oxygénée de la colonne d'eau (Grossart et Simon, 1993).

Le méthane est principalement produit dans les sédiments et l'hypolimnion anoxique par les archées méthanogènes (Ferry, 1993), et dans les microhabitats anoxiques du système digestif du zooplancton (de Angelis et Lee 1994), des poissons (van der Maarel et al. 1999) et sur les particules en suspensions ou les aggrégats en cours de sédimentation (Grossart et Simon, 1997). Divers mécanismes tels que le clivage des methyl-esters par les radiations UV, la production de méthane induit par hypoxie dans les mitochondries, le méthane libéré par les organismes soumis à un stress oxydatif, et la décomposition des composés méthylés ont aussi été suggérés comme une cause de la supersaturation en CH_4 dans les eaux oxygénées (Keppler et al., 2009). Récemment, Grossart et al. (2011) et Bogard et al. (2014) ont émis le postulat que le CH_4 peut être produit en présence d'oxygène par des méthanogènes attachés à du phytoplancton en suspension, lui-même présentant des concentrations en acétate et en H_2 dans les environs de la cellule algale plus importante; c'est ce que confirme la récente étude de

Tang et al. (2014). Les auteurs soulèvent un autre concept intéressant; celui que la thermocline pourrait agir comme une barrière face aux bulles de CH_4 provenant des profondeurs de la colonne d'eau, conduisant à leur diffusion subséquente dans l'hypoliminion oxique. Néanmoins dans leur étude, Tang et al. (2014) réfutaient aussi leurs propres hypothèses puisque la modélisation de leurs résultats montrait que les bulles s'échappant des sédiments auraient perdu tout leur CH_4 avant d'atteindre la thermocline. Dans la mesure où les microbulles de CH_4 peuvent provenir des eaux saturées situées bien au-dessus des sédiments (Prairie et del Giorgio, 2013), cette hypothèse nécessite des études complémentaires.

Généralement, le méthane est considéré comme étant principalement consommé dans la colonne d'eau à travers des réactions de méthanotrophie aerobique, néanmoins son oxydation in situ è l'intérieur des sédiments par des réactions de méthanotrophie anaérobique a aussi été observé (Eller et al., 2005). Il a été reporté que l'oxydation anaérobique du CH_4 s'accompagne d'une réduction des sulfates (Valentine et Reeburgh, 2000) et d'une dénitrification (Deutzmanna et al., 2014). Il a été prouvé récemment que la méthanotrophie entraînée par la réduction des sulfates était stimulée par la réduction du fer (Sivan et al., 2014).

L'oxyde nitreux (N_2O) dans les lacs est produit à la fois au cours de la nitrification bactérienne et la nitrication des *Archeae* sous des conditions suboxiques voire hypoxiques (Mengis et al., 1997), mais aussi pendant la dénitrification partielle se produisant dans les micro-habitats pauvres en oxygène (Codispoti et Christensen, 1985). Le N_2O dans la colonne peut, par ailleurs, être un sous-produit du métabolisme des méthanotrophes (Yoshinari, 1985). Dans les lacs, d'autres sources potentielles de N_2O encore peu décrites dans la littérature existent comme la production de ce composé par le phytoplancton (Cohen et Gordon 1979, Guieysse et al. 2013, Fig. 1.1.1) et le zooplancton. Dans la colonne d'eau les possibles pertes du N_2O se ferait sous l'action d'une réduction dissimulative de celui-ci en N_2 et de sa consommation par certaines cyanobactéries (Farias et al., 2013). Les trois principaux GES produit dans les lacs de faible envergure peuvent être émis soit par diffusion, soit par ébullition (Bastviken et al., 2004) ou par transport facilité par les plantes (Dacey et Klug, 1979) (Fig. 1.1.2).

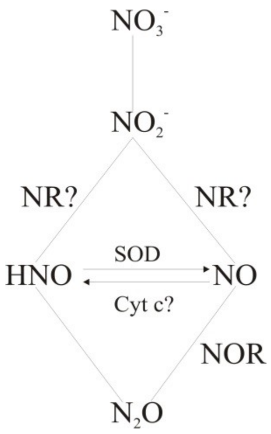

Figure 1.1.1 Voies possibles de la formation du NO2 chez le phytoplancton (NO_3^- = nitrate, NO_2^- = nitrite, NO = nitric oxide, HNO = nytroxyl, NR = nitrate reductase, SOD = superoxide dismutase, NOR = NO reductase, Cyt c = cytochrome c oxidase. Modifié deGuieysse et al. (2013).

Ils peuvent aussi être stoqués dans l'hypolimnion durant l'été, dans la colonne d'eau au cours de l'hiver (Ducharme-Riel et al., 2015) ou encore être piégés dans la couche de glace recouvrant le lac (Greene et al., 2014, Langer et al., 2015). Cette réserve de GES sera partiellement émise dans l'atmosphère au cours du brassage automnale ou au printemps lorsque la glace fond.

Le transport par diffusion à l'interface eau-atmosphère dépend de la solubilité du gaz, de la différence entre la teneur du gaz dans l'eau et dans l'air, et enfin de la vélocité du transfert (appelée aussi vitesse du piston). Les gaz se diffusant à partir de la surface des lacs sont soit produits dans la colonne d'eau, soit dans les sédiments. Dans le cas de GES fortement solubles tels que le CO_2 et le N_2O (Abrilet al., 2005; Bauch et al., 2011), la diffusion est probablement la voie d'émission la plus significative. En ce qui concerne le CH_4, gaz le moins soluble, l'ébullition a souvent été soulignée comme la voie d'émission dominante (Casper et al., 2000; Bastviken et al., 2004). Les bulles de GES peuvent se former dans les sédiments et à l'interface sédiment-eau lorsque la pression partielle des gaz dissous dépasse la pression hydrostatique (Chanton et Whiting, 1995). Il est possible de séparer l'ébullition stable de celle épisodique (Green, 2013). La première apparaît lorsqu'un courant continu de bulles est libéré par les sédiments (bullage fin), alors que la seconde consiste en des évènements occasionnels de bullage (bullage grossier) qui peuvent être déclenché par des changements dans la pression atmosphérique ou encore être la conséquence de perturbations à la surface des sédiments. Selon le type d'ébullition, les GES s'échappant des sédiments pourront être mis en contact avec de l'eau oxigenée pour des durées plus ou moins longues. La probabilité qu'ils soient consommés, transformés et incorporés dans le réseau trophique pélagique sera dépendante de cette durée d'exposition (Bastviken et al., 2003).

Malgré l'engouement croissant du monde scientifique pour l'étude des voies d'émission des GES à partir des eaux continentales, la quantification des phénomènes d'ébullition dans ces écosystèmes demeure un point d'incertitude considérable. Bien que la majorité des études présentent l'ébullition comme le mécanisme principale d'émission du CH_4, en lui attribuant jusqu'à 95 % des flux de CH_4 totaux (e.g. Schubert et al., 2012; Walter et al., 2006). Green et Baird (2012) émettaient l'hypothèse que les évènements occasionnels de bullages ne contribuaient pas significativement à l'émission totale de CH_4 provenant des tourbières submergées. De la même manière, Repo et al. (2007) reportaient que l'ébullition de CH_4 ne participait que faiblement au flux de CH_4 totaux émis par les petits lacs de Sibérie, et constituait une composante négligeable pour les flux de CO_2. Cette controverse pourrait s'expliquer en partie par le fait que le taux et la fréquence des événements d'ébullition sont affectés par la température de l'eau (qui régule l'activité microbienne au sein des sédiments) et par la quantité d'énergie incorporée dans la colonne d'eau due au stress induit par le vent ou par le différentiel réchauffement/refroidissement (Wik et al., 2014). Elle peut aussi être le résultat de l'utilisation de méthodes inappropriées pour estimer correctement un phénomène à ce point variable dans le temps et dans l'espace.

Les transports de GES directement à partir des sédiments vers l'atmosphère par les plantes aquatiques ont été rapportés par Dacey et Klug (1979) dans des nénuphars. Ce transport produit via le réseau des espaces internes de gaz dans les tiges de plantes, représentant un système d'écoulement sous pression qui force l'oxygène vers les racines et le rhizome situés dans les sédiments anaérobies, et inversement, déplace les sédiments de GES vers l'atmosphère (Dacey, 1980). Cette voie de libération de GES permet à une partie du CH_4 ou du N_2O (Jørgensen et al., 2012) de s'échapper du lac sans être oxydée dans la colonne d'eau. D'autre part, les plantes aquatiques peuvent également introduire de l'oxygène dans les sédiments (Gerard et Chanton, 1993) et ainsi réduire les conditions optimales pour la production de CH_4.

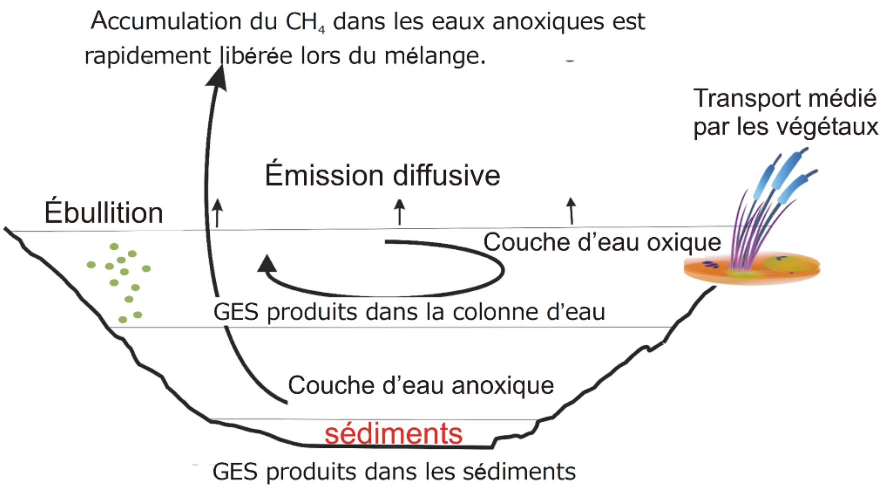

Figure 1.1.2 Voies d'émission des GES (modifié de Bastviken et al., 2008).

1.1.3 Méthodologie pour estimer les émissions et le stockage de GES dans les petits lacs

Alors que les émissions de GES provenant des lacs ont été intensivement étudiées au cours des dernières décennies (Bastviken et al., 2011; Raymond et al., 2013), la grande majorité des études ont porté sur les flux de gaz de diffusion à la surface du lac. Les flux de diffusion résultent de gradients de concentrations de GES entre l'eau et l'atmosphère et du mélange turbulent à la surface du lac. La turbulence peut être généré en profondeur ou à la surface du lac, et est entraînée par le vent forçant et par les échanges de chaleur à la surface du lac (MacIntyre et al., 2010). À des vitesses de vent faibles, la turbulence résulte tout autant des différences de vitesses des courants, mais aussi de l'eau froide conduisant à mouvements convectifs naturels (Wüest et Lorke, 2003). À des vitesses de vent modérées à élevées, le cisaillement est suffisamment fort pour provoquer des ondulations à la surface de l'eau, qui devient peu à peu turbulente. À des vitesses de vent élevées, l'échange de chaleur résultant de l'évaporation représente une force supplémentaire au forçage du vent directement causant la turbulence (MacIntyre et al., 2002). La turbulence est rarement mesurée directement (par exemple, avec des vélocimètres acoustiques à effet Doppler); à la place un coefficient de transfert de gaz (k) et les échanges gazeux sont calculés avec des modèles basés sur le vent, des modèles de renouvellement de surface, des tours de covariance de Foucault, ou des mesures in situ à l'aide de chambres flottantes.

Chacune de ces méthodes peut donner différentes valeurs de k dans des conditions venteuses ou calmes, des conditions chaudes, conduisant à des biais dans les estimations de flux de diffusion.

Les modèles de vent sont basées sur des gaz traceurs (par exemple, Cole et Caraco, 1998; Crusius et Wanninkhof, 2003) ou des techniques de covariance de Foucault (par exemple, Jonsson et al., 2008) pour obtenir des relations quadratiques ou cubiques entre les vitesses de vent et le coefficient k. Pour ces modèles, la contrainte du cisaillement, la dynamique des flux, la dissipation d'énergie et la production de microbulles sont les principaux facteurs qui contrôlent le transfert de gaz à travers l'interface air-eau (Wanninkhof et al., 2009). Les modèles de renouvellement de surface comprennent une description quantitative du mélange des eaux de surface et du budget de la chaleur (Imberger, 1985) pour calculer le flux d'énergie de la turbulence totale et le taux de dissipation de l'énergie cinétique turbulente à l'interface. Ces modèles (Soloviev et Schluessel, 1994; MacIntyre et al., 1995; Tedford et al., 2014) donnent des valeurs de k en proportion du taux de dissipation de l'énergie cinétique et la viscosité cinématique. Récemment, Tedford et al. (2014) ont suggéré deux équations distinctes pour estimer k: i) le cisaillement du vent comme étant le mécanisme dominant causant les turbulences près de l'interface air-eau sous des conditions de chaleur, et ii) avec une augmentation des taux de dissipation d'énergie (et k) via les mouvements convectifs sous les conditions de refroidissement.

Le coefficient de transfert de gaz peut également être calculé en utilisant les flux instantanés mais très localisés de CO_2 estimés par les chambres flottantes (Frankignoulle 1988) couplées à un détecteur de gaz en ligne. Pendant de nombreuses années, des mesures de flux directs utilisant des chambres flottantes ont été discrédités en raison de l'hypothèse que la chambre peut amortir la turbulence ou, d'une certaine façon, l'augmenter (Vachon et al., 2010). Les travaux récents de Cole et al. (2010) indiquent que la conception de la chambre peuvent aider à contourner ces problèmes (voir aussi Alin et al., 2011), en particulier dans des conditions de vents faibles à modérés (Kramer et al., 2003). Le travail de Vesala et al. (2006) présente des valeurs similaires de coefficients de transfert de gaz estimés par les chambres flottantes et ceux des systèmes de covariance de Foucault installés sur les rivières, pour soutenir l'exactitude des flux estimés avec des chambres flottantes. Lorsqu'elles sont utilisées de la bonne façon, les chambres flottantes peuvent être considérées comme la norme pour tester les modèles. En outre, tandis que la diffusion provoque une augmentation linéaire de tout gaz donné dans la chambre, l’apparition de bulles de gaz provoque une déviation soudaine et distincte de la linéarité dans les ensembles de données à haute résolution. Ainsi, la chambre flottante peut être utilisée pour estimer la contribution de l'ébullition à efflux totale des GES d'un lac (Matthews et al., 2003; Soja et al., 2014).

L'utilisation de tours de covariance de Foucault et de traitement du signal numérique (Eugster et Pluss, 2009) pour mesurer les flux de diffusion de gaz est plus complexe sur le plan logistique, mais il fournit des flux à une plus grande échelle spatiale. Les systèmes de covariance de Foucault fonctionnent sur l'hypothèse que les flux verticaux de gaz covarient avec les vitesses verticales du gaz d'intérêt (Fig. 1.1.3). La méthode de covariance de Foucault implique quelques hypothèses simplificatrices telles que des fluctuations de densité négligeables et des flux verticaux dans un terrain homogène horizontal. Une fois ces hypothèses admises, les systèmes de covariance de Foucault peuvent fournir des estimations directes des flux de GES sur une zone du lac. Cependant, il existe de plus en plus de preuves que la deuxième hypothèse ne peut pas être remplie lorsque les masses d'eau sont situés sur une petite pente. Voilà pourquoi chaque site expérimental doit être traité individuellement, et les effets possibles des flux verticaux doivent être corrigés. En outre, des mesures de covariance de Foucault représentent les flux cumulés de diffusion des gaz et d’ébullition, de ce fait des méthodes alternatives peuvent être nécessaires pour distinguer les différentes voies d'émissions de GES. L’ébullition des GES peut être évaluée directement en utilisant des entonnoirs submergées et renversés. Cet appareil se compose d'un corps en forme de cône avec une seringue en plastique ou un tuyau avec soupape de 'luer-lock' installé sur le dessus, entretenu par des flotteurs juste en dessous de l'interface de l'eau. Les entonnoirs d’ébullition peuvent être déployés soit pour de courtes périodes de temps et échantillonnés à des intervalles de temps courts (Martens et Klump, 1980; Keller et Stallard, 1994), ou pour des périodes plus longues avec une relativement plus faible fréquence d'échantillonnage (par exemple, Mattson et Likens, 1990; Huttunen et al., 2001).

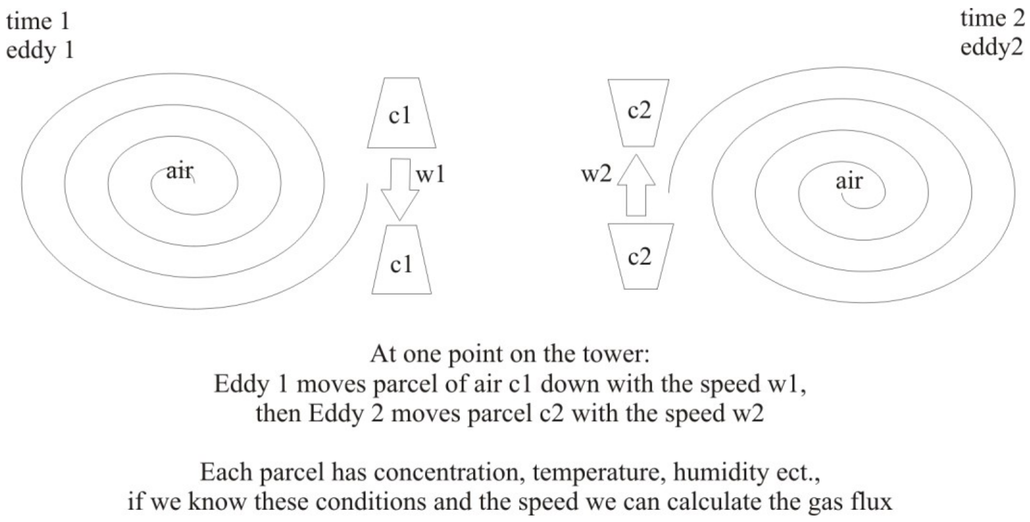

Figure 1.1.3 Schéma présentant un aperçu de l'approche de covariance de Foucault pour calculer les flux de gaz sur un lac (modifié à partir de Burba, 2013).

Les déploiements de courte durée ont l'inconvénient d'augmenter la probabilité de manquer des événements majeurs de l'ébullition, et les longs déploiements diminuent la probabilité d'évaluer avec précision la chronologie des événements bouillonnants (Varadharajan et al., 2010). Cela peut être évité lorsque les entonnoirs sont équipés de systèmes automatisés qui permettent d'enregistrer des volumes de gaz cumulés et de prélever des échantillons de gaz. Ces pièges à bulles automatisés (ABT) facilitant l'évaluation des flux d'ébullition de type stables et de type épisodiques sont actuellement développés (H. Hoffman, pers commun.). Les flux d'ébullition peuvent également être évalués avec précision en utilisant un écho sondeur possédant une haute précision (Ostrovsky et al., 2008).

Le budget des GES d'un lac peut être évalué de manière globale en utilisant le bilan massique (Encinas-Fernandez et al., 2014). Cette approche, basée sur les profils verticaux de concentrations de gaz, permet non seulement d'estimer les quantités d'un gaz donné dans le lac (via la multiplication des volumes des couches en profondeur), mais aussi de comprendre les principaux processus internes et externes responsables des changements temporels des GES dans les lacs exposés à différentes conditions météorologiques, situés dans différents paysages, et présentant des différences de fonctionnement de l'écosystème (Bastviken et al., 2008; Åberg et al., 2010).

1.1.4 Les processus allochtones de régulation de la dynamique des GES dans les petits lacs

Le fonctionnement des petits lacs est régulé par des facteurs externes (allochtones) à une étendue beaucoup plus grande que dans le cas des grands lacs (et plus profondes). Les caractéristiques des bassins versants comme l'altitude, la délimitation, l'hydrologie, la couverture végétale, le type de sol et l'utilisation des terres, contrôlent la quantité de nutriments allochtones et la matière organique qui sont transférées vers le lac (Fig. 1.1.4).

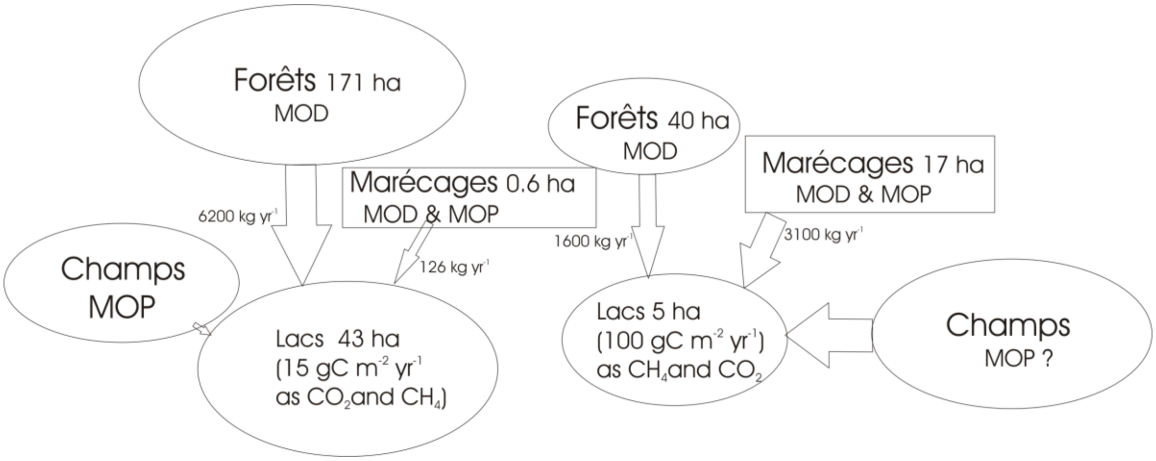

Figure 1.1.4 Diagramme schématique de l'effet de l'utilisation des terres sur les MOD allochtones et les apports en MOP dans les grands lacs par rapport aux petits lacs, et l'effet ultérieur sur les émissions annuelles de GES (uniquement de CO_2 et de CH_4). Modifié à partir de Canham et al. (2004).

Les caractéristiques météorologiques telles que la température de l'air et l'humidité, l'insolation relative et le rayonnement UV, l'ampleur et l'intensité des précipitations, ainsi que la vitesse et la direction du vent influencent la structure thermique du lac, la turbulence au sein de la colonne d'eau et au niveau des interfaces (air-eau et eau-sédiments), la quantité et la qualité des intrants de carbone, et l'emplacement de ces entrées (des intrusions dans la colonne d'eau vers le bas ou au milieu, Fig. 1.1.5). Les effets du bassin versant et des conditions météorologiques / climatiques sur les concentrations de GES dans les petits lacs peuvent être directs, grâce à des intrusions d'eau contenant du CO_2 (Maberly et al., 2013), du CH_4 ou possiblement du N_2O.

Alternativement ils peuvent être indirects, par le biais de la régulation de l'activité microbienne. La production microbienne de GES (respiration, méthanisation, production de N_2O) est contrainte par des changements de température et d'apports de matière organique (Canham et al., 2004; Urabe et al., 2011). La régulation indirecte des GES par le climat et le bassin versant peut également se répercuter sur la quantité d'oxygène qui est introduit, dissous et consommé dans la colonne d'eau du lac. Par exemple des petits lacs à haute altitude ou avec des bassins versants dans les hautes terres sont susceptibles de recevoir des flux d'eau froide bien aérés dans la partie inférieure de la colonne d'eau. Ces intrusions entraînent une dé-stratification partielle ou complète de la colonne d'eau (Botelho and Imberger, 2007), la remise en suspension de sédiments de surface, ou l'oxygénation des sédiments de surface et des eaux hypolimniques (Laborde et al., 2010). Alors que les sédiments remis en suspension peuvent entraîner une augmentation de la libération du CH_4 et du N_2O provenant des sédiments vers la couche d'eau sus-jacente (par l'intermédiaire de l'ébullition et de la diffusion), des niveaux élevés d'oxygène vont faciliter l'oxydation des deux gaz, et conduire à des émissions de CO_2 et de N_2 à la surface du lac.

La majorité des petits lacs situés à des altitudes et latitudes élevées sont nets hétérotrophes (Cole et al., 1994) en raison du climat froid et humide qui est typique dans les régions montagneuses et arctiques, laquel entraîne une diminution des taux de décomposition microbienne de la matière organique dans le bassin versant et un lessivage accru des basin versant où la matière organique est transformée en GES (Negandhi et al., 2013). Pour les petits lacs situés à des altitudes et des latitudes plus basses, les effets de l'utilisation des terres sur les concentrations de GES dans l'eau, par le biais du transfert de la matière organique et de nutriments qui diffèrent dans la forme, la quantité et la qualité. Selon Mattsson et al. (2005) la proportion de matière organique particulaire par rapport à la dissoute déposée dans les lacs augmente avec l'augmentation de l'impact humain (agriculture). La différence entre le flux de MOD provenant des terres et le MOP dans les réseaux trophiques aquatiques peut conduire à des effets différents sur les concentrations de GES en fonction de la dimension de lac D'une part, une grande partie de la MOD allochtone est biologiquement inaccessible (Søndergaard et Middelboe, 1995), mais elle peut affecter la production primaire planctonique et benthique par son influence sur l'atténuation du rayonnement photosynthétiquement actif (Carpenter et al., 1998; Markager et Vincent, 2001), ainsi que par son influence sur la stratification de la colonne d'eau (Fee et al., 1996). La partie instable de la MOD dérivant du bassin versant vers le lac est utilisée par des bactéries planctoniques, ayant un taux de croissance considérablement faible, respirent une grande partie de celui-ci sous forme de CO_2 (35-90%, del Giorgio et Cole, 1998). Certains auteurs ont également suggéré que la MOD allochtone peut être directement exploitée par le phytoplancton (Vincent et Goldman, 1980; Znachor et Nedoma, 2010) ou le zooplancton (Salonen et Hammar, 1986), et régule ainsi la production planctonique et les contrôles autochtones sur les GES. D'autre part, la MO particulaire allochtone est plus susceptible de sédimenter une fois qu'elle arrive dans les lacs, par la décomposition anaérobie en carburant (Kortelainen et al., 2004). Le dépôt de COP dans les sédiments peut entraîner des conditions plus anoxiques et favoriser la production de GES plus réduits tels que le CH_4 et le N_2O. Des quantités élevées de COP allochtones peuvent stimuler la formation d'agrégats organiques macroscopiques limnétiques (lac de neige) dans la colonne d'eau oxydée du lac (Grossart et Simon, 1993). Ces agrégats peuvent devenir des points chauds pour la décomposition microbienne et les émissions de GES. Des processus comparables à ceux qui se produisent dans « la neige marine », en la formation de micro-zones anaérobies, sont susceptibles de se produire dans les lacs (Karl et Tilbrook, 1994).

1.1.5 Les processus autochtones de régulation dynamique des GES dans les petits lacs

Les processus internes qui régulent directement ou indirectement la production, le stockage, la distribution et les émissions de GES sont particulièrement importants dans les petits lacs ou la colonne d’eau est étroitement couplée aux sédiments sur l’ensemble de la zone du lac. La température de la colonne d’eau et la stratification contrôlent la concentration en oxygène, qui à son tour renforce ou inhibe l’activité de microbes spécifiques responsables de la production et de la consommation de certains GES. Les niveaux d’oxygène contrôleront aussi la disponibilité des nutriments autochtones et le carbone organique dissous via la décomposition de la MO et la respiration, la deposition des nutriments et leur remobilisation dans les sédiments.

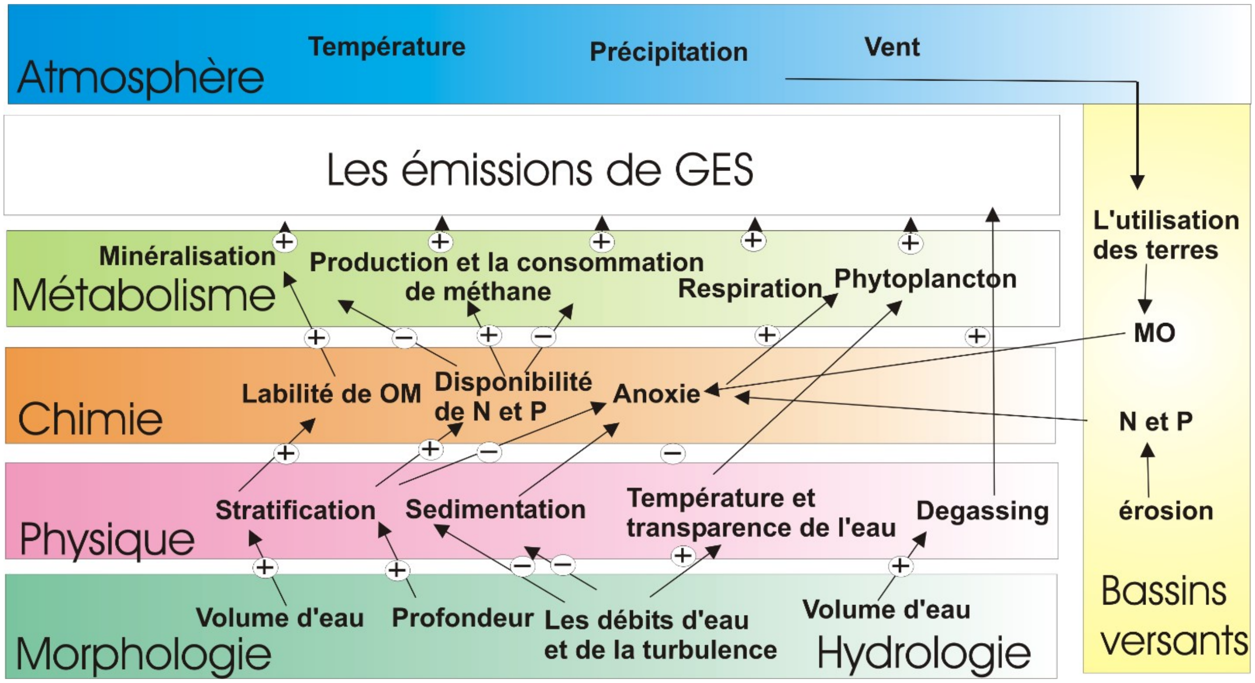

Figure 1.1.5 Schéma illustrant certains des contrôles allochtones et autochtones de la dynamique des GES dans les lacs. OC = carbone organique; PP = production primaire. Des flèches avec le symbole (+) représentent un effet positif; et celles avec le symbole (-) représentent un effet négatif. Redessiné et modifié de Mendonça et al. (2012). Courtoisie de Peter Casper.

L'abondance planctonique et la composition de leurs communautés affecteront les niveaux d'oxygène dans la colonne d'eau mais leur activité sera régulée par la température de l'eau, la disponibilité en nutriments et les interactions trophiques. Les processus biologiques reliés à l'abondance du plancton et à leur activité, ainsi que les interactions entre et au sein des niveaux trophiques, exercent un contrôle interne sur le cycle des GES dans les lacs, soit directement ou indirectement (Fig. 1.1.5).

Bastviken et al. (2003) ont montré que les bactéries méthanotrophes ont contribué à environ 0,5-10% du bactérioplancton hétérotrophe total au cours de la stratification estivale, tandis que pendant l'hiver et les périodes de brassage des couches d'eau, les méthanotrophes ont contribué de 68 à 110% de la production bactérienne. L'efficacité de croissance des méthanotrophes a varié de 6 à 80% et a abouti à de grandes différences dans la quantité de CH_4 qui s'est évacué (sans avoir été oxydé) des sédiments vers la colonne d'eau. Bien que Kankaala et al. (2006) ont supposé une gamme de l'efficacité de croissance des méthanotrophes plus étroite dans les lacs finlandais (25-50%, selon del Giorgio et Cole, 1998), les résultats des calculs ont montré que les méthanotrophes peuvent contribuer à près de 47% de la production bactérienne totale pendant l'été, et à plus de 81% durant la période automnale de mélange. En plus du CH_4, le bactérioplancton hétérotrophe, incluant les méthanotrophes, vont réguler les concentrations en CO_2 dans les lacs. L'augmentation de la pCO_2 peut résulter d'une plus grande abondance en bactéries et de leur activité liée à l'augmentation de l'exsudation photosynthétique (Grossart et al., 2006).

Le bactérioplancton autotrophe peut directement influencer les concentrations en CO_2 dans les lacs par l'absorption photosynthétique et les émissions respiratoires. L'abondance de cette fraction du phytoplancton autotrophe a également été démontrée comme étant corrélée aux concentrations de CH_4 (Schmidt et Conrad, 1993). Bien que cet effet peut être dû à la production directe de CH_4 dans la colonne d'eau oxygéné par certains microbes inconnus associés au phytoplancton, il est tout à fait possible que le bactérioplancton autotrophe abondant est plus facilement décomposé que les plus grandes cellules phytoplanctoniques plus robustes, dans les sédiments de l'hypolimnion anoxique servant de ravitaillement au phénomène de méthanisation. Schulz et Conrad (1995) ont mis en évidence une augmentation significative de l'acétate et du CH_4 dans les boues sédimentaires couvertes par les suspensions de picophytoplancton autotrophe, avec les plus fortes concentrations de CH_4 produit par decomposition du picophytoplancton de haute qualité (*Porphyridium* sp.) par opposition à ceux produits quand la matière organique de plus faible qualité (*Synechococcus* sp.).

L'abondance, l'activité et la qualité du phytoplancton peut directement influencer les concentrations en GES dans les lacs à travers sa consommation de CO_2, ou par sa production de CH_4 associé (Grossart et al., 2011; Bogard et al., 2014). Le phytoplancton affecte aussi indirectement les concentrations en GES dans les lacs par leurs effets sur les niveaux d'oxygène dans la colonne d'eau (Mallin et al., 2006) la stabilité de la surface des sédiments, le pH (Balmer et Downing, 2011), la température (Kahru et al., 1993) et les concentrations en nutriments. La biomasse du phytoplancton, produite dans les conditions de fleurs d'eau et déposée vers les sédiments du lac, a montré une stimulation de la synthèse ribosomiale des méthanogènes et augmente la production de CH_4 (Schwarz et al., 2008). Les fleurs d'eau de phytoplancton peuvent varier en qualité (e.g. la teneur en acides gras et leur composition) selon les espèces ou groupes dominants. Par conséquent, il est possible que la méthanogénèse soit stimulée à différents degrés en fonction de la qualité de la matière organique déposée.

Le zooplancton peut directement influencer les GES via leur respiration individuelle qui était (jusqu'à 20% de la variance de CO_2 pour les lacs japonais; Urabe et al., 2010), et par l'intermédiaire de la production de CH_4 (De Angelis et Lee, 1994) et peut-être aussi par le N_2O, dans leur tube digestif. Le zooplancton peut aussi indirectement influencer les GES dans les lacs à travers plusieurs autres mécanismes. Tout d'abord, les herbivores planctoniques peuvent transférer du carbone et des nutriments provenant des couches d'eau plus profondes pendant leur migration verticale (Haupt et al., 2010), affectant le phytoplancton (consommation de CO_2) et la production bactérienne (agissant sur tous les 3 GES). Deuxièmement, le zooplancton peut paître d'une manière non-sélective les cellules phytoplanctoniques ce qui pourrait être lié à l'azote ammoniacal et la libération de phosphore soluble (Urabe, 1993) ou ils peuvent brouter d'une manière sélective sur certaines espèces phytoplanctoniques responsables de changements sur la composition de la communauté, avec des effets sur la photosynthèse et l'efficacité de la respiration s'en suivent, ainsi que sur les taux de dépôts détritiques. Troisièmement, le zooplancton peut regle concentrations des GES par son effet de pâturage sur le bactérioplancton, et plus particulièrement sur les bactéries méthanotrophes (Kankaala et al., 2007).

1.1.6 La variabilité spatio-temporelle de la production et des émissions des GES dans les petits lacs

Les taux des processus responsables de la production de GES et de leurs flux peuvent varier dans le temps et l'espace menant à des points chauds, ainsi qu'a des « moments » chauds dans la production et les émissions de GES dans les petits lacs (McClain et al., 2003). L'hétérogénéité spatiale dans la production et dans les émissions de GES dans les lacs reste toujours mal comprise en raison de la résolution spatiale limitée. Récemment, les zones littorales ont émergées comme étant de possibles points chauds pour la production de GES (Hofmann et al., 2010) et leurs émissions (Wang et al., 2006; Hofmann, 2013). Les sédiments côtiers reçoivent généralement plus de MO (à la fois autochtone et allochtone), ainsi plus de substrats sont disponibles pour la production de GES. Les sédiments sont aussi généralement plus chauds sur le littoral que dans la zone pélagique (Thebrath et al., 1993). Cela peut conduire à des taux proportionnellement plus élevés de la décomposition microbienne et à de plus forts gradients de GES entre les sédiments de surface et la fine couche d'eau du dessus. Les conditions environnementales côtières sont susceptibles d'être plus instables que celle du large, ce qui peut contribuer à une plus forte diversité de microbes responsables de la production de GES. Les sédiments littoraux sont plus exposés aux effets prolongés de la pression d'oscillations et aux grands courants oscillatoires dus aux ondes de surface (Hofmann et al., 2008). Bien que les changements de pression peuvent augementer les flux de gaz provenant des sédiments par un effet de pompage d'ondes (Precht et al., 2004), les courants oscillatoires peuvent avoir un effet similaire via leur réduction de l'épaisseur de la couche d'eau limite (Lorke et Peeters, 2006). En outre, la libération directe de l'eau interstitielle dans la colonne d'eau est possiblement suivie de la re-suspension des sédiments causée par les courants de cisaillement (Hofmann, 2007).

La zone littorale de nombreux petits lacs est occupée par une abondance de macrophytes. Les macrophytes affectent la résistance de la stratification thermique (Dale et Gillespie, 1977), les taux de remise en suspension des sédiments (Barko et James, 1998; Vermaat et al., 2000), et les échanges d'eau entre la zone littorale et la zone pélagique (Barko et James, 1998). Les macrophytes affectent les mouvements de sédiments, le cycle des nutriments et la dynamique de l'oxygène dans le sédiment, dans ce cas ils changent les conditions pour la production de GES, et peuvent agir comme des vecteurs pour le transport de gaz des sédiments vers la surface de l'eau et l'atmosphère. De plus, la décomposition de la litière de macrophytes peut accélérer l'établissement de conditions anoxiques et fournir des substrats facilement disponibles pour les méthanogènes (Weisner et al., 1994; Brix, 1997). Tous ces effets peuvent contribuer à des différences dans la dynamique des GES entre les zones occupées par des macrophytes et les zones libres des lacs. Leur développement au printemps et leur flétrissement vers la fin de l'été peut en outre contribuer à la répartition spatiale observée dans la production et les émissions de GES.

Il y a évidemment des variations temporelles dans la production de GES, leurs émissions et leur stockage, liées aux variations saisonnières et diurnes dans le mélange vertical et le flux d’énergie à l’interface eau-sédiments, et les changements saisonniers du taux de transfert horizontal entre le littoral et les zones pélagiques. Dans les lacs monomictiques et dimictiques, les GES stockés dans l’hypolimnion sont libérés principalement pendant les périodes de mélange des couches d’eau. La variabilité de la libération des GES et leur stockage dans les petits lacs peu profonds dépendent notamment de l’étendue de la stratification de la colonne d’eau. Dans les lacs turbides, la chaleur peut être facilement piégée dans les eaux de surface renforçant ainsi la stratification thermique et prévenir le mélange au printemps et en automne. Dans le cas des lacs plus clairs avec peu de phytoplancton et une abondance de macrophytes, la chaleur peut être répartie plus uniformément à travers la colonne d’eau et le mélange des couches se fera plus facilement. Cette différence dans le processus de libération des GES peut avoir de vastes répercussions lorsque l’on considère que de nombreux petits lacs de faible profondeur peuvent passer d’une eau claire (dominance de macrophytes), à une eau turbide et trouble (dominance du phytoplancton) dû à l’eutrophisation et le réchauffement climatique.

1.1.7 Les influences possibles des changements climatiques sur les processus de régulation des GES dans les lacs

Le climat influe sur le fonctionnement des petits écosystèmes lacustres, y compris les processus allochtones et autochtones, responsables de la production de GES, leurs émissions et leur stockage (Fig. 1.1.6). Les effets des changements climatiques sur les milieux terrestres peuvent inclure une productivité accrue, des taux plus élevés du turn-over de la matière organique dans les sols (Pare et al., 2006) et des changements dans les communautés végétales. Ceux-ci peuvent conduire à une augmentation du carbone exporté vers les lacs avec une augmentation de l’apport de CO_2 allochtone. Les changements climatiques, comme l’augmentation de la fréquence de la vague de chaleur estivale et des évènements extrêmes (tempêtes), peuvent entraîner une exportation plus élevée du COD provenant du bassin versant (Schindler et al., 1997). Le réchauffement du climat conduira à ce que de matière organique enfouie dans les sédiments des lacs soit minéralisée plus vite (Gudasz et al., 2010).

Le réchauffement climatique pronostiqué résultera en une plus forte stratification des petits lacs ce qui pourra conduire à une désoxygénation plus rapide et plus permanente des couches inférieures de la colonne d'eau (Moss et al., 2011). Des conditions du lac plus chaudes et plus réduites stimuleront la production de CH_4 et de N_2O et, après leur incomplète oxydation, stimuleront également leur libération vers la surface. Les énergies plus élevées requises pour l'activation de la méthanogénèse (Conrad et Wetter, 1990) peuvent être fournies aux sédiments lacustres étant donné que le réchauffement climatique conduit à des proportions plus importantes de la production primaire émises sous la forme de CH_4 (Yvon-Durocher et al., 2010).

De même, le réchauffement climatique pourrait avoir des effets prononcés sur le fonctionnement des réseaux trophiques aquatiques. Moss et al. (2011) suggèrent que de tels effets peuvent se manifester comme la croissance de la prédation des poissons conduisant à une diminution de la taille et de la capacité du zooplancton à contrôler le phytoplancton. Dans les lacs plus chauds, où les individus zooplanctoniques les plus larges ont été décimés par des poissons planctonivores, les fleurs d'eau d'algues sont plus susceptibles de se développer, avec une certaine prévalence de quelques espèces de cyanobactéries. Ceci peut être accompagné par une stratification d'autant plus forte de la colonne d'eau, avec une prolifération des fleurs d'eau d'algues à la surface piégeant la majeure partie de la chaleur, une anoxie dans les couches inférieures de la colonne d'eau, des taux plus élevés d'apports en nutriments et de plus fortes émissions en GES (CH_4, N_2O).

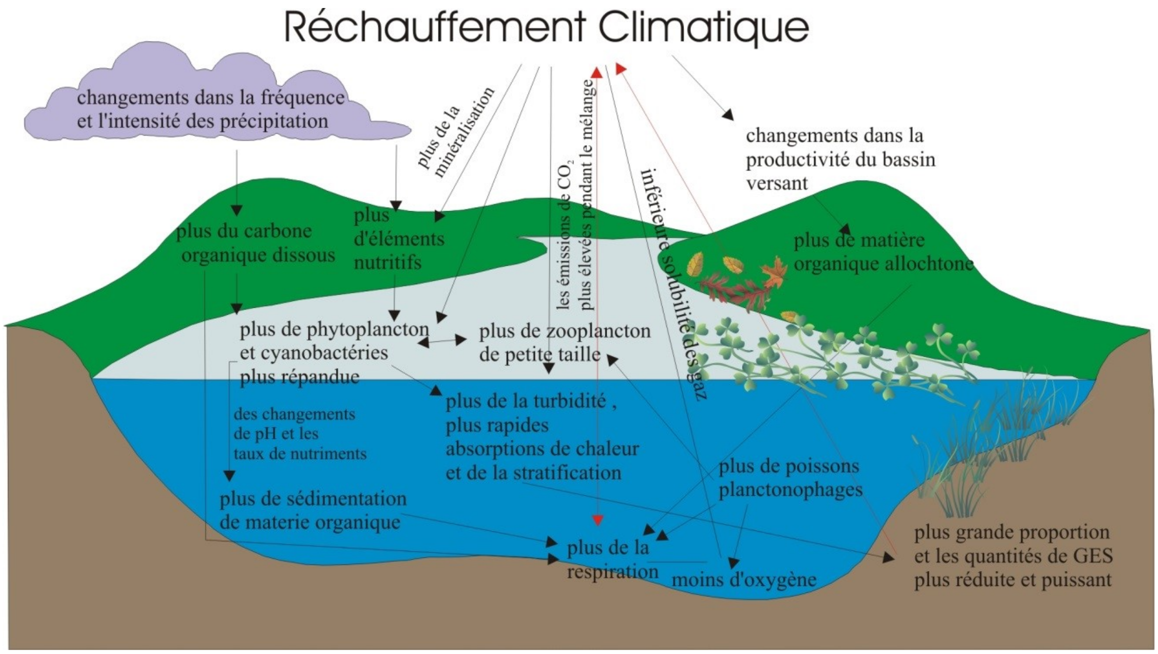

Figure 1.1.6 Les possibles effets des changements climatiques et de l'eutrophisation sur le fonctionnement des lacs peu profonds, tout en mettant l'accent sur l'interaction mutuelle entre ces deux processus (modifié de Moss et al., 2011).

1.2 Objectifs et Hypothèses

Les conditions limnologiques des petits lacs changent rapidement, affectant les taux, le moment et le mécanisme dominant de libération des GES. De plus, la méthode sélectionnée pour mesurer les flux peut affecter notre capacité à estimer de façon précise leur contribution au budget global des émissions de GES. L'objectif général de cette thèse est d'étudier l'interaction entre les caractéristiques limnologiques des lacs, les forçages météorologiques et les émissions de GES.

Chapitre 1. Les émissions et le stockage des GES dans un petit lac peu profond

Objectifs

1. Caractériser la variation saisonnière des flux diffusifs et d'ébullition du CO_2 et du CH_4 dans la zone pélagique d'un petit lac peu profond exposé à des conditions contrastées du climat.
2. Comparer l'ampleur et la variabilité des flux de GES estimés par les modèles basés sur le vent et le renouvellement de surface, relativement aux estimations de chambres flottantes.
3. Décrire la variabilité temporelle de la distribution verticale du CH_4 aqueux en réponse aux conditions météorologiques variables.
4. Caractériser la saisonnalité du stockage des GES dans un petit lac en fonction de l'étendue et de la force de la stratification de la colonne d'eau.

Hypothèses

1. Les petits lacs eutrophes peuvent à la fois agir comme un puits et comme une source de CO_2 selon les conditions de pluie et le niveau de stratification de la colonne d'eau.
2. Les flux de CO_2 et de CH_4 dans un petit lac varient sur une échelle de temps saisonnière et interannuelle selon les conditions météorologiques.
3. L'approche choisie pour estimer le coefficient de transfert de gaz affecte de manière significative l'ampleur et la variabilité des flux de GES dans les lacs.

Chapitre 2. Effets des vagues de chaleur sur l'oxygène, les nutriments et le potentiel de réchauffement climatique d'un lac peu profond et de petite taille

Objectifs

1. Caractériser les effets de la vague de chaleur estivale sur l’oxygène et les nutriments et leur distribution verticale dans un petit lac peu profond.
2. Caractériser les effets des régimes de précipitations sur les concentrations en nutriments et sur la biomasse du phytoplancton.
3. Décrire les différences spatiales du mélange des couches d’eau, de l’oxygène, des nutriments et de la biomasse du phytoplancton entre les zones littorales (avec des macrophytes) et les zones pélagiques (sans macrophytes).
4. Étudier la relation entre l’oxygène, les nutriments et la biomasse du phytoplancton, et les concentrations de CO_2, CH_4 et N_2O, ainsi que le couplage (ou découplage) entre les différents GES.
5. Suivre les variations saisonnière et spatiale du potentiel de réchauffement climatique (PRC) dans un petit lac.

Hypothèses

1. Un été exceptionnellement chaud et sec conduit à une stratification prononcée de l’oxygène dans un petit lac.
2. Pendant la vague de chaleur estivale, l’anoxie hypolimnique apparaît dans la zone pélagique mais pas sur la zone littorale (avec macrophytes).
3. La vague de chaleur estivale entraîne de plus fortes concentrations en nutriments totaux mais des concentrations plus faibles en nutriments dissous dans l’épilimnion comparé à un été pluvieux précédent.
4. Contrairement à des conditions bien mélangées, une stratification stable pendant l’été conduit à l’apparition de points chauds et de moments chauds dans la saturation en GES et PRC dans un petit lac.

Chapitre 3. Effet de la structure du réseau alimentaire planctonique et des fleurs d'eau d'algues sur les émissions de gaz à effet de serre par un lac hypereutrophe

Objectifs

1. Caractériser les dynamiques temporelles et spatiales de la saturation des GES (CO_2, CH_4, N_2O) et leur stockage dans un petit lac hyper-eutrophe souffrant d'une récurrence d'apparition de fleurs d'eau phytoplanctoniques.
2. Décrire les effets directs et indirects des fleurs d'eau de phytoplancton sur les concentrations de GES.
3. Décrire les effets de la composition des fleurs d'eau phytoplanctonique sur les concentrations de GES et leur stockage.

Hypothèses

1. Les émissions et les flux de GES varient selon une échelle de temps saisonnière et interannuelle dans les zones pélagiques et littorales dans un petit lac hyper-eutrophe.
2. Il y a un effet direct d'une fleur d'eau phytoplanctonique sur les concentrations de GES dans l'eau du lac.
3. La composition des fleurs d'eau de cellules phytoplanctoniques affecte la quantité et les rapports entre différents GES dans la colonne d'eau.
4. Le zooplancton joue un rôle dans la régulation des concentrations de GES à travers leurs effets sur le réseau trophique et le cycle de la matière organique.

1.3 Sites étudiés

Le Lac Jacques est un petit lac (0.18 km^2), peu profond (profondeur maximale = 1,9 m, profondeur moyenne = 0,75 m) situé à 30 km au nord de la ville de Québec (QC, Canada). Ce les fourni de l'eau à partir de deux ruisseaux dans la partie orientale du lac, ainsi que d'une source d'eau souterraine dans sa partie sud. Le lac peut être classé comme étant méso-eutrophe en termes de phosphore total (valeur moyenne d'été de 21 µg L^{-1}), d'azote total (0,7 mg L^{-1}), et de concentrations en Chl-*a* (17 µg L^{-1}). Environ cinquante pour cent de la surface du lac est recouverte de la macrophytes *Brasenia schreberi* en été. Les 10 cm supérieurs des sédiments ont une forte teneur en carbone organique (perte par le feu = 38 ± 5%, n = 10). Les temps de rétention d'eau moyens sont 14-20 jours en été. La concentration de carbone organique dissous varie entre 2,2 à 3,0 mg L^{-1}.

Le Lac St Augustin est aussi relativement petit (0,62 km^2). C'est un lac urbain situé à la périphérie de la ville de Québec (Canada), le long de la rive nord du fleuve St-Laurent. Le lac est alimentés en eau à partir de deux petites entrées de surface et des sources souterraines, avec un temps de renouvellement de l'eau d'environ 6 mois (Roberge et al., 2002). Le lac St-Augustin peut être classé comme eutrophe à hypereutrophes en termes de nutriments et de Chl-a (40 µg L^{-1}), et stocke des quantités relativement élevées de matière organique dans les sédiments de surface (LOI = 18 ± 7%, n = 10). Au cours des trois derniers siècles, le lac était sous la forte incidence du développement anthropogénique et de l'urbanisation (Pienitz et al., 2006). Le développement de l'agriculture et de la construction d'autoroutes à proximité ont entraîné une forte eutrophisation au milieu du XXème siècle, et des proliférations d'algues persistantes estivales ont été observées depuis 1970. Dans les années chaudes, les fleurs d'eau se composent presque exclusivement de cyanobactéries planctoniques (Deshpande et al., 2014), tandis que les dinoflagellés et diatomées peuvent devenir abondants en d'autres années. Malgré l'abondance du phytoplancton, les zones peu profondes (moins de 2 m de profondeur) du lac sont occupées par de denses lits de macrophytes submergés.

1.4 Méthodologie

L'approche méthodologique globale est décrite ci-dessous, alors que certains détails sur les campagnes de terrain et la modélisation des échanges gazeux sont donnés dans les sous-sections plus basses. L'échantillonnage sur le terrain a été mené pendant la saison d'eau libre entre 2011 et 2012 sur deux lacs de petite taille (surface < 1 km^2) et peu profonds (profondeur < 5m). Un large éventail de caractéristiques météorologiques et limnologiques ont été échantillonnées, incluant la vitesse des vents, les précipitations, la température de l'air, l’humidité relative, les profils verticaux de pH, d’oxygène et de température, les concentrations en nutriments solubles et totaux, ainsi que l’abondance et la taxonomie du phytoplancton et du zooplancton. Les GES dissous ont été échantillonnés dans les zones littorales et pélagiques des deux lacs en utilisant la méthode d’échange gazeux. Le mélange et la structure physique des lacs ont été suivis à l'aide de chaînes de thermographes installées dans la zone pélagique afin de calculer les flux de gaz. Les modèles de renouvellement de surface et basés sur la vitesse du vent ont été appliqués puis les résultats comparés aux flux de gaz obtenus directement à l'aide d'une chambre flottante. Les flux d'ébullition ont été obtenus en utilisant des entonnoirs inversés submergés dans les zones pélagiques et littorales des lacs. Le stockage de GES près du fond des lacs a été calculé à l'aide de profils verticaux des gaz dissous dans la zone littorale et pélagique et la bathymétrie.

1.4.1 Les campagnes de terrain

Les caractéristiques limnologiques du lac Jacques, incluant la température de surface, la conductivité, l’oxygène dissous (DO), les éléments nutritifs des eaux de surface et la chlorophylle-a, ont été évaluées au cours de la saison des eaux libres (Mai-Octobre) en 2011 et 2012. En 2011, les données horaires sur la température de l’air, l’humidité relative, la vitesse du vent, la direction du vent, le point de rosée, et les précipitations ont été obtenues à partir de la station météorologique d'Environnement Canada située à 7 km du lac. En 2012, une station météorologique WeatherHawk 511 montée à 25 m de la rive du lac et à 7 m au-dessus du niveau de l'eau, a enregistré la température de l'air, l'humidité relative, la vitesse du vent et la direction du vent toutes les 30 minutes. Les données de l'éclairement solaire pour la période étudiée ont été obtenues à partir de la

NASA (Outil interactif Giovanni pour la visualisation des données et de l'analyse). Les chaînes de thermographes étaient amarrées dans les zones pélagique (2011 et 2012) et littorale du lac (2012 seulement). Les conditions météorologiques ont été remarquablement différentes entre les deux années étudiées, nous offrant l'occasion de comparer les effets de conditions contrastées sur les flux de GES dans ce petit lac peu profond.

Les profilages des concentrations de GES (CO_2, CH_4 et N_2O) ont été effectués pendant l'intégralité de l'étude aux zones littorale et pélagique du lac en utilisant la méthode d'échange gazeux développée par Hesslein et al. (1991). En bref, 2 litres d'eau du lac sont équilibrés avec 20 ml de l'air ambiant. Le volume d'air est ensuite injecté dans un flacon Labco préalablement évacué et rincé avec de l'hélium. Les échantillons de gaz ont été analysés par un chromatographe en phase gazeuse équipé d'un détecteur FID ou DPE. Les concentrations de GES dissous à la surface du lac ont été calculées à l'aide de la loi de Henry, et comparées à la concentration des gaz en équilibre avec l'atmosphère (degré de saturation). Ces valeurs ont été utilisées pour les estimations des flux diffusifs de gaz, que ce soit pour le modèle de renouvellement de surface ou pour les modèles basés sur le vent. Pour évaluer le stockage des GES au lac Jacques, la masse totale de CO_2 et de CH_4 au-dessus (ou en dessous) de la saturation a été calculée en multipliant la concentration moyenne du gaz sur un intervalle de profondeur donné par le volume d'eau dans cet intervalle, et en sommant sur toute la profondeur du lac (Rudd et Hamilton, 1978).

Simultanément au profilage des GES, une chambre flottante a été déployée à la surface du lac. La chambre a été construite à partir de plastique en PVC de 10 mm d'épaisseur, équipés de flotteurs répartis uniformément sur la paroi latérale, dont la paroi était submergée de 4 cm sous l'eau. Le volume d'air de la chambre était relié au détecteur de CO_2 EGM4 (PP-Systems). Cela a permis d'évaluer directement les flux de CO_2 (ainsi que le coefficient k_{600}) qui ont été comparés aux flux de GES estimés avec les modèles de renouvellement de surface et basés sur le vent. Des mesures simultanées permettent de minimiser les effets possibles de la variabilité spatiale des concentrations de GES (Heiskanen et al., 2014) et d'éviter de grandes différences entre les flux mesurés et estimés (Schubert et al., 2012). Les flux de GES émis par ébullition ont été mesurés à l'aide d'entonnoirs inversés submersibles qui ont permis de recueillir les bulles de gaz dans une seringue située au sommet du cône. Cette approche de mesures directes et modélisées des flux de diffusion et d'ébullition a permis de détecter des différences entre la spéciation des GES et leurs voies d'émission pendant deux années présentant des conditions climatiques contrastées.

Une approche méthodologique similaire a été utilisée au lac St-Augustin où les caractéristiques limnologiques et les concentrations de GES ont été évaluées tous les quinze jours ou chaque semaine dans les zones pélagique et littorale du lac durant deux étés (2011-2012). Les profils de GES (CO_2, CH_4 et N_2O) de la surface jusqu'au fond à des intervalles de 1 m ont été mesurés afin d'évaluer le stockage de gaz dans le lac. La station météorologique d'Environnement Canada a fourni des données horaires sur les conditions météorologiques. De plus, l'échantillonnage au lac St-Augustin incluait l'évaluation de la structure de la communauté planctonique. En bref, à chaque date de prélèvement, l'eau non filtrée a été recueillie dans des bouteilles de 2L opaques pour évaluer l'abondance et le biovolume du picoplancton par cytométrie de flux, et le biovolume des plus grandes cellules de phytoplancton par microscopie inversée. Le zooplancton a été recueilli avec un filet à plancton conique (taille des mailles de 63 µm; diamètre de 20 cm) qui a été remorqué verticalement à travers la colonne d'eau dans les stations d'échantillonnage littorales et pélagiques.

1.4.2 Le mélange du lac, les échanges de chaleur et la modélisation des échanges gazeux

Les flux de chaleur sensible et de chaleur latente utilisés dans le modèle de renouvellement de surface ont été calculés en utilisant la formule aérodynamique de Macintyre et al. (2002). La méthode d'Imberger (1985) a été utilisée pour évaluer la dynamique diurne de la couche de mélange, alors que le flux de chaleur surfacique a été calculé selon la méthode de Kim (1976). Le flux de poussée nécessaire pour les estimations du taux de dissipation de l'énergie cinétique a été calculé suivant Imberger (1985) et MacIntyre et al. (2002, 2009). La vitesse de pénétration convective a été calculée suivant Deardoff (1970) et MacIntyre et al. (2002). Pour les détails sur les calculs spécifiques, voir l'Annexe 4.1. Le coefficient de transfert de gaz k a été calculé en utilisant le modèle de renouvellement de surface développé par MacIntyre et al. (1995) et Zappa et al. (2007) (ci-après SC) suivant l'équation:

$k_{600} = a_1 (\varepsilon/v)^{1/4} Sc^{-1/2}$

où a1 est une constante expérimentale (fixée à 0,5 dans cette étude), ε est le taux de dissipation de l'énergie cinétique, et v la viscosité cinématique. Nous avons utilisé la similitude d'échelle de Tedford et al. (2014) pour calculer les taux de dissipation d'énergie pendant le refroidissement grâce à l'équation:

$\varepsilon = 0.56\, u_{*w}^3/kz + 0.77\beta$

et pendant le réchauffement avec:

$\varepsilon = 0.6\, u_{*w}^3/Kz$

où u*w est la vitesse de frottement de l'eau calculée à partir du stress de cisaillement par le vent, K la constante de Von Karman, z la profondeur (égale à 15 cm), et β le flux de poussée en surface.

Pour estimer les flux de diffusion, nous avons sélectionné les modèles basés sur le vent de Cole et Caraco (1998, ci-après CC), et de Crusius et Wanninkhof (2003, ci-après CW). Ces deux modèles sont basés sur des expériences effectuées sur les lacs de petites et moyennes tailles (0.13 à 1.35 km^2), mais l'équation de Cole et Caraco résultante a été développée en incluant des données issues d'expériences par gaz traceurs sur des grands lacs (par exemple le lac Pyramide, 500 km^2). L'équation de Cole et Caraco s'applique pour des vents allant jusqu'à 9 m s^{-1}, tandis que celle de Crucius et Wanninkhof s'applique uniquement à des vents allant jusqu'à 6 m s^{-1} et a été forcée à 0 pour les vents de 0 m s^{-1}. Cependant, les auteurs signalent que pour des vents d'une vitesse inférieure à 3,7 m s^{-1}, k_{600} est indépendante de la vitesse du vent et montrent des écarts considérables à des faibles vitesses de vents.

1.5 Résultats et discussion

Cette section résume les résultats les plus importants présentés dans chaque chapitre de la thèse. Pour une description plus détaillée des résultats et une discussion approfondie de leur importance, veuillez vous référer aux chapitres 2, 3 et 4.

1.5.1 Article 1 intitulé "Émissions de gaz à effet de serre et leur stockage dans un lac peu profond et de petite taille"

À l'inverse de la conception générale selon laquelle les petits lacs possèdent des couches considérablement bien mélangées, le lac Jacques a montré une stratification thermique persistante en 2011 et 2012. La stratification a été plus forte pendant l'été chaud et sec de 2012 que pendant l'été pluvieux et froid de 2011. Par exemple, les différences de température entre la surface et le fond ont atteint 14°C en 2012, mais seulement 8°C en 2011. La stratification peut être quantifiée par la fréquence de Brunt-Väisälä (N). En 2011, le métalimnion débutait entre 0.8-1.0 m de profondeur et s'étendait jusqu'au fond du lac, pour des valeurs de N atteignant 60 cycles par heure (cph). En revanche, pendant l'été caniculaire de 2012, le métalimnion débutait entre 0.3-0.6 m et s'étendait jusqu'à 0.8 m, avec des valeurs de N atteignant près de 100 cph. Il est intéressant de considérer que les océanographes supposent qu'une colonne d'eau est fortement stratifiée quand N atteint des valeurs supérieures à 20 cph.

Les différences dans la stratification et les précipitations entre ces deux années ont conduit à des différences dans la distribution verticale des gaz dissous. En 2011, le CO_2 a augmenté après les orages, avec des indications d'intrusions d'eau autant dans les couches situées au milieu de la colonne d'eau ou près du fond. Les eaux de surface ont été continuellement supersaturées en CO_2 et le CH_4 ne s'est pas accumulé dans la couche d'eau inférieure de la colonne d'eau. En revanche, l'eau en surface était sous-saturée en CO_2 au milieu de l'été 2012, tandis que le CH_4 s'est accumulé dans la partie inférieure de la colonne d'eau en juillet et en août.

La comparaison entre les méthodes utilisées pour l'estimation des flux de diffusion a révélé que le modèle basé sur le renouvellement de surface (SC) montrait des flux de GES supérieurs aux modèles basés sur le vent (CW et CC). Les flux estimés en utilisant SC étaient mieux corrélés aux mesures directes issues des chambres flottantes que les estimations obtenues par les modèles basés sur le vent, avec un écart maximal de 39%. La plus grande variance obtenue avec le modèle CW (loi de puissance possédant une courbe plus abrupte par rapport à celle utilisée pour le modèle CC) peut limiter l'intéret de ce modèle pour des estimations précises et une mise à l'échelle des flux de GES dans les lacs. Les flux diffusifs de CO_2 estimés avec le modèle SC variait en 2011 de 8 à 89 mmol m^{-2} d^{-1} et de -20 à 97 mmol m^{-2} d^{-1} en 2012. Les flux de CH_4 variaient entre 0.4 et 1.7 mmol m^{-2} d^{-1} en 2011 et entre 0.4 et 14.4 mmol m^{-2} d^{-1} en 2012. Ces valeurs sont comparables aux flux mesurés par Natchimuthu et al. (2014) variant entre -6.4 et 31.8 mmol CO_2 m^{-2} d^{-1} et entre 3.3 et 15.1 mmol CH_4 m^{-2} d^{-1} pour un petit étang peu profond en Suède, ou encore à ceux de Huttunen et al. (2003) variant entre -1.8 à 32.0 mmol CO_2 m^{-2} d^{-1} et entre 0.09 et 8.3 mmol CH_4 m^{-2} d^{-1} dans un ensemble de petits lacs boréaux.

Un petit lac peut être polymictique ou il peut rester stratifié de manière stable pendant une grande partie de l'été. Mes travaux démontrent que la fréquence des pluies et la température moyenne ont eu un effet modérateur sur la spéciation, le stockage et les émissions de GES dans un lac peu profond de petite taille. La variabilité intra et inter annuelle des émissions de GES dépend des conditions météorologiques et de la méthode choisie pour les estimations des flux diffusifs. Les modèles basés sur le vent (CW) génèrent une forte variabilité dans les estimations et peuvent conduire à un biais dans la contribution des lacs au cycle planétaire du carbone. Les changements climatiques provoquent une fréquence plus élevée d'étés chauds et secs (vagues de chaleur) accompagnés de pluies soudaines et intenses, pouvant conduire à des différences quant aux voies d'émission de GES et au cycle du carbone dans les lacs peu profonds.

1.5.2 Article 2 intitulé "Effets des vagues de chaleur sur l'oxygène, les nutriments et le potentiel de réchauffement climatique d'un lac peu profond et de petite taille".

La vague de chaleur estivale de 2012 a généré une stratification persistante de l'oxygène et une anoxie dans la couche inférieure de la colonne d'eau du lac Jacques. Il y avait moins d'oxygène dans l'hypolimnion pendant l'été caniculaire que pendant l'été précédent (en moyenne 4,8 mg L^{-1} et 8,2 mg L^{-1}, respectivement). Pendant la vague de chaleur, une pluviométrie réduite, une diminution du mélange et une plus faible disponibilité en oxygène ont causé des différences dans la disponibilité des nutriments, mais également une perturbation du cycle du carbone par rapport à l'été précédent. Au cours de la période de stratification, les concentrations en phosphore total (TP) et azote totale (TN) étaient plus élevées en 2012 qu'en 2011. En revanche, il y avait beaucoup moins de SRP et NO_3^- en 2012 qu'en 2011. Les fleurs d'eau d'algues, constituées en partie d'espèces cyanobactériennes en 2012, ont montré des concentrations de Chl-a atteignant jusqu'à 65 µg L^{-1}. Le niveau de saturation en GES était corrélé avec la biomasse du phytoplancton, la température de l'eau et la concentration en nutriments (TP).

Le potentiel de réchauffement climatique (PRC, en équivalent CO_2) des GES dissous (CO_2, CH_4 et N_2O) en 2011 était comparable pendant tout l'été, avec seulement une légère augmentation du PRC dans les zones pélagiques lorsque le lac se mélangeait. En revanche, la vague de chaleur de l'été de 2012 a entraîné des moments chauds d'émission dans les zones pélagiques et littorales du lac. En 2011, le PRC a atteint des valeurs maximales de 8.9 et 14.1 × 10^3 mol CO_2 d^{-1} dans les zones littorale et pélagique du lac, respectivement. En 2012, le PRC a représenté un moment chaud en septembre 2012 où il a atteint 33.1 et 41.0 × 10^3 mol CO_2 d^{-1} dans les zones littorale et pélagique, respectivement. Pour sa part, le PRC des GES émis par ébullition était significativement plus élevé en 2011 qu'en 2012 (14.0 comparativement à 7.1 × 10^3 moles de CO_2 d^{-1}).

Mes résultats montrent les effets prononcés de la vague de chaleur estivale sur le fonctionnement de l'écosystème d'un petit lac, avec notamment un fort impact sur les concentrations de GES, leur spéciation et leurs voies d'émission. Les flux diffusifs plus élevés de l’année caniculaire étaient accompagnés par une baisse des flux par ébullition. Contrairement à l'opinion généralement acceptée, le réchauffement climatique ne mène pas nécessairement à une hausse du PRC dans le cas des petits lacs peu profonds où la stratification est plus intense et les couches inférieures de la colonne d'eau demeurent plus froides pendant l'été. La présence de macrophytes dans la zone littorale du lac a entraîné des émissions de GES plus élevées pendant un moment chaud en 2012. Pour estimer de façon précise les émissions de GES et faire des prévisions sur les effets futurs du réchauffement climatique, les différentes voies d'émission devront être évaluées. Si la méthanogénèse est stimulée par l'augmentation de la température (Yvon-Durocher et al., 2011), cela ne conduit pas nécessairement à des concentrations plus élevées en CH_4 à la surface du lac si la colonne d'eau reste bien oxygénée et l'activité des méthanotrophes est stimulée.

Au lac Jacques, l'ébullition des GES était significative en 2011 et 2012, mais les flux diffusifs étaient beaucoup plus importants en 2012 (ils contribuaient 10 fois plus au PRC pendant la période de retournement automnal). En effet, l'accumulation des trois GES dans l'hypolimnion pendant la vague de chaleur estivale a causé par la suite un moment chaud d'émission pendant la période retournement automnal, avec des concentrations atteignant les valeurs les plus élevées jamais signalées dans les eaux de surface des lacs tempérés (par ex. Huttunen et al. 2003). Par conséquent, 70% des émissions de GES en eau libre ont été observées au cours de l'automne suivant la vague de chaleur. Ce résultat est en accord avec ceux d'Encinas-Fernandez et al. (2014) montrant que la fraction de CH_4 stockée puis émise par diffusion pendant le retournement automnal est largement plus grande que celle émise durant le mélange printanier et la période estivale stratifiée. Nos résultats indiquent également que l'hétérogénéité temporelle des émissions de CO_2, CH_4, et N_2O peut augmenter suite au réchauffement climatique.

Le «point de basculement des flux d'énergie» d’un moment chaud a été atteint plus tôt dans la zone littorale après un orage estival (2012). En revanche, dans la zone pélagique, les moments chauds se sont produits lorsque les pluies intenses de la mi-septembre ont été suivies par une diminution des températures de l’air menant à de grands flux de stockage pour les trois GES. Les concentrations en CO_2 et CH_4 des eaux de surface étaient en partie expliquées par la variabilité dans la biomasse phytoplanctonique. L'absorption par l'activité photosynthétique des algues explique sa relation avec le CO_2, alors que la relation avec le CH_4 pourrait être expliquée par la production de CH_4 dans la colonne d'eau associée aux cellules phytoplanctoniques qui produisent des composés labiles pour les méthanogènes (Grossart et al., 2011; Tang et al., 2014). Même si ce phénomène est potentiellement significatif, il est probablement secondaire par rapport à la production anoxique de CH_4 dans les sédiments lacustres (Bogard et al., 2014), stimulée par la sédimentation de matière organique suivant le déclin d'une fleur d'eau (Fig. 2.2.S3).

1.5.3 Article 3 intitulé "Effet de la structure du réseau alimentaire planctonique et des fleurs d'eau d'algues sur les émissions de gaz à effet de serre par un lac hypereutrophe"

En 2011 (une année pluvieuse), la flore du lac St-Augustin était principalement dominée par des dinoflagellés (*Glenodinium sp.*), avec une fleur d'eau massive culminant en septembre avec des concentrations en Chl-a allant jusqu'à 55 µg L^{-1}. En 2012 (une année chaude et sèche), le lac a subi une importante efflorescence de cyanobactéries en été, incluant des espèces de cyanobactéries toxiques (*Aphanizomenon* sp. et *Anabaena* sp.), avec des concentratrions en Chl-*a* atteignant 60 µg L^{-1}. Suivant l'établissement des fleurs d'eau, les eaux de surfaces devenaient sous-saturées en CO_2 de façon périodique (2011) ou persistante (2012). Cette sous-saturation persistante en CO_2 lors de l'été caniculaire était accompagnée de fortes concentrations en CH_4 (0.36 comparé à 0.27 µM en 2011) et de faibles concentrations en N_2O (0.8 comparativement à 1.2 nM en 2011).

Le lac St-Augustin est polymictique mais il présentait un gradient vertical de GES (Fig 4.S1), avec des accumulations périodiques près du fond. Les variations saisonnières et interannuelles dans le stockage des GES dans la colonne d'eau du lac suivaient le même patron que les concentrations en surface dans le cas du CO_2 et du CH_4, mais pas dans le cas du N_2O. Contrairement aux niveaux de saturation mesurés en surface du lac, la colonne d'eau entière du lac (les 2 zones additionnées) contenait plus de N_2O durant l'année la plus chaude (5.9 moles comparé à 4.2 moles en 2011). Une analyse en composantes principales (ACP) sur les données de la zone pélagique révèle que les niveaux de saturation en CO_2 étaient le mieux expliqués par la température, la vitesse du vent et la Chl-a (PC1; $R^2 = 0.51$). Le niveau de saturation en CH_4 était le mieux expliqué par les mêmes 3 variables (PC1; $R^2 = 0,60$), ainsi que par la biomasse du picoplancton et les nutriments (PC2). La saturation en N_2O était le mieux expliquée par la température de surface et la concentration en O_2 (PC1; $R^2 = 0.20$). L'analyse de régression linéaire a révélé des relations significatives entre les différentes composantes du réseau trophique planctonique et les GES. Il y avait des effets positifs des diatomées et du zooplancton sur le CO_2, et des effets négatifs des Dinophyceae. Pour le CH_4, il y avait des effets positifs des Chlorophyceae, des cyanobactéries et des Dinophyceae, et un effet négatif de la biomasse des bacteria. Enfin, il y avait un effet positif des Cryptophyceae sur le N_2O, et un effet négatif des cyanobactéries et des bacteria. De façon globale ces résultats suggèrent que la structure thermique et la biomasse et la composition du plancton sont fortement liées à la concentration des GES.

L’approche choisie dans mon étude a permis d'étudier les effets des fleurs d'eau d'algues sur les autres composantes du réseau trophique planctonique et sur la saturation et l’accumulation des GES dans la colonne d’eau d’un lac hypereutrophe. Sous des conditions météo plus chaudes et sèches, les lacs eutrophes et hypereutrophes sujet à de fortes efflorescences cyanobactériennes peuvent devenir des puits efficaces du CO_2 atmosphérique. La majorité des lacs autour du monde sont présentés comme des sources importantes de CO_2 par Cole et al. (1994; 2007) et Sobek et al. (2005). Toutefois dans la majorité des cas, les lacs ont été échantillonnés durant l'automne ou l'hiver. Un échantillonnage limité à cette période de l'année réduit la chance d'observer la sous-saturation en CO_2 associée à la croissance du phytoplancton. Les concentrations en CO_2 au lac St-Augustin, de la même façon que dans plusieurs autres lacs productifs (Balmer et Downing, 2011), sont tombées bien en-dessous du niveau de saturation durant les épisodes de fleurs d'eau, en particulier durant les fleurs d'eau de cyanobactéries. Je postule que les petits lacs eutrophes absorbe de larges quantités de CO_2, peuvent relâcher ce carbone sous la forme de CH_4. Cela pourrait également accélérer la minéralisation du carbone allochtone à travers le "priming effect" (Guenet et al., 2010).

Les 3 GES étaient fortement reliés à la communauté planctonique du lac St-Augustin. Les relations inverses entre le CO_2 et les fleurs d'eau ou le picoplancton s'expliquent par l'activité photosynthétique. En contrepartie, l'augmentation synchrone entre le CO_2 et les diatomées indiquent que ce groupe surpasse les autres groupes lorsque la colonne d'eau est bien mélangée, la météo est pluvieuse et venteuse, et les GES stockés dans la colonne d'eau sont relâchés vers l'atmosphère. La forte relation inverse entre le picoplancton et le N_2O suggère sa consommation par les bactéries ou sa réduction plus efficace associée à une plus grande activité bactérienne. Cette consommation a pu se produire dans les microhabitats anoxiques de l'épilimnion. Contrairement au CO_2 et N_2O, le CH_4 était corrélé positivement à la biomasse du phyto et du picoplancton. Les fleurs d'eau mènent à une augmentation de la déposition de matière organique au fond du lac. Le CH_4 peut avoir été produit en anaérobie dans les microzones anoxiques des amas de matière organique qui sédimentent (Schulz et al., 2001) ou en présence d'oxygène en association avec le picoplancton autotrophe (Grossart et al., 2011).

Peu d'études ont quantifié les changements saisonniers des concentrations en GES dans les petits lacs eutrophes subissant des fleurs d'eau (Duarte et Prairie, 2005; Downing et al., 2008), encore moins en mesurant les 3 GES de façon simultanée. Ces écosystèmes sont surveillés de près étant donné leur eutrophisation anthropique accélérée, mais ils ont encore très peu été étudiés du point de vue de leur rôle dans le cycle global du carbone. Mes résultats montrent comment l'occurrence et la persistance d'une fleur d'eau liée à un été caniculaire ont modifié la structure du réseau trophique planctonique et ont mené vers une plus forte émission de CH_4, au détriment des émissions de CO_2. Cette relation peut avoir des conséquences fondamentales dans notre compréhension du rôle des lacs sur le climat global, et procurer un argument supplémentaire pour restaurer sans tarder les lacs pollués et subissant une eutrophisation accélérée.

1.6 Limites des études et perspectives futures

Les concentrations et flux de GES dans les lacs peuvent varier grandement sur l'échelle spatiale et temporelle (Schmid et al., 2007; Hofmann et al., 2010). Mon approche au lac Jacques inclut l'échantillonnage des gaz dissous à plusieurs reprises sur 3 à 6 sites dépendant de l'année, pour un total de 33 mesures (incluant l'échantillonnage pendant la nuit; Annexe Fig. 4.2.1). Cependant, la principale limitation de notre étude demeure cette résolution spatio-temporelle relativement faible étant donné la grande variabilité pouvant exister. La distribution hétérogène des gaz dissous peut notamment résulter du transport diffusif et advectif entre les zones peu profondes près du rivage et les zones d'eau libre (Hofmann, 2013). Dans les petits lacs peu profonds, un tel transport peut se produire sur une courte période de temps, c'est pourquoi l'estimation manuelle et ponctuelle des concentrations en GES n'est pas idéale pour évaluer les différences sur un court délai entre les zones littorales et pélagiques. Le développement récent de senseurs automatisés pour le monitorage des concentrations de CO_2 (par ex. le senseur Vaisala; Jackowicz-Korczyński et al., 2010) et de CH_4 (par ex. le senseur Panterra, Neodym Technologies, Laurion et al., 2010; ou des senseurs plus précis mais plus chers) pourrait permettre une évaluation plus précise des GES dans la colonne d'eau et les sédiments, et ainsi mieux évaluer le transport horizontal du littoral vers la zone pélagique puis son relâchement vers l'atmosphère lors d'une remontée d'eau (*upwelling*). De telles données permettraient également de mieux comprendre et modéliser les effets des facteurs météorologiques et des processus physiques sur ces échanges.

La seconde limitation de cette étude concerne les données temporellement et spatialement des flux d'ébullition. En effet, j'ai installé 4 à 10 entonnoirs par lac permettant un total de 54 mesures de flux d'ébullition à travers deux saisons d'eau libre, ce qui est déjà bien supérieur à ce qui a été publié dans plusieurs autres études (en fait, la plupart des études n'ont pas mesuré de flux d'ébullition). La majorité des études sur l'ébullition utilisent des entonnoirs déployés sur une courte période de temps (incluant mon étude, de 6 à 24 heures), et avec une faible fréquence d'échantillonnage. Avec cette approche, il est probable que des événements d'ébullition intenses et soudains soient manqués. Ce problème peut être contourné par le déploiement de pièges à bulles automatisés permettant de connaître le patron temporel des événements d'ébullition (Juutinen et al. 2004). Lorsque ces pièges automatisés sont couplés à des détecteurs de GES (quantification), ils permettent une détermination complète des flux de gaz (Walter et al. 2010). Néanmoins, les effets du déploiement de gros appareils sur la turbulence et ainsi sur notre capacité d'estimer les flux avec précision demeurent inconnus, en particulier dans le cas de petits plans d'eau peu profonds. Par ailleurs, de tels instruments étant couteux, ils ne permettent pas d'adresser l'hétérogénéité spatiale de l'ébullition. Finalement, il est nécessaire de choisir entre un déploiement à un seul endroit (précision, compréhension des différences spatiales) ou si on permet aux entonnoirs de bouger librement au-dessus du lac pour estimer un flux moyen (mise à l'échelle).

Des incertitudes persistent sur les meilleures méthodes devant être utilisées pour calculer les flux diffusif des gaz dans les petits lacs. Dans le cadre de ma thèse, mon objectif était de comparer des modèles basés sur le vent au modèle plus complet de renouvellement de surface (incluant les effets du vent et des échanges thermiques), et de comparer ces estimations aux mesures directes obtenues à l'aide de chambres flottantes. Nous avons conclu que pour obtenir une meilleure congruence avec les mesures directes des flux, les composantes convectives de la turbulence près de la surface doivent être considérées dans la formulation de k (modèle de renouvellement de surface). Cependant, la contribution de la convection à la turbulence peut dépendre de la taille et de la forme du lac tel que démontré par Read et al. (2012) (voir aussi Vachon et Prairie, 2013). Mes données suggèrent également que cela peut dépendre de la variabilité interannuelle dans le forçage météorologique. Les mesures directes de la turbulence à la surface du lac, par exemple à l'aide d'un vélocimètre acoustique Doppler, permettraient de développer des modèles d'échange gazeux plus appropriés aux conditions physiques spécifiques des lacs peu profonds et de petite taille, et de mieux évaluer les effets du réchauffement climatique sur les facteurs contrôlant la turbulence et les flux de gaz.

De futurs travaux devraient aussi se concentrer sur les effets indirects du réchauffement climatique sur la production et l'émission des GES par les petits lacs, notamment les effets reliés au changement du niveau de l'eau, au recouvrement par les macrophytes et à la structure du réseau trophique. Les effets de la réduction du niveau de l'eau sur la production et l'émission de GES peut agir via un apport d'énergie plus élevé à la surface des sédiments donnant lieu à des flux d'ébullition plus élevés (Wik et al. 2013), et une diminution de la profondeur de la colonne d'eau pourrait mener à de plus faibles taux d'oxydation des GES, dépendamment des profils en oxygène. Un changement dans l'abondance et la composition de la communauté des macrophytes suivi de changements dans le niveau d'eau et la température de l'eau peut affecter la circulation de la chaleur et de la matière organique dans la zone littorale, modifiant les conditions de production des GES. D'un autre côté, les macrophytes plus abondantes peuvent devenir un vecteur majeur pour le transfert de gaz entre les sédiments et l'atmosphère. Les flux de GES à travers les plantes n'ont pas encore été étudiés en profondeur dans les lacs tempérés. Néanmoins, les études démontrent que cela peut représenter jusqu'à 90% des émissions totales de CH_4 dans certains lacs peu profonds ayant une dense couverture de macrophytes (Chanton et al., 1992). Par conséquent, plus d'attention devrait être portée à ce mode de transport, aussi potentiellement affecté par le climat.

Des changements dans le niveau d'eau et le régime thermique de la colonne d'eau peuvent aussi affecter la structure du réseau trophique planctonique et générer des fleurs d'eau d'algues toxiques récurrentes (Paerl, 1988). La production de CH_4 et de N_2O dans les accumulations denses de cellules à la surface (et dans les biofilms) pourrait contribuer significativement au potentiel de réchauffement global des gaz émis par les lacs affectés par de tels changements. La formation de films à la surface du lac composés de surfactants ou de cellules peut aussi affecter la turbulence et contrôler la response des coefficients de transfert gazeux au vent et à la convection (Banerjee et MacIntyre, 2004). Des études expérimentales sur les taux spécifiques de production de GES en milieu oxique associée au phytoplancton devraient être réalisées en utilisant différentes espèces de phytoplancton. Ceci est particulièrement intéressant compte tenu des récentes découvertes de Grossart et al. (2011) qui ont montré une plus forte production de CH_4 associée aux cyanobactéries. L'importance relative de la production aérobique de CH_4 associée au phytoplancton (Tang et al., 2014) en relation avec la production anaérobique de CH_4 dans les sédiments pendant la décomposition de la matière organique fraîche devrait également être étudiée. J'ai réalisé des expériences préliminaires de décomposition de la biomasse fraiche de phytoplancton et de macrophytes (Annexe; Fig. 4.4.1). Les résultats indiquent des productions plus élevées de CO_2 et de CH_4 pendant la décomposition des macrophytes que celle du phytoplancton. Les résultats suggèrent aussi une biodisponibilité plus élevée de la matière organique phytoplanctonique étant donné une augmentation plus soudaine de la production de CH_4 dans les 4 premiers jours, quoique la production chute ensuite. Idéalement, de telles expériences bénéficieraient de l'application d'outils moléculaires permettant d'évaluer la présence de taxons spécifiques impliqués dans la production et la consommation de GES via les voies métaboliques moins connues, telles que la méthanogenèse aérobique et la méthanotrophie anaérobique (Eller et al., 2005).

Des changements dans la structure de la communauté de phytoplancton seront accompagnés de changements dans la biomasse et la taille du zooplancton, ce qui peut avoir un effet sur la production de CH_4 et N_2O. La seule étude existante à ce jour portant sur la production de GES dans le zooplancton a montré des taux faibles mais significatifs de production de CH_4 dans les copépodes marins broutant le phytoplancton (de Angelis et Lee, 1994). Dans plusieurs petits lacs peu profonds, le zooplancton est dominé par les cladocères, pourtant aucune étude examinant la production de CH_4 ou de N_2O par ces herbivores aquatiques clés n’a été réalisée.

Enfin, une autre question intéressante formulée au cours de mon étude est de connaitre si le forçage météorologique différentiel affecte l’importance du CH_4 comme source de carbone dans la chaîne trophique pélagique. Par l’intermédiaire des bactéries methanotrophique, le CH_4 agit comme source de carbone pour les organismes benthiques, le plancton hétérotrophe et les poissons consommant le plancton (Bastiviken et al., 2003). L’importance de cette source d’énergie peut toutefois dépendre de la prédominance des voies d’émission du CH_4 (diffusion ou ébullition), de l’activité et de l’abondance des MOB contrôlées par les concentrations en oxygène et en nutriments (Kaankala et al., 2006), de la distribution verticale et du taux de broutage du zooplancton, ainsi que de leur capacité à exploiter la nourriture bactérienne (Geller et Müller, 1981). Le transfert de C-CH_4 à travers la chaîne trophique pélagique peut être évalué en exploitant le fait que le fractionnement typique des isotopes stables du CH_4

biogénique ($\delta^{13}C$ typiquement autour de -60 à -80‰) est différent de la signature du phytoplancton (-25‰ à -35‰) et de celle du matériel terrestre. Les résultats de telles études pourraient être utilisés dans la paramétrisation d'un modèle de méthane biogénique lacustre qui fournirait le moyen d'évaluer les effets possibles des changements climatiques futurs sur le cycle du CH_4 dans les plans d'eau possédant différentes caractéristiques limnologiques.

1.7 Conclusions, contribution scientifique et innovation

Récemment, les eaux intérieures ont été reconnues pour leur importance dans le cycle global du carbone (Tranvik et al., 2009). Les lacs de petite taille sont les plans d'eau les plus abondants et biologiquement les plus actifs sur notre planète (Downing, 2009), et pourtant leur rôle est encore mal connu. Les résultats de cette thèse mettent l'emphase sur l'importance des lacs de petite taille et peu profonds comme source de GES pour l'atmosphère, et montrent que la synchronisation et l'ampleur des émissions de gaz dépendent de la méthode utilisée pour estimer les coefficients de transfert de gaz et du forçage météorologique (Article 1), de la stratification en oxygène et des concentrations en nutriments (Article 2), ainsi que de l'abondance et la structure du plancton (Article 3).

Très peu d'études jusqu'à maintenant ont investigué la variabilité spatiotemporelle des niveaux de saturation et du stockage des trois principaux GES pendant des conditions climatique contrastées. Seuls des effets récents sur le cycle du CO_2 sous des conditions estivales extrêmes ont été présentés par Vachon et del Giorgio (2014) pour des lacs tempérés, et par Pokrovsky et al. (2013) pour des mares arctiques. À ma connaissance, les résultats présentés dans ma thèse sont les premiers décrivant les effets d'un été extrêmement chaud et sec sur les cycles et les émissions de CO_2, CH_4 et N_2O par diffusion et ébullition, et faisant le lien avec la météorologie de surface et les caractéristiques de l'environnement. À ma connaissance, cette thèse constitue également la première étude sur les effets des fleurs d'eau d'algues et de la structure du réseau trophique planctonique sur la dynamique des GES dans les zones pélagiques et littorales d'un lac hypereutrophe. Cette thèse est organisée de manière à pouvoir d'abord évaluer les effets de la structure physique du lac et des méthodes d'estimation des coefficients de transfert des gaz sur les flux de gaz à l'interface eau-air pendant l'été sous des conditions climatiques contrastées. Dans un deuxième temps, la thèse étudie l'étendue de l'effet de la vague de chaleur estivale sur l'oxygène, les nutriments et le cycle des GES, incluant les effets sur la spéciation et les voies d'émission des gaz. Enfin, étant donné que les vagues de chaleur en été dans les lacs peu profonds sont très souvent associées à des fleurs d'eau de phytoplancton (incluant des cyanobactéries), j'ai étudié les effets des fleurs d'eau sur la structure du réseau trophique planctonique et sa relation avec les trois GES.

La méthode sélectionnée pour la quantification des flux diffusifs peut introduire des biais dans l'estimation des émissions de GES par les lacs de petite taille peu profonds. Dans cette thèse, j'ai évalué la précision de divers modèles d'échanges gazeux en comparaison avec les mesures directes de flux obtenues avec une chambre flottante branchée sur un détecteur de gaz. Mes résultats indiquent que le modèle de renouvellement de surface de MacIntyre et al. (2010) et celui basé sur le vent de Cole et Caraco (1998) fournissent à eux deux une enveloppe permettant d'anticiper les coefficients de transfert des gaz. La fenêtre d'étendue devrait être plus large à des faibles vitesses de vent. Le coefficient de traînée augmente quand l'atmosphère est instable et diminue quand il est stable, avec un effet plus marqué pour des vitesses de vent inférieur à ~2 m s^{-1} et un rétrécissement quand le vent augmente. Pour des vitesses de vent inférieures à 2 m s^{-1}, le multiplicateur de la vitesse du vent dans l'équation de k augmente dans le cas d'une atmosphère instable. Ainsi, la variabilité dans la stabilité de l'atmosphère (facteur non étudié dans cette thèse) peut aussi contribuer à la variabilité de k, avec les effets les plus prononcés aux faibles vitesses de vent (Annexe 4.1).

Les travaux récents de Cole et al. (2010) impliquent qu'une conception améliorée des chambres flottantes permet d'éviter certains biais causés par le déploiement des chambres sur la turbulence (Vachon et al., 2010), avec un rendement adéquat pour des conditions de vent faibles à modérées (Kramer et al., 2003; Alin et al., 2011). De la même façon, les travaux récents de Huotari et al. (2013) ont montré des valeurs similaires entre les coefficients d'échange gazeux obtenus à partir de chambres flottantes à ceux obtenus par les systèmes de covariance des turbulences en rivière, venant appuyer la justesse des estimations par ces chambres. Les résultats de ma thèse démontrent le même type de concordance quand on travaille à un seul endroit et qu'on applique différents modèles de mesure des flux, valorisant l'utilité des chambres flottantes qui permettent d'améliorer les modèles de transfert gazeux dans les lacs de différentes tailles et formes.

L'augmentation de la fréquence de périodes exceptionnellement chaudes et sèches accompagnées par des pluies torrentielles et des ouragans est une des conséquences des changements climatiques (Schär et Jendritzky 2004). Ce changement affecte aussi le fonctionnement des petits lacs peu profonds, avec des effets prononcés sur la magnitude et les voies d'émission des GES. La seconde partie de ma thèse révèle que 1) la réponse des petits lacs aux conditions climatiques pendant les vagues de chaleur peut être utilisée pour évaluer les effets des changements climatiques sur de tels systèmes, 2) avec le réchauffement climatique, la qualité de l'eau de ces lacs peut se détériorer suite à la mobilisation des nutriments accumulés dans les sédiments et des fleurs d'eau d'algues plus fréquentes, 3) les zones lacustres colonisées par les macrophytes peuvent être particulièrement susceptibles aux effets de la canicule et peuvent réduire leur efficacité de rétention des nutriments, et 4) malgré l'augmentation de la diffusion des GES durant la vague de chaleur, globalement le PRC du lac n'a pas augmenté en raison de la baisse des flux d'ébullition du CH_4. Ces résultats sont contraires à la perception générale.

Au dernier chapitre de ma thèse, je montre comment la biomasse et la composition du plancton affectent le niveau de saturation et l'accumulation des GES dans un lac hypereutrophe. La fréquence des efflorescences d'algues toxiques dans les lacs est un problème important pour la gestion de la qualité de l'eau. Cette thèse offre un aperçu du processus de rétroaction pouvant exister entre le climat, le plancton et la spéciation des GES dans un petit lac, et valorise le besoin d'inclure des mesures de GES dans les programmes de surveillance des lacs. L'étude de Moss et al. (2011) suggère que l'eutrophisation des lacs peu profonds en réponse au réchauffement climatique pourrait accélérer le réchauffement par une boucle de rétroaction positive, car les lacs plus chauds, plus stratifiés et plus chargés en nutriments pourraient émettre une plus grande proportion de GES plus puissants (CH_4 et N_2O). Aussi, l'étude d'Yvon-Durocher et al. (2011) montre que le réchauffement peut donner lieu à de plus fortes émissions de CH_4

par les lacs car une hausse des températures stimulent particulièrement l’activité des méthanogènes. Mes résultats supportent l'hypothèse selon laquelle le PRC des GES diffusifs peut augmenter en raison du réchauffement et de l'eutrophisation (lac Jacques et lac St-Augustin), mais ils montrent également que cela peut être accompagné par une baisse de l'ébullition résultant d'une réduction globale du PRC.

Finalement, les résultats présentés dans ma thèse supportent le fait que l’apparition et la persistance de fleurs d'eau d'algues, reliées à un été chaud et sec, affecte la structure du réseau trophique planctonique et mène à une plus grande accumulation de CH_4 et N_2O dans les eaux du lac, au détriment du CO_2. Ces effets sont importants pour comprendre l’impact des lacs sur le climat global et fournir des arguments pour une restauration plus efficace des lacs pollués. Avec le réchauffement du climat et l’eutrophisation, de plus en plus de lacs sur la planète sont susceptibles de subir des efflorescences d'algues toxiques. La composition des efflorescences et la réponse du réseau trophique contrôleront le potentiel de réchauffement climatique des gaz émis par les lacs. La mise à l'échelle et la modélisation de la contribution des petits lacs au budget global de carbone exigera plus de données sur la réponse des processus autochtone et allochtones au forçage météorologique.

Influence des processus externes et internes sur les émissions de gaz à effet de serre dans les lacs de petite taille

PARTIE 2. Articles

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2.1 Greenhouse gas emission and storage in a small shallow lake

Émission de gaz à effet de serre et leur stockage dans un petit lac peu profond

(Hydrobiologia, Springer, doi: 10.1007/s10750-015-2240-2)

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Les données limnologiques ont été collectées et analysées par M. Bartosiewicz sous la supervision d'Isabelle Laurion. Les données météorologiques ont été collectées et analysées par M. Bartosiewicz ou téléchargées du serveur NASA (Giovanni 3) sous la supervision de Sally MacIntyre. Le modèle de renouvellement de surface a été développé par Sally MacIntyre. Le manuscrit a été écrit par M. Bartosiewicz et révisé par Sally MacIntyre et Isabelle Laurion. La version finale du manuscrit a été soumise à Hydrobiologia le 1er février. Le manuscrit a été accepté le 2 mars et publié en ligne le 7 mars 2015.

Résumé

Les petits lacs sont plus susceptibles de montrer une forte variabilité temporelle des émissions de gaz à effet de serre en raison de leur stratification transitoire et leur temps de résidence court. Pour déterminer à quel point les émissions de CO_2 et de CH_4 et leur stockage dépendent de la météorologie de surface, nous avons étudié un lac peu profond pendant deux années montrant des conditions contrastantes en termes de précipitation et de stratification thermique. Les flux de gaz ont été estimés grâce à des modèles basés sur la vitesse du vent ou sur le renouvellement de surface, et ces estimations ont été comparées à des mesures directes obtenues à l'aide de chambres flottantes. Les résultats indiquent que le lac a accumulé du CO_2 en association avec le régime des précipitations, à la fois durant l'été pluvieux (2011) et l'été le plus sec (2012), suggérant un apport externe de carbone. En 2011, la stratification était moins intense et fréquemment perturbée. Le lac était alors une source en CO_2 et CH_4 pour l'atmosphère, et l'ébullition excédait les flux diffusifs en CH_4. En 2012, la stratification était plus persistante et le lac était un puits de CO_2 pendant les périodes sèches. Le CO_2 et le CH_4 se sont accumulés dans l'hypolimnion plus tard pendant l'été lorsque les précipitations ont augmenté. Les flux diffusifs de CH_4 en 2012 étaient similaires à ceux retrouvés au coeur de l'été 2011, et quatre fois plus élevés pendant le retournement automnal. Les flux d'ébullition étaient pour leur part plus faible pendant l'été le plus sec. Les estimations issues du modèle basé sur le renouvellement de surface se rapprochaient le plus des flux de gaz mesurés à l'aide des chambres flottantes, alors que le modèle basé sur le vent sous-estimait les flux de deux fois.

Abstract

Small lakes are likely to show considerable temporal variability in greenhouse gas emissions given their transient stratification and short residence time. To determine the extent that CO_2 and CH_4 emission and storage depends on surface meteorology, we studied a shallow lake during two years with contrasting rainfall and thermal stratification. Gas fluxes were estimated with wind based and surface renewal models and compared to direct measurements obtained with floating chambers. The assessment of GHG storage revealed that the lake gained CO_2 in association with rainfall in both the rainier (2011) and drier summer (2012). In 2011, stratification was less extensive and disrupted frequently. The lake was a source of CO_2 and CH_4, and ebullition exceeded diffusive fluxes of CH_4. In 2012, stratification was more persistent, the lake was a sink for CO_2 during dry periods, CO_2 and CH_4 accumulated in the hypolimnion later in the summer when rainfall increased, diffusive fluxes of CH_4 were similar to those in 2011 mid-summer and over four times higher during overturn. Ebullition was lower in the drier summer. Fluxes measured with chambers were closer to estimations from the surface renewal model and about two times values estimated with wind based models.

Key words: small lake, weather variability, thermal stratification, greenhouse gases, storage, diffusive flux, ebullition, gas transfer modelling.

2.1.1 Introduction

Small and shallow lakes and impoundments are abundant in the landscape (Downing et al., 2006; Downing, 2010) and emit substantial quantities of atmospheric greenhouse gases (GHG) (Casper et al., 2000; Repo et al., 2007). They are active reactors where carbon derived from the catchment, atmosphere and ground waters can be stored, utilized, chemically altered or released as CO_2 and CH_4 (Kling et al., 1992; Cole et al., 2007). The rate and direction of CO_2 transport from lakes depends on the ratio of primary production to ecosystem respiration which in turn are driven by inputs of inorganic nutrients and allochtonous and autochthonous organic material (Kortelainen et al., 2006). The rate of CH_4 transport to the atmosphere depends on the rates of methanogenesis (Zeikus and Winfrey, 1976) and methanotrophy (Rudd and Hamilton, 1975; Whiticar et al., 1986). Methane can diffuse from the lake surface or be released *via* gas bubbles formed in the sediments (Bastviken et al., 2011; Wik et al., 2013). The amount of gas released to the atmosphere in these two modes of transport depends on water depth and temperature (Hofmann et al., 2010), on the composition, temperature and chemical characteristics of the sediments (Martinez and Anderson, 2013; Wik et al., 2014), and on turbulent mixing as it causes fluxes within the water column and across the air water interface.

Estimations of GHG flux from inland waters are increasing in accuracy due to improvements in modelling of the gas transfer coefficient *k*, more extensive data on dissolved gas concentrations (Raymond et al., 2013), and more detailed description of seasonal dynamics in GHG storage (Fernandez et al., 2014). Further improvement requires understanding how weather and its effects on stratification and turbulence control the production, emission and storage of GHG in lakes of different morphometry. For example, GHG emissions from a dimictic lake were shown to increase during cold fronts when water from below the mixed layer was entrained (Aberg et al., 2010), and during fall as summer stratification breaks down (Vesala et al., 2006). The study by Ojala et al. (2011) illustrates between year variability in a dimictic lake due to intrusions from rainfall. Few studies have been undertaken in shallow polymictic lakes in which diel stratification and mixing would be expected to lead to similar GHG storage and fluxes in summer and fall.

In shallow polymictic lakes water temperature and oxygen concentrations may change rapidly with changing weather (Wilhelm and Adrian 2008). Hence the seasonal and inter-annual variability in GHG storage and fluxes, both diffusive and ebullitive, may be markedly different depending on weather conditions (Natchimuthu et al., 2014). These conditions affect GHG through their effect on sediment re-suspension (Bussmann 2005; Hofmann et al., 2010) and on inflows of ground and stream waters, which can be considerable given the short residence time of such small lakes. They also affect the water column thermal structure and the intensity of turbulent mixing controlling gas diffusion at the air-water interface (Jahne et al., 1987; MacIntyre 1993). Diffusive gas fluxes at the lake surface can be assessed using wind based models (e.g. Cole and Caraco, 1998), surface renewal models (MacIntyre et al., 2010; Tedford et al., 2014), or measured directly using floating chambers (Soja et al., 2014). Depending on the heterogeneity in surface water GHG concentrations and on factors controlling turbulent mixing, the choice of calculation method can introduce biases. The amount of energy incorporated into the water column due to wind stress or heating/cooling of the water column and sediments has also been shown to affect ebullitive fluxes (Wik et al., 2014).

In this study, we describe the inter-annual differences in stratification and surface meteorology between a cool rainy summer and a dry hot summer for a small 2 m-deep lake. We hypothesized that when exposed to contrasting weather conditions, the lake will alternatively be polymictic or stably stratified during the summer, with subsequent effect on GHG emission and storage. We compare GHG diffusive fluxes calculated with wind based and surface renewal models, and evaluate these against floating chamber measurements. We tested if stable stratification as opposed to conditions with more frequent mixing events results in a delayed release of diffused GHG until autumnal overturn, and globally higher CH_4 emissions. We also hypothesized that the amount and composition of the gas released via ebullition will depend on water column stratification and hypolimnetic temperature.

2.2.2 Methods

Study site

Lake Jacques is a small (0.18 km^2), shallow lake (maximum depth = 1.9 m, mean depth = 0.75 m) located 30 km north of Quebec City (QC, Canada), and supplied with water from two creeks in the eastern part of the lake, as well as from a ground water spring in its southern part. The lake can be classified as meso-eutrophic in terms of total phosphorus (mean summer value of 21 µg L^{-1}), total nitrogen (0.7 mg L^{-1}), and Chl-*a* concentrations (17 µg L^{-1}). Approximately fifty percent of the lake surface area is covered with the macrophyte *Brasenia schreberi* in summer. The upper 10 cm of the sediments has high organic carbon content (loss on ignition = 38 ± 5%, n = 10). The water retention time averages 14-20 days in summer (R. Tremblay, *pers. comm.*). The concentration of dissolved organic carbon ranged between 2.2 to 3.0 mg L^{-1} during the study and an algal bloom formed on the surface in 2012.

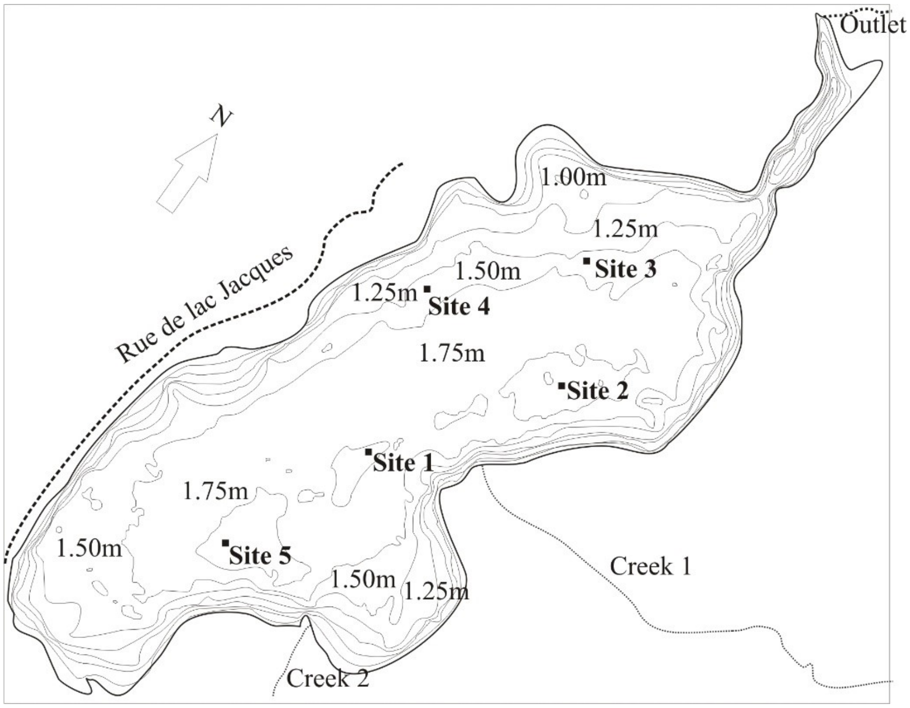

Figure 2.1.1 The morphometry and bathymetry of Lake Jacques. The thermistor chain was positioned at site 1 and ebullition funnels at sites 1-4 (2011) and 1-5 (2012).

Meteorological and physicochemical characteristics

In 2011, hourly data on air temperature, relative humidity, wind speed, wind direction, dew point, and precipitation were obtained from the meteorological station of Environment Canada located 7 km from the lake (anemometer threshold 1 m s^{-1}). In 2012, a meteorological station WeatherHawk 511 (anemometer threshold 0.6 m s^{-1}) mounted 25 m from the lake shore and 7 m above the water level, recorded air temperature, relative humidity, wind speed and wind direction every 30 minutes. Between 21 July and 6 August 2012, the on-site anemometer had to undergo maintenance and wind speed data were replaced by those from the Environment Canada station. To correct the wind speed data from the Environment Canada meteorological station, we performed a linear regression analysis when both sensors were operational in 2012. We used the resulting equation, $u_1=0.442 + 0.791u_2$ (R^2=0.65, p<0.001), to correct wind speeds (u) from 2011 and those from 21 July and 6 August in 2012. Downwelling hourly solar irradiance data (short wave and long wave) for both years were obtained from high spatial resolution (0.125-0.125°) surface meteorological forcing model from NASA (Giovanni Interactive Tool for Data Visualization and Analysis). Upwelling short wave and long wave were computed as in MacIntyre et al. (2014). Diffuse attenuation coefficient for downward visible light (LI-COR spherical quantum sensor LI-193) was calculated as the slope of the linear regression of $ln\ (E_z/E_0)$ versus depth, where E_Z is the irradiance at depth z and E_0 is the surface irradiance, and used in surface renewal model computing. The value was on average equal to 2.4 m^{-1}.

In 2011, surface temperature, conductivity, dissolved oxygen (DO) and pH were assessed fortnightly with a 600R multiparametric probe (Yellow Springs Instrument). The oxygen probe was calibrated at the beginning of each sampling day in water-saturated air. Surface water samples (100–500 ml) were filtered on GF/F glass fibre filters (0.7 µm nominal mesh size; Advantec MFS Inc.) for the determination of chlorophyll-a concentration (Chl-a) using a UV–Vis spectrophotometer at 750 and 665 nm (Wintermans and De Mots, 1965). In 2012, all measurements were performed weekly from May until the end of August, and then fortnightly until the ice cover was formed. The lake properties were always assessed between 10:00 AM and 2:00 PM.

Time series of water temperature were obtained with a thermistor chain installed from June to September 2011 and from May to October 2012. In 2011, seven temperature loggers (Onset Tidbit v2; accuracy 0.2°C, resolution 0.2°C, response time 5 min) were deployed at 0, 0.2, 0.4, 0.7, 1.0, 1.5 and 2.0 m, and acquired data every 15 minutes (sampling site 1.9 m deep at time of deployment, Fig. 1). In 2012, the loggers were deployed at the same site (depth of 1.7 m at time of deployment) at 0, 0.2, 0.35, 0.5, 0.75, 1.2 and 1.5 m, and acquired data every 8 minutes. Isotherms were calculated using linear interpolation. The surface energy budget and computation of dissipation rate of turbulent kinetic energy used in gas transfer coefficient calculations were computed following MacIntyre et al. (1995; 2002) and Tedford et al. (2014).

Indirect estimations of k with wind based and surface renewal models

The wind speed normalized at 10 m (u_{10}), according to the logarithmic wind profile relationship including atmosphere stability effects (Smith, 1988), was used to calculate the gas transfer coefficients (k_{600}) standardized to a Schmidt number (Sc) of 600 following Wanninkhof (1992). Using the equations of Cole and Caraco (1998; hereafter *CC),* we calculated:

$$k_{600} = 2.07 + 0.215 u_{10}^{1.7} \qquad (1)$$

and the equation of Crusius and Wanninkhof (2003; hereafter *CW*):

$$k_{600} = 0.168 + 0.228 u_{10}^{2.2} \qquad (2)$$

The equation in Cole and Caraco (1998) applies for wind speeds up to 9 m s^{-1} whereas that of Crucius and Wanninkhof (2003) to winds up to 6 m s^{-1}.

We also computed the gas transfer coefficient with the surface renewal model (MacIntyre et al. 1995; Zappa et al. 2007, hereafter *SC*):

$$k_{600} = a_1 (\varepsilon/v)^{1/4} Sc^{-1/2} \qquad (3)$$

where a_1 is an experimental constant (assumed as 0.5 in this study), ε *is the* kinetic energy dissipation rate, and v the kinematic viscosity. The kinetic energy dissipation rate, describing turbulence in the surface mixing layer where mixing is directly energized by wind shear and convection (Imberger 1998), was calculated during heating as $\varepsilon = 0.6\ u_{*w}^3/kz$ and during cooling as $\varepsilon = 0.56\ u_{*w}^3/kz + 0.77\beta$, where u_{*w} is the water friction velocity computed from wind shear stress, k is the von Karman constant, z is the depth equal to 15 cm, and β is the surface buoyancy flux (Tedford et al., 2014). For details on computing u_{*w} and β, see MacIntyre et al. (2002, 2014). The first equation implies wind as the dominant factor responsible for turbulence at the lake surface, while the second one includes cooling as an additional factor that generates turbulence.

Dissolved GHG measurements and flux estimations

Aqueous concentrations of CO_2 and CH_4 were determined fortnightly from June to mid-September in 2011, and weekly from May to the end of August in 2012, then fortnightly until the ice cover was formed. Gas sampling was performed by equilibrating 2 liters of lake water with 20 mL of ambient air, shaking for 3 min, and then injecting the headspace gases into a 5.9 ml Exetainer (Labco Scientific) previously flushed with helium and vacuumed (Hesslein et al., 1990). The procedure was always repeated 3 times and yielded CV on average less than 10%. Gas samples were taken within 5 min after collecting the lake water, and were kept in the dark at 4°C until analyzed by gas chromatography (Varian 3800 with a COMBI PAL head space injection system). In addition, vertical profiles of CO_2 and CH_4 (resolution 0.25 m) were taken monthly or fortnightly from June to September in 2011 and from May to October in 2012 following the procedure described above. To assess gas storage, the total mass of CO_2 and CH_4 above (or below) saturation was calculated by multiplying the average concentration over a depth interval by the water volume within that depth interval, and summing over the depth of the lake (Rudd and Hamilton, 1978).

Diffusive CO_2 and CH_4 flux ($Flux_d$) was calculated using the gas transfer coefficients for a given gas k (cm h^{-1}) estimated with either *CC, CW* or *SC* models as:

$$Flux_d = k\,(C_{sur} - C_{eq}) \tag{4}$$

where C_{sur} is the gas concentration in surface water (mmol L^{-1}) and C_{eq} is the gas concentration in the water at equilibrium with the atmosphere. Global values of atmospheric partial pressures (IPCC 2007) were used to determine C_{eq}. The gas transfer coefficient was calculated as:

$$k = k_{600}\,(Sc/600)^c \tag{5}$$

where c equals -0.5 for rough surfaces (Csanady, 1990).

Synchronized with the determination of aqueous CO_2 and CH_4 concentrations, a floating chamber (circular, 23.4 L, hereafter *FC*), made of 10 mm thick PVC plastic with floaters distributed evenly on the sidewall, extending 4 cm into the water, was coupled with an infra-red gas analyser (EGM-4, PP-Systems), and deployed 2 m from the boat on the surface of the lake to directly assess CO_2 flux ($Flux_c$) according to:

$$Flux_c = S\, M_w\, V_{ch}\, (V_m\, A)^{-1} \qquad (6)$$

where S is the slope of the linear regression of gas concentration in the chamber versus time (measurements taken for a maximum of 20 min depending on flux rate), M_W the gas molecular weight, V_{ch} the volume of the chamber, V_m the gas molar volume at ambient temperature, and A the area of the chamber. Atmospheric and surface water dissolved CO_2 together with direct measurements of CO_2 flux were used to estimate the floating chamber gas transfer coefficients using the equation:

$$k = Flux_c / (C_{sur} - C_{eq}) \qquad (7)$$

k was standardized to k_{600} using equation 5. Gas transfer coefficients obtained from the *FC* were used to calculate the chamber CH_4 flux following equation 4.

Ebullition measurements

The rate of gas ebullition was measured with four (2011) or five (2012) submersible inverted funnels installed monthly from June to September 2011 (n=14 measurements, only 2 funnels worked in August), and 2012 (n=20 measurements). The funnels consisted of a soft PVC cone-shaped body mounted on a metal frame, with a plastic syringe and luer-lock valve installed on top, and maintained with floaters at approximately 20 cm below the water surface. At each sampling date, the funnels were submerged in the lake at one of the 4 or 5 pelagic sites (Fig. 1.) and moored with three weights deployed at least 2 m away from the funnel to avoid the collection of bubbles escaping at installation and to keep the funnel upright in the same position for 4 to 24 h depending on the flux rate. The collected bubbles were sampled in triplicate vials, and the samples diluted 100 times with helium before they were analyzed by gas chromatography as described above.

The ebullitive flux was calculated as:

$$Flux_e = C\ V_g\ V_m^{-1}\ A^{-1} \quad (8)$$

where C is the concentration of a given gas in the syringe, V_g is the total volume of the gas in the syringe, V_m is the gas molar volume at ambient temperature obtained from meteorological station, and A is the area of the funnel.

Finally, the total global warming potential over a 20 year period (GWP) was calculated as the sum of diffusive and ebullitive GHG fluxes with CH_4 having 34 times higher GWP than CO_2 (nominal 1; Myhre et al., 2013).

2.1.3 Results

Meteorological conditions and mixing of the lake

Air temperatures varied on seasonal and diel time scales. Maxima in summer were near 30°C; shifts between warm and cold fronts caused 5°C to 10°C changes in air temperatures, similar to variations over the course of a day (Fig. 2.1.2). Three to four day long cold fronts prevailed in 2011 starting from the end of June. In early July 2011 they were associated with rainfall and wind speeds reaching 5 m s^{-1}. Warm fronts later in July caused the lake surface to warm to 26°C, brought heavy rains and winds up to 8 m s^{-1} during the daytime. Winds often declined at night below the anemometer's threshold (1 m s^{-1}). Cold fronts with air temperature falling to 15°C and rainfall dominated the weather of early August 2011, but the winds at this period did not exceed 5 m s^{-1}. Early in September cold fronts were associated with heavy rains, but the highest winds, 6 m s^{-1}, were recorded later in the month during a warm front. The rate of dissipation of turbulent kinetic energy computed at 15 cm depth following Tedford et al. (2014) indicated that ε ranged between 10^{-8} and 10^{-6} m^2 s^{-3}, that is, the upper water column was moderately turbulent (MacIntyre et al., 2009). In 2012 warm fronts prevailed, starting from May when air temperatures reached 28°C and afternoon winds remained above 6 m s^{-1}. As in 2011, winds were low at night. In early June cooler air masses were associated with rain and daytime winds above 6 m s^{-1}. At this time the lake was cooling also during the day and buoyancy contributed to the turbulence at the surface. The weather in July and August 2012 was dominated by warm fronts with little rain particularly in July. On average, 2011 was cooler and windier than 2012 and had almost two times higher rainfall (Table 1).

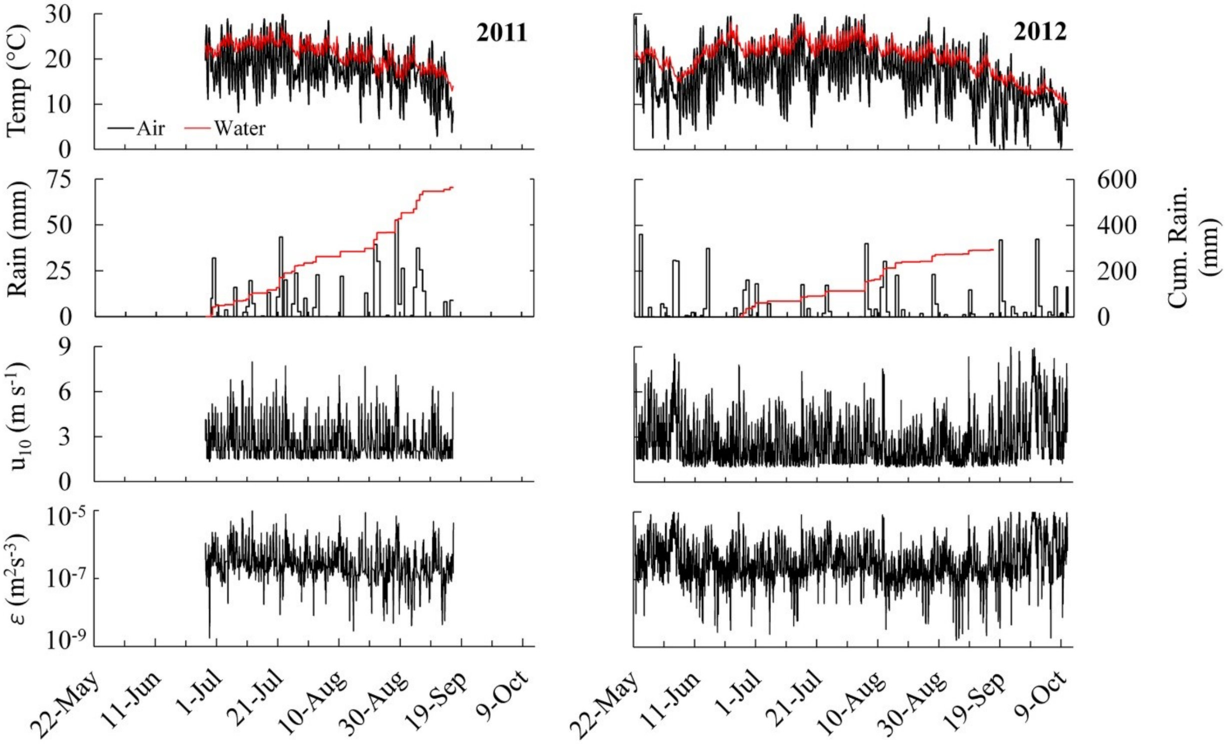

Figure 2.1.2 Meteorological conditions including air and surface water temperatures, daily and cumulative rainfall, wind speed at 10 m (u_{10} when winds were above 1 m s^{-1}), as well as kinetic energy dissipation rate (ε) following Tedford et al. (2014).

Thermal stratification, pronounced in both years despite the lake's shallow depth, was greater in 2012 than 2011 (Fig. 2.1.3). For example, the temperature difference from top to bottom was 8°C in mid-July 2011 and 13°C in mid-July 2012. The strong near surface heating is a result of the relatively high diffuse attenuation coefficient in this lake, 2.4 m^{-1}. In both summers, shallow diurnal thermoclines formed in the upper 20 cm to 60 cm. In 2011, the afternoon winds caused episodes of apparent mixed layer deepening. These patterns result from winds tilting the thermocline; on relaxation, the warm water was found at shallower depths and cooler water upwelled.

Meteorology	**2011**	**2012**
Air T (°C)	17.5	18.2
Cum. rain (mm)	563	292
Daily rain (mm d^{-1})	8.4	4.3
Wind (m s^{-1})	2.9	2.4
SW (W m^{2})	213	222
LW (W m^{2})	349	365
RH (%)	75.6	73.5
$\varepsilon \times 10^{8}$ (m^{2} s^{-3})	62.4	49.6

Table 2.1.1 Meteorological conditions, including air temperature (air T), cumulative precipitation (cum. rain), daily precipitation (daily rain), wind speed (wind), downwelling short wave (SW) and long wave (LW) irradiation, relative humidity (RH), and the dissipation rate of turbulent kinetic energy (ε) as a mixing index at Lake Jacques in summer 2011 and 2012. The comparison of wind speeds and dissipation rates in the actively mixing layer was done for winds exceeding 1 m s^{-1}.

Similar events occurred in 2012, but the penetration of warm water was restricted to shallower depths. Intervals with warm temperatures in the upper meter were interspersed with ones with cooler due to the passage of cold fronts during which near surface temperatures dropped by ~10°C. In consequence, the temperature differences within the water column were reduced to 2°C in 2011 and 7°C in 2012 (Fig. 2.1.3).

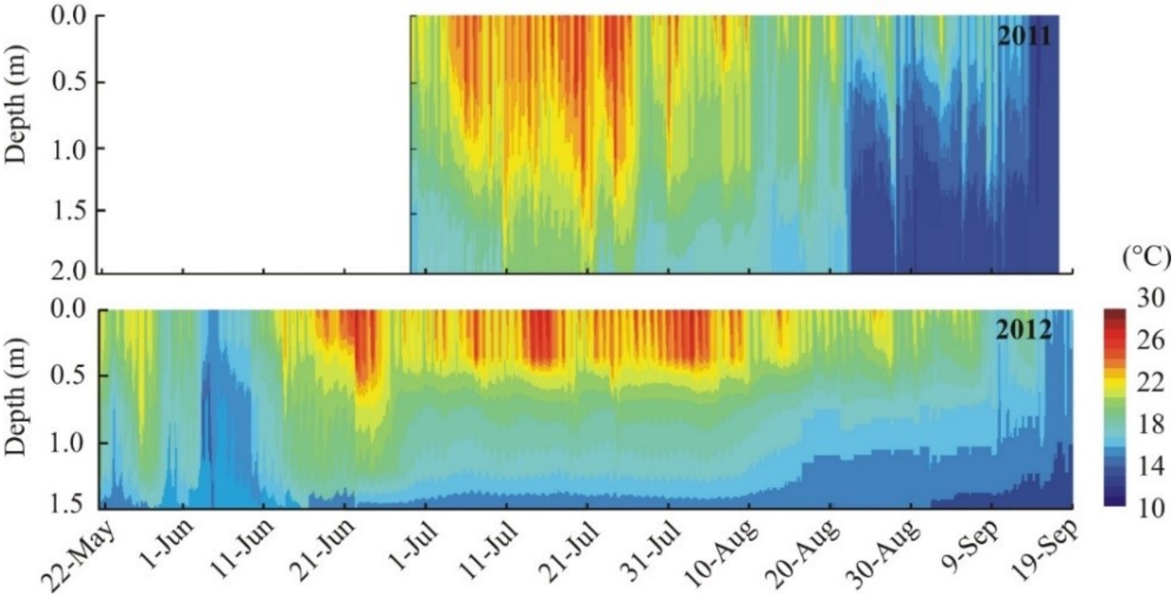

Figure 2.1.3 Thermal structure of the lake during rainy, cooler summer 2011 and dry, hotter summer 2012.

During cold fronts, temperatures often decreased in the lower water column. The large decreases in temperature in the lower water column in 2011 co-occurred with rain events and may be the signature of cold inflows of stream water. The transport could also be associated with differential cooling between pelagic and littoral zones. For example, at the pelagic site, temperatures cooled from 20-22 July in the thermocline between 40 and 80 cm but not at one meter depth. As inflows from rain were low at that time, the mid-water column cooling, with temperatures similar to the inshore site, is indicative of offshore flows from differential cooling. Even in 2012, cold fronts tended to be associated with rainfall, so cool water intrusions may have been due to the combination of increased stream/ground water inflows and/or differential cooling.

Stratification, quantified as buoyancy frequency, $N^2 = g\ \rho^{-1}\ d\rho/dz$, where g is the acceleration constant due to gravity, ρ is the water density and $d\rho/dz$ is the vertical density gradient, showed layering with features typical of seasonally stratified lakes. The upper water column stratified in the day and mixed at night, and comprised the epilimnion. For the first half of summer 2011, the metalimnion, with more persistent stratification, began at 0.8 or 1 m and extended nearly to the bottom with N having maximal values of 60 cycles per hour (60 cph = 0.1 rad s^{-1}). To provide context, oceanographers consider water to be strongly stratified when $N > 20$ cph. For most of summer 2012, the metalimnion began between 0.3 and 0.6 m and extended to 0.6 to 0.8 m with maximal values of N near 100 cph. Below the metalimnion, buoyancy frequencies were higher than in 2011. Importantly, in 2011 the stratification across the metalimnion intermittently weakened throughout its depth, with values dropping to 30 cph or 40 cph. In contrast, this weakening did not occur in 2012. The weakening is indicative of periods with increased mixing, which reduce the stratification and enable transport of dissolved gases across the metalimnion. In both years, increased rainfall after mid-August weakened the stratification in the lower water column, with N dropping below 30 cph in 2011 and below 50 cph in 2012. During this latter period in 2011 there were intervals when buoyancy frequencies became nearly uniform throughout the water column. In 2012, buoyancy frequencies only became near uniform with values below 20 cph in concert with the large rainfall event on 16 September (Fig. 2.1.2). Thus, rainfall contributed to weakening of vertical stratification. We infer that the persistent rainfall in summer 2011 enabled frequent exchanges between the upper and lower water column. The persistently high metalimnetic values of N in 2012 (50 cph), until the rainfall events later in the summer, imply reduced vertical fluxes between the metalimnion and epilimnion.

Variability in GHG vertical distribution and departure from saturation

The differences in stratification and rainfall between years led to differences in vertical distribution of dissolved gases. The oxycline tended to be located between 0.5 m and 1.5 m in both years, with anoxia prevalent below 1.5 m in 2012 (Fig. 2.1.4). Concentrations of dissolved gases differed between sampling periods in both years, with large increases prevalent either near the bottom or in mid-water intrusions (Fig. 2.1.4). In 2011, increases of CO_2 were associated with the higher frequency of rainstorms. For example, the large increase of CO_2 in the hypolimnetic waters on 6 August followed heavy rains on 5 August. Dissolved CH_4 did not accumulate in the lower water column during summer 2011. The higher temperature and oxygen levels in the lower water column in 2011 relative to 2012 provide evidence for larger vertical exchanges in 2011. Were CH_4 produced in 2011, it would have been oxidized quickly. In 2012 high concentrations of CO_2 and CH_4 did not occur in bottom waters during July but increased in August, and de-gassing occurred when stratification was eroded in September. Increases in gas concentrations in August 2012 co-occurred with increased rainfall, decreases in near-bottom temperatures, and cooling indicated by heat budgets (averaged over 2 days). Concentrations of CH_4 also increased inshore (data not shown). Thus, rainfall events appear to have caused loading of either organic matter or CO_2 in both years, with stronger stratification, anoxia and weaker mixing in 2012 enabling the persistence of CH_4 in the hypolimnion.

Surface CO_2 concentrations were significantly higher in 2011 than in 2012 (mean of 16.3 μM and 8.6 μM respectively, $p<0.05$). On average, surface CH_4 concentrations were higher in 2012 than in 2011 (0.89 μM and 2.1 μM respectively, $p<0.05$), but this latter difference was mainly due to the large increase of dissolved CH_4 in the water column during fall overturn. The variability in GHG saturation levels was higher in 2012 (CV=145% and 210% for CO_2 and CH_4 respectively) than in 2011 (CV=58% and 45%). The comparison of gas transfer coefficients was done for data with wind speeds above 1 m s^{-1} (anemometer thresholds) and below 6 m s^{-1} (upper wind speed limit for the *CW* model), thus allowing similar conditions for all models used in calculations. Overall, *k* was higher when estimated with the surface renewal (*SC*) model than when estimated with wind based models and *k* was higher with *CC* than *CW* (Table 2).

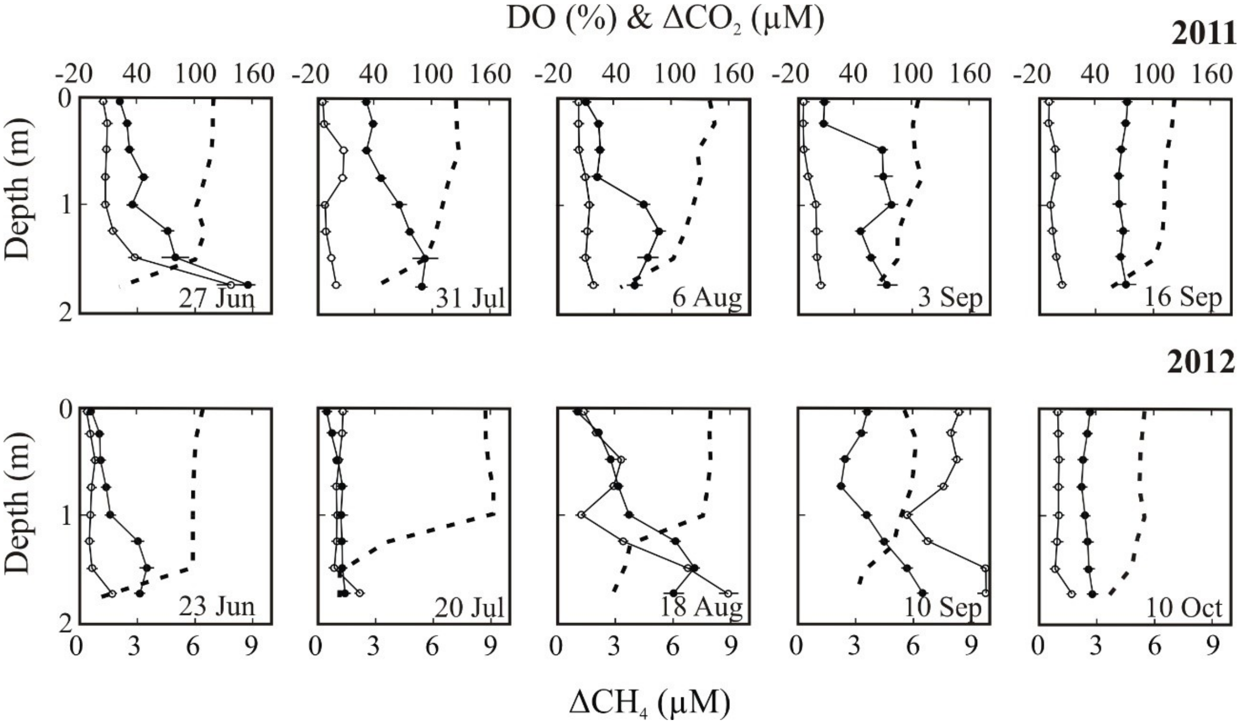

Figure 2.1.4 Profiles of dissolved oxygen saturation (dotted line, upper axis) as well as the departure from saturation of CO_2 (closed circles, upper axis) and CH_4 (open circles, lower axis) in Lake Jacques during 2011 and 2012.

Comparison of gas transfer coefficients calculated with wind based and surface renewal models

The contribution from buoyancy flux was negligible. The application of different models to estimate k_{600} revealed significant differences between years in average values (Mann-Whitney test, $p<0.05$), significant differences between models ($0.001< p < 0.05$) in the same year, and significant differences in the magnitude of k_{600} variability (coefficient of variation, CV) calculated with wind based (*CW*) and surface renewal models ($p<0.05$). The higher values using the surface renewal model result because k_{600} increases more rapidly with wind speed than in the Cole and Caraco (1998) formulation, similar to results shown in MacIntyre et al. (2010) under heating. The larger variance with *CW* results because it is a power law formulation with steep curvature relative to *CC*, and the lower average values result because the curve goes through zero at low winds and provides lower estimates of k at low winds than *CC* (Banerjee and MacIntyre 2004). Because of this bias, we do not include *CW* in the comparisons with the floating chamber to follow.

	2011		2012	
Model	k_{600} cm h^{-1}	CV %	k_{600} cm h^{-1}	CV %
CW	2.94	80	2.22	100
CC	3.47	27	3.15	28
SC	5.22	30	5.28	29

Table 2.1.2 The gas transfer coefficient k_{600} and its variability (CV) for two wind based models (*CW* and *CC*) and a surface renewal model (*SC*) at Lake Jacques during the rainy and dry summer.

Comparison of GHG fluxes estimated with floating chambers and models

The relative magnitude of fluxes measured with the floating chambers was similar to modeled values (Table 3).

$Flux_d$ (CO_2 / CH_4)	*FC*	*CC*	*SC*	$R_{FC:CC}$	$R_{FC:SC}$
	mmol m^{-2} d^{-1}				
2011	29.1 / 0.6	18.7 / 0.65	29.4 / 1.02	0.84	0.96
2012	9.0 / 1.35	4.5 / 1.52	9.2 / 2.80	0.90	0.98

Table 2.1.3 Average GHG diffusive fluxes (CO_2 / CH_4) in 2011 (June-September) and 2012 (May-October) measured with floating chambers (FC) or calculated using a wind based model (*CC*) and a surface renewal model (*SC*), as well as the correlation coefficients (R) between modelled and measured CO_2 fluxes considering *FC* as the reference ($p<0.01$).

The winds were above the anemometer threshold when chamber measurements were conducted. With respect to CO_2, chamber fluxes were more similar to fluxes estimated using the surface renewal model (*SC*) than with *CC* wind based model (Fig. 2.1.5 A, B). The correspondence was still good between measured fluxes and *SC* estimates during autumnal overturn. With respect to CH_4, results of both models differed at most by 30% from these calculated with the floating chamber in 2011 (Fig. 2.1.5 C). During 2012, *FC* fluxes were again similar to both models early in the season, they were lower than both models in mid-summer, and similar to *CC* modeled fluxes during autumnal overturn (Fig. 2.1.5 D).

Temporal variability in GHG diffusive fluxes and storage

On average, higher CO_2 diffusive fluxes occurred in 2011 than in 2012 (June to September; $p<0.01$, ANOVA with Tukey test; Table 2.1.4, Fig. 2.1.5). In summer 2011, the lake was continuously a net source of CO_2 to the atmosphere. Similarly it was a source in early and late summer 2012, but a sink in mid-summer.

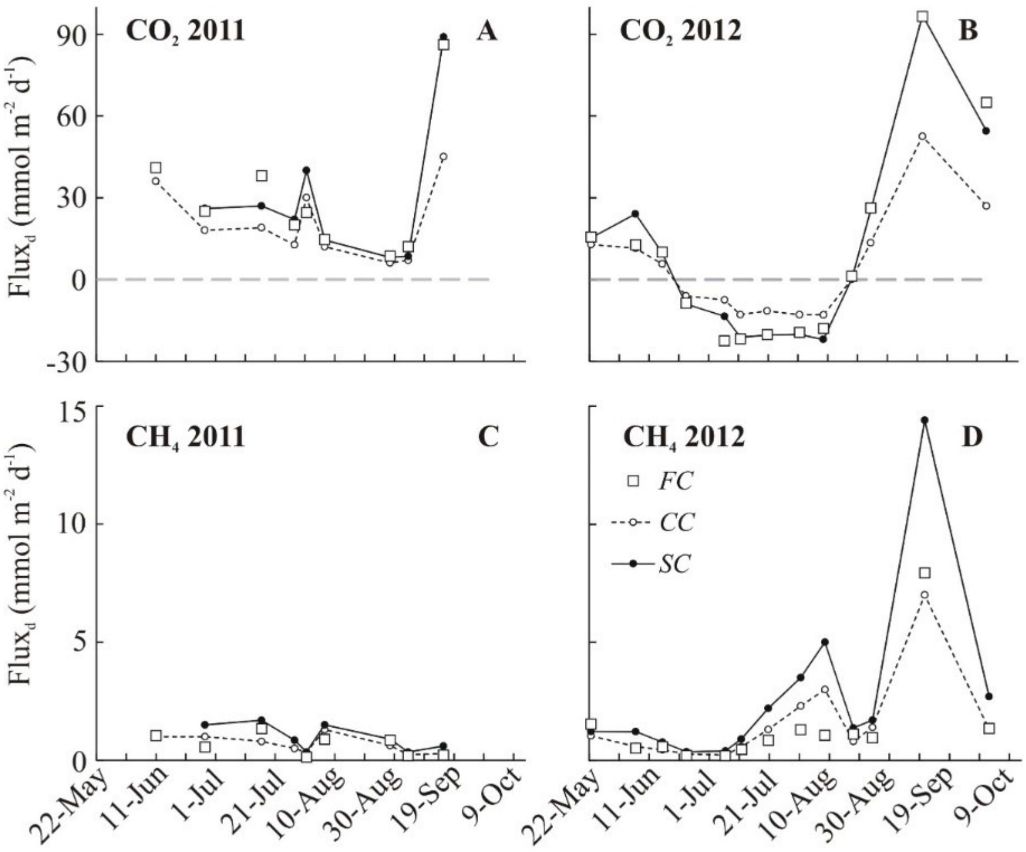

Figure 2.1.5 The CO_2 and CH_4 diffusive fluxes ($Flux_d$) in 2011 and 2012 estimated with wind based (*CC*, dashed line) and surface renewal model (*SC,* solid line) or measured directly (CO_2) and estimated with floating chambers (*FC*, open squares).

In contrast, diffusive CH_4 fluxes were higher on average in 2012 than in 2011 ($p<0.01$). The highest CO_2 fluxes occurred in September when stratification was eroded, with estimates averaging 89 and 97 mmol m^{-2} d^{-1} for *SC* model and 86.5 and 96.5mmol m^{-2} d^{-1} for *FC*, in 2011 and 2012, respectively. The CH_4 emissions also increased at the autumnal overturn, but only in 2012, with diffusive fluxes reaching 14 mmol m^{-2} d^{-1} for *SC* model and 7.95 mmol m^{-2} d^{-1} for *FC* measurements (Fig. 2.1.5).

The assessment of GHG storage revealed that, despite an efflux at the surface, the lake was gaining CO_2 early in 2011 (June-July) in association with heavy rainfalls (Fig. 2 and 6). By the end of August rainfall increased again, and so did the CO_2 concentrations in the lake. This rise continued until mid-September when CO_2 exceeded 16×10^3 moles. The CH_4 tended to follow a pattern opposite to CO_2 as it decreased early in the summer during the rainy period, increased when rainfall was relatively low, and decreased again later in the season. In 2012, the lake lost CO_2 between June and July when rainfall was low and Chl *a* increased from 13 µg L^{-1} to 37 µg L^{-1}. The lake gained CO_2 beginning in August with large increases in mid-August and in September when rainfall increased and temperatures in the lower water column decreased (Fig. 2.1.2 and 2.1.3). The stable stratification resulted in the development of anoxia in the hypolimnion in the summer, and accumulation of CO_2 and CH_4 reaching 12.6×10^3 and 19.7×10^2 moles respectively, despite the efflux of both gases at the surface (Fig. 2.1.5). While a considerable amount of GHG was lost during the overturn period in 2012, the mass of each gas above saturation was still significant in October.

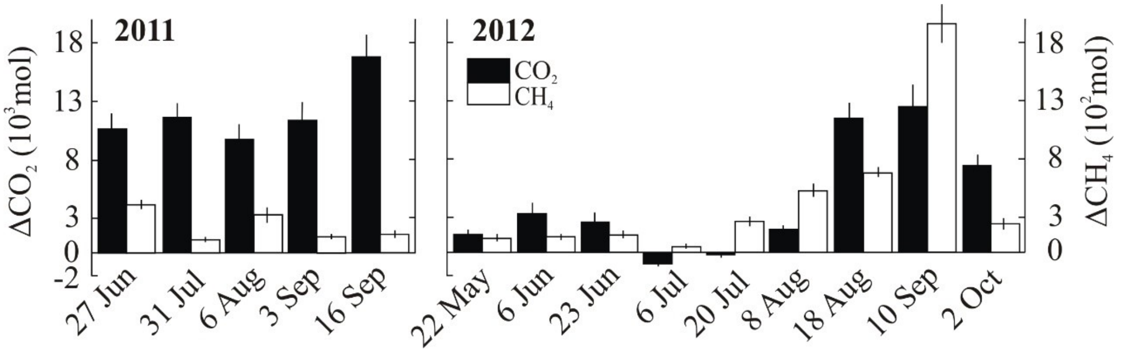

Figure 2.1.6 The storage (mass relatively to saturation ±SD) of CO_2 (closed bars, scale on left axis) and CH_4 (open bars, scale on right axis) in Lake Jacques in 2011 and 2012.

Greenhouse gas ebullition

Methane ebullition was higher during the rainier summer 2011 (4.1 mmol m^{-2} d^{-1}; n=14) than 2012 (0.6 mmol m^{-2} d^{-1}, n=20; $p<0.01$, Table 4). It was also higher during rainier months of 2012. Overall in 2011, ebullition contributed 66% of the total lake CH_4 flux in June, and 83% in July and September (comparing with *SC* diffusive flux). In 2012, the contribution of ebullition decreased from 58% in June, to 24% in July, 16% in August, and 5% in September. The collected gas bubbles contained, on average, more CH_4 in 2012 than in 2011 ($p<0.01$; n=34, partial pressures are provided in Table 4). The bubbles contained small amounts of CO_2, thus ebullition contributed only from 0.02 to 4% of the total CO_2 flux respectively for both years. Interestingly in 2012, when diffusive CO_2 flux was negative, CO_2 ebullition flux ranged from 0.28 mmol m^{-2} d^{-1} in July to 0.015 mmol m^{-2} d^{-1} in August. The total GWP of the lake was 32% higher in 2011 (201 mmol CO_2 equivalent m^{-2} d^{-1}) than in 2012 (152 mmol CO_2 equivalent $m^{-2}d^{-1}$; Table 2.1.4).

	CO_2 Flux$_d$	CH_4 Flux$_d$	CH_4 Flux$_e$	P_eCH_4	P_eCO_2	GWP
		mmol m^{-2} d^{-1}		%	%	mmol m^{-2} d^{-1}
2011						
June	26.5	1.4	3.4	21.4	4.1	192
July	29.7	1.0	5.3	30.5	0.8	242
August	11.3	1.2	5.1	31.2	1.8	225
September	48.8	0.5	2.4	42.3	0.6	146
2012						
June	8.3	0.7	1.1	14.4	5.0	72
July	-18.7	1.7	0.5	39.1	2.3	57
August	1.2	2.6	0.3	42.5	0.7	102
September	75.5	8.5	0.5	51.3	0.02	376

Table 2.1.4 Monthly average diffusive fluxes (Flux$_d$) using surface renewal model (*SC*), ebullition fluxes (Flux$_e$), partial pressure of CH_4 and CO_2 in the collected bubbles (P_eCH_4 and P_eCO_2 in %), and the global warming potential (GWP) of GHG emitted by Lake Jacques in summer 2011 and 2012 (given as CO_2-equivalent).

2.1.4 Discussion

Despite its small size, Lake Jacques had a persistent layered structure over the summer, similar to deeper lakes, and stratification was strong. For example, buoyancy frequencies in the metalimnion were higher and more persistently high than those reported during diel stratification in tropical lakes with high insolation (MacIntyre et al., 2002), higher than observed in the pycnocline in a much larger meromictic lake (MacIntyre et al., 1999), and similar to values computed using data from a turbid subarctic thaw pond (Laurion et al., 2010). The stratification was weakened during the passage of cold fronts, with such events occurring more frequently and causing a greater weakening of the stratification in 2011 than in 2012. The variability in stratification moderated exchanges between the upper and lower water column, the extent of oxygenation of the lower water column, and the speciation of GHG.

The between-year differences in weather led to variability in storage and emissions of GHG. Frequent cold fronts associated with rain in 2011 resulted in accumulation of CO_2, but not CH_4, in the lower water column of the lake and higher effluxes of CO_2 at the surface than in the warmer dryer year. The strong thermal stratification in 2012 resulted in persistent anoxia starting early in the summer. However, increases of both CO_2 and CH_4 in the lake occurred later in summer 2012 in association with rainfall. As a result of the stratification, emissions were delayed until a large rainfall event weakened the stratification in mid-September. The approach taken here, which included time series data and gas storage assessment, similar to that in Aberg et al. (2010), enabled us to illustrate that increased CO_2 within the lake was associated with rainfall and how the between-year differences in meteorology moderated stratification, which in turn moderated concentrations and proportions of CO_2 and CH_4, and their evasion to the atmosphere.

Our comparison of CO_2 fluxes measured with chambers versus surface renewal and wind based models was good, with comparisons within 30% between the chamber and surface renewal approach (Fig. 2.1.5). The comparisons with CH_4 were also good, with the largest discrepancy reaching a factor of two. These comparisons differ from those in Schubert et al. (2012) who report greater than four-fold differences between floating chamber and modeled fluxes of CH_4. In our experiments, the flux measurements and water samples for the model calculations were obtained at the same location. In contrast, Schubert et al. (2012) studied a larger lake and deployed multiple chambers to obtain appropriate coverage. As indicated in the analysis in Heiskanen et al. (2014), the combination of internal wave motions and convective cooling can induce spatial variability in greenhouse gas concentrations. This variability should be taken into account for robust comparisons of measured and modeled results and for model development. The good agreement we demonstrate by working at one location indicates floating chambers and model approaches can obtain similar results and points to the utility of the approach for developing accurate models of the gas transfer coefficient.

Accuracy in predicting GHG emissions also requires understanding controls on temporal variability. Temporal variability in GHG fluxes depends on the size of water bodies (small > large, Roulet et al., 1997) and on sampling frequency (Weyhenmeyer, 1999). Besides our results showing that emissions varies between years in association with difference in ambient meteorology, they also show they depend on the method used for estimating *k*. Models, such as the power law formulation of Crucius and Wanninkoff (2003), which force *k* to low values at low wind speeds, generate more variable GHG fluxes (max. CV= 100%). The surface renewal model approach gives less variable estimates. As it also includes the various processes mediating turbulence (Zappa et al., 2007; MacIntyre et al., 2010), it offers an approach for inclusion of a range of hydrodynamic controls on the gas transfer coefficient. In support of that model, we obtained better congruence with CO_2 fluxes from floating chamber and the surface renewal model than with the two wind based models used in our comparison. Since the differences in gas transfer coefficient parameterizations can cause biases in predicting the contribution of lakes to the global carbon cycle (Rypdal and Winiwarter, 2001), model results should be evaluated against direct measurements particularly in changeable weather conditions.

During the rainy summer 2011, Lake Jacques was continuously a source of CO_2 to the atmosphere indicating the prevalence of respiration and net heterotrophy. The increased CO_2 concentrations lower in the water column tended to co-occur with rainfall, thus it may have resulted from direct inputs of CO_2 or labile organic carbon from the drainage basin which fueled bacterial respiration in the lake. The higher winds in that year caused more frequent water column mixing, enabling evasion and possibly more frequent re-suspension of organic matter from the sediments, which also may have stimulated bacterial respiration. In 2012, surface waters were under-saturated in CO_2 from the end of June until early August, concomitant with supersaturation in dissolved oxygen. This depression indicates a drawdown of CO_2 by photosynthesis combined with reduced vertical fluxes due to stable stratification separating the lower and upper water column. In 2012, if CH_4 flux was not taken into account, the lake would have been a net sink for atmospheric carbon. However, when CH_4 was included in the carbon budget, the lake became a source of carbon to the atmosphere except in July (GWP in Table 2.4).

Weather conditions also affected the ratios of CO_2 to CH_4 diffusing from the lake and the amount of CH_4 emitted via ebullition. High amounts of oxygen introduced to the water column during stormy weather led to lower concentrations of dissolved CH_4, likely resulting from enhanced methanotrophy and supressed methanogenesis (Huttunen et al., 2006; Juutinen et al., 2009). When oxygen concentration was low, CH_4 accumulated in the lower water column. We did not determine whether CH_4 was derived from microbial processing of the CO_2 associated with rainfall events or directly from the sediments as in other studies (Rudd and Hamilton, 1978; Huttunen et al., 2003). Nevertheless, since the CH_4 only began to accumulate after rainfall, even though the lower water column was anoxic for most of 2012, it suggest that methanogenesis was stimulated by the microbial processing of the organic matter (Huttunen et al., 2003) or CO_2 (Wand et al., 2006) introduced with runoff. Moreover, stormy weather resulted in significantly higher CH_4 release from the lake *via* ebullition, possibly linked to warmer sediment temperatures (Wik et al., 2014) due to weaker stratification enabling a larger downward flux of heat. The higher frequency of frontal activity would have led to rapid changes in atmospheric pressure that also induces ebullition (Fechner-Levy and Hemond, 1996; Tokida et al., 2007). In Lake Jacques, ebullition was a more important CH_4 emission pathway than diffusion, as demonstrated in several recent studies (e.g., Delsontro et al., 2010; Schubert et al., 2012; Shakhova et al., 2013), but only during stormy weather. Interestingly, during the dry, hot summer the volume of gas evading from the sediments was considerably lower, but it contained a higher proportion of CH_4. This inter-annual variability may have resulted from the difference in the size of bubbles, with smaller ones rising more often from the sediments during the more frequent mixing events in 2011. Relatively smaller bubbles would have lost relatively more of their initial CH_4 (Ostrovsky et al., 2008).

Weather patterns and oxygen availability may have also mediated the cycling of CH_4 by microbial processes. Significantly more dissolved CH_4 was found in lake water after summer rainfall events when the lake was more strongly stratified. The stratification could have allowed a more efficient exploitation of this carbon source by methanotrophic bacteria and a greater input of CH_4-derived carbon to higher trophic levels (Bastviken et al., 2003; Sanseverino et al., 2012). In contrast, during stormy weather, most of the CH_4 was released from the sediments as gas bubbles avoiding oxidation in the water column and incorporation to the lake food web. Hence, CH_4 may be a larger carbon source for bacteria during hot and dry weather or in wind-sheltered lakes than during stormy weather or in wind-exposed lakes (Kankaala et al., 2013).

In summary, depending on weather conditions, we show that small and shallow temperate lakes can be polymictic or remain stably stratified for periods longer than a month. Our correlative data indicated that rainfall and associated runoff introduced carbon directly into the lake with inflows generally near the bottom. The duration and extent of stratification moderated both the vertical distribution and processing of CO_2 and CH_4, and their flux at the air-water interface. The frequency of frontal events and the magnitude of wind and rain associated with these fronts caused year-to-year variability in GHG emissions and storage. Climate induced modification of the frequency of weather events such as rainstorms and concomitant changes in thermal stratification thus can lead to different pathways of GHG cycling in shallow lakes.

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2.2 Heat wave effects on oxygen, nutrients and global warming potential in a small lake

Effets des vagues de chaleur sur l'oxygène, les nutriments et le potentiel de réchauffement climatique d'un lac de petite taille

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Les données limnologiques et météorologiques ont été collectées et analysées par M. Bartosiewicz, les données de gaz ont été collectées et analysées par M. Bartosiewicz sous la supervision d'Isabelle Laurion. Les données météorologiques ont été collectées et analysées par M. Bartosiewicz. Le manuscrit a été écrit par M. Bartosiewicz et révisé par Isabelle Laurion et Roxane Maranger. Le manuscrit est en revision.

Résumé

Les écosystèmes lacustres sont susceptibles aux changements climatiques qui génèrent une plus forte stratification thermique, ce qui peut altérer le recyclage des nutriments et la production de gaz biogéniques. La présente étude nous permet de tester l'impact du forçage climatique en comparant l'influence de la stratification thermique sur les concentrations d'oxygène, de nutriments et de gaz à effet de serre (CO_2, CH_4 et N_2O), ainsi que sur leurs voies d'émission (diffusion et ébullition) au cours d'une année moyenne (2011) et d'une année record en chaleur (2012). Les résultats montrent que le CH_4 est le plus important contributeur au potentiel de réchauffement climatique (PRC) du lac pendant les deux années. Cependant, les différences de stratification thermique et d’oxygène ont causé des changements dans les voies d’émission, passant d'une dominance par l’ébullition en 2011 (61% du PRC total), à une dominance par la diffusion en 2012 (55% du PRC total). Cette stabilité thermique a aussi causé une forte hétérogénéité temporelle des concentrations en GES, incluant une période de sous-saturation estivale du CO_2 et N_2O et d'un moment chaud pendant la période automnale de mélange alors que 70% des émissions estivales sont survenues. Le lac est demeuré un puits de CO_2 pendant les efflorescences estivales d’algues associées aux conditions chaudes et stratifiées. Fait intéressant, la vague de chaleur n’a pas donné lieu à une hausse du PRC du lac car elle était accompagnée d’une plus forte stratification thermique et des températures hypolimniques plus froides, probablement associées à des taux plus faibles d’ébullition de CH_4 vers l’atmosphère. En effet, le PRC du lac était deux fois plus élevé durant l'année moyenne alors que l'ébullition était stimulée à la fois par l'injection d'eau de surface plus chaude vers les sédiments où la majorité du CH_4 est probablement produite et par le relâchement de bulles emprisonnées dans les sédiments suite aux changements de pression atmosphérique.

Mots clés : lac peu profond, stratification, vague de chaleur, climat, gaz à effet de serre, potentiel de réchauffement global, moments chauds, fleur d’eau de phytoplancton, CO_2, CH_4, N_2O.

Abstract

Lake ecosystems are susceptible to the effects of climate change where warming conditions may result in stronger thermal stratification potentially altering patterns in nutrient cycling and biogenic gas production. In this study we were able to test the impact of climate forcing by comparing the influence of stratification patterns on oxygen, nutrient and greenhouse gas (GHG) concentrations (CH_4, CO_2, N_2O) and emission pathways (diffusion and ebullition) during an average and a heat wave year of a small shallow lake. Results show that CH_4 was the most important contributor to lake global warming potential (GWP) for both years. However, the differences in thermal stratification and oxygen levels resulted in a shift in the dominant CH_4 emission pathway, from ebullition to diffusion during the average versus the heat wave year (61% versus 55% of the total GWP). This stable stratification also lead to a high temporal heterogeneity in GHG concentrations, including periods of undersaturation in CO_2 and N_2O, and a hot moment at fall overturn, when the lake released 70% of total GHG emitted during the entire open water period. Interestingly, the heat wave did not result in higher GWP of the lake as it was accompanied by stronger thermal stratification and colder hypolimnetic temperatures, resulting in lower rates of CH_4 ebullition. Indeed the GWP potential was twice as high during the non heat wave year, due to increased CH_4 ebullition stimulated both by injections of warmer surface waters near active sediments and concomitant release of bubbles from changes in pressure.

Key words: shallow lake, stratification, heat wave, climate, greenhouse gases, macrophyte, global warming potential, hot moments, phytoplankton bloom, CO_2, CH_4, N_2O.

2.2.1 Introduction

Small and shallow lakes are abundant water bodies that play major roles in global carbon and nutrient cycles (Cole et al., 2007; Harrison et al., 2009) and controlling regional climate (Krinner 2003; Downing et al., 2006). The thermal structure, mixing patterns and energy fluxes of lakes are strongly influenced by meteorological conditions (e.g., MacIntyre, 1993). This meteorological forcing drives turbulence both at the air-water interface and within the water column and can therefore influence the gas exchange dynamics within the lake and between the lake and the atmosphere (MacIntyre et al. 1995). Weather conditions during the ice-free season are also responsible for year-to-year differences in nutrient inputs from the watershed, be it total load or timing of delivery (Wilhelm and Adrian, 2008). Consequently, changes in weather conditions can affect both biotic and abiotic components of lake ecosystems, with possible implications on water quality and the biogenic production of greenhouse gases (GHGs).

Lakes of different sizes and mixing regimes have been shown to respond differently to the strength and persistence of meteorological forcing (Gerten and Adrian, 2001). Models simulating the effects of future warming on lakes predict that an increase in average summer temperature and irradiance may lead to greater water column stability in both large (e.g. Peeters et al., 2002) and small lakes (De Stasio et al., 1994; Mooij et al., 2007). However, while changes in stratification of large lakes will be relatively slow in time, the mixing regimes in small lakes will be more strongly influenced by warming over short time-scales mainly due to their larger surface-to-volume ratios. The possible effects of a stronger stratification in shallow lakes may include higher rates of carbon and nutrient remobilization, and a switch from macrophyte domination to phytoplankton blooms (Scheffer et al., 2001).

The extent to which the pelagic and littoral zones of small lakes will respond to warming may differ, particularly when the littoral zone is dominated by macrophytes. For example even in the absence of climate effects, the presence of submerged macrophytes can result in in stronger temperature stratification and a reduction in the mixed layer depth (Herb and Stefan, 2005). Plants also decrease sediment re-suspension rates (Vermaat et al., 2000), reduce littoral-pelagic water exchange (Barko and James 1998) and can also act as vectors for gas transport from the sediment to surface waters and atmosphere (Dacey, 1980). Furthermore, macrophyte litter decay can enhance anoxic conditions and provide readily available substrates for methanogens (Chasar et al., 2000) as well as heterotrophic bacteria including denitrifying bacteria (Brix, 1997). Through these various effects, macrophytes influence oxygen dynamics and nutrients cycling in both the sediments and water column, changing conditions for GHG production (Van Der Nat and Middelburg, 1998; Caraco et al., 2006; Tall et al., 2011). Thus a differential or stronger response in both physical structure and subsequent biogeochemical transformations could be expected between pelagic and littoral zone of lakes under conditions of warmer climate (Havens and Steinman, 2013).

Summer heat waves have become more frequent over the last few decades (Stott et al., 2004) as a result of global climate change (Schär and Jendritzky, 2004) and these events will likely influence the physical structure and subsequence functioning of lakes. Indeed the summer heat wave of 2003 lead to a modification in the abiotic and biotic characteristics of Lake Zurich, where a dramatic increase in water column stability resulted in significantly higher hypolimnetic oxygen depletion (Jankowski et al., 2006). This observation supports the predictions of climate models that suggest warming will increase oxygen deficits in the hypolimnion of lakes, which may subsequently result in enhanced release of phosphorus (P) from the sediments and increased primary production (Sondergaard et al., 2001; Jeppesen et al., 2003). Other studies have shown that stronger stratification, higher P availability and primary production in lakes could also lead to an increased production and storage of more potent GHG such as methane (CH_4) (Bastviken et al., 2008; Bartosiewicz et al., 2015) and possibly nitrous oxide (N_2O) (Huttunen et al., 2003). However the release of these GHG from lakes depends on mixing and turbulence, which is controlled by both wind and heat fluxes (MacIntyre et al., 2010). Therefore specifically how the change in stratification during a heat wave will influence nutrient cycling, algal biomass as well as GHG speciation and emissions remains poorly understood.

In this study we assessed the influence of hot and dry weather during the most recent North American heat wave (Mills et al., 2013) on the stratification, nutrients, chlorophyll and oxygen levels in the pelagic and littoral zones of a small and shallow lake. We compared the resulting pattern of CO_2, CH_4 and N_2O concentrations, including GHG fluxes from both diffusion and ebullition and the overall global warming potential (GWP) of a shallow lake during a relatively wet and an extremely dry year.

2.2.2 Methods

Study site

Small (0.18 km^2) and shallow Lake Jacques (mean depth 0.75 m) is located 30 km from Quebec City (Canada, Fig. 2.2.1). The littoral zone of the lake (0.07 km^2) is occupied by an invasive macrophyte (*Brasenia schreberi* sp.), commonly known as the water shield. The surface sediments of the lake have a high organic matter content both in the littoral (loss-on-ignition: LOI=45 ± 7%, n=10) and pelagic zones (LOI=38 ± 5%, n=10). The catchment of Lake Jacques covers 5.92 km^2 and is occupied by residences (on the west side of the lake) and forest (on the east side of the lake). The lake is supplied with water from two small creeks, both located in its eastern part and an underground spring located in its southern part. The water outflow ranges from 0.2 to 0.3 m^3 s^{-1} in spring and 0.02 to 0.05 m^3 s^{-1} in summer. Water retention time varies from 2 to 14 days in spring and summer, respectively.

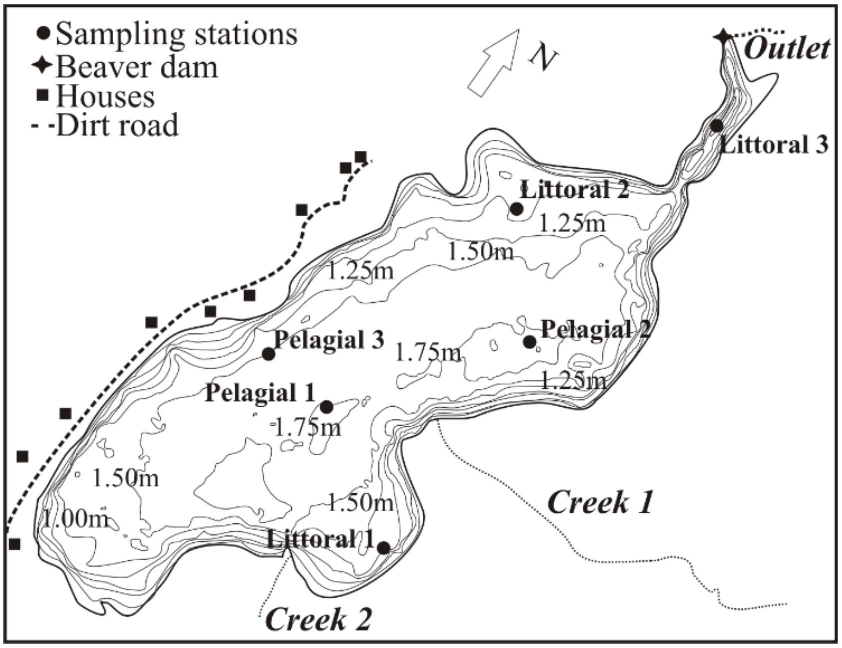

Figure 2.2.1 Morphometry and bathymetry of Lake Jacques. The thermistor chains were located at sites Pelagic 1 and Littoral 1 (only in 2012), and the greenhouse gases were sampled at sites 1-2 in 2011 and Sites 1-3 in 2012.

Physicochemistry

Surface meteorology and physicochemistry was followed in Lake Jacques as described in details by Bartosiewicz et al. (2015). Furthermore, in both 2011 and 2012 water samples were collected at the surface of the littoral and pelagic sampling sites (Fig. 2.2.1), filtered through pre-rinsed cellulose acetate filters and analyzed for soluble reactive phosphorus (SRP, persulfate digestion after Stainton et al., 1977) and dissolved nitrogen (NO_3^-, alkaline persulfate digestion). Non-filtered water was acidified with H_2SO_4 (0.15% final concentration) and stored at 4°C until samples were analysed for total phosphorus (TP, spectrophotometry after persulfate digestion) and total nitrogen (TN, colorimetric method after potassium persulfate digestion in alkaline conditions as nitrate-nitrite by reduction with hydrazine). Water samples (100–500 ml) were filtered on MFS GF/F filters for the determination of chlorophyll-a concentrations (Chl-*a*) following the extraction method of Wintermans and De Mots (1965) and quantified by UV–Vis spectrophotometry.

Time series on the thermal structure of the water column at the pelagic (as in Bartosiewicz et al. 2015) and littoral station were obtained with a thermistor chain installed from June to September 2011 and from May to October 2012. The mixed layer depths were calculated following MacIntyre et al. (2002) based on density criteria. Buoyancy frequency (N, in cycles per hour or cph), describing water column stability was computed as $N^2 = g\,\rho^{-1}\,d\rho/dz$, where g is the gravity constant and ρ the water density.

Greenhouse gas concentration and global warming potential

Aqueous CO_2 and CH_4 concentrations were assessed by equilibration of 2 liters of lake water (4 sites in 2011 and 6 sites in 2012, Fig. 2.2.1) into 20 mL of ambient air during 3 min of vigorous shaking. The headspace was sampled in 5.9 ml Exetainers (Labco) previously flushed with helium and vacuumed sealed (Hesslein et al., 1990), and the procedure was repeated 3 times at each sampling site. Gas samples were taken within 5 min after collecting the lake water and kept in the dark at 4°C until analyzed by gas chromatography (Varian 3800 with a COMBI PAL head space injection system). Aqueous N_2O concentrations were collected in a similar manner, but the samples were injected into 9 mL pre-evacuated vials and concentrations were determined by gas chromatography using an ECD detector on a Schimadzu 2014 GC with a Tekmar 7050 autosampler. We used a Poropaq Q (80/100) column to separate gases with P5 (95% argon and 5% methane) as the carrier gas.

In addition, the sediments porewater samples were collected by in situ dialysis at 1-cm vertical sampling resolution using acrylic peepers (Carignan et al., 1985). The peepers were deployed, by skin diving, at the littoral and pelagic zones of Lake Jacques in July and August 2012 (at the beginning and by the end of summer stratification) and left for 14-21 days to equilibrate. Peepers were installed to collect water samples down to 10 cm below the sediment interface and up to 5 cm above. Before use, the peepers were prepared under a N_2 atmosphere as in Tessier et al. 2014. Upon retrieval, the peepers were sampled as described by Alfaro-De La Torre and Tessier (2002) in order to obtain two or three replicate profiles of CH_4 by equilibration of 2 ml of porewater from each depth in evacuated 3.7ml Exetainer. Porewater CH_4 profiles were used to obtain CH_4 production and consumption rates in the sediments of Lake Jacques following method of Berg et al. (1998). Fluxes of CH_4 at the sediments-water inteface were calculated using Fick's equation (Boudreau 1997) and assuming temperature dependent transfer coefficient from Oelkers (1991). For porosity data, sediments cores (0-15cm) were obtained at the peeper deployment sites, sectioned on the shore at 1-cm intervals from the sediment interface to 15 cm depth, transported to the laboratory in plastic bags located in a container filled with fresh bulk sediments to limit atmospheric exchange and oxidation, and stored in -80°C until analyzed for water content within 2-3 weeks after collection.

The diffusive GHG flux at the lake surface was estimated as: $Flux_d = k\ (C_{sur} - C_{eq})$, where k is the gas transfer coefficient (cm h^{-1}), C_{sur} is the gas concentration in surface water (mmol L^{-1}), and C_{eq} is the gas concentration in equilibrium with the atmosphere. The gas transfer coefficient was taken from Cole and Caraco (1998) wind based model, where $k_{600} = 2.07 + 2.15\ (u_{10})^{1.7}$, after transformation with $k = k_{600} \times (Sc/600)^{1/2}$, where u_{10} is a wind speed at 10m and Sc is the Schmidt number for a given gas. The use of Cole and Caraco model may result in conservative estimations of fluxes as demonstrated by Bartosiewicz et al. (2015) in the pelagic zone of this lake. However, for any given wind, turbulence at the lake surface will be greater offshore than inshore (Schilder et al. 2013) thus such conservative estimates may be better suited to evaluate GHG diffusion in the littoral zone, and over the entire lake area when meteorological and thermal structure data are limited. The chemical enhancement factor, by which gas transfer coefficient is multiplied for CO_2 flux estimations, was calculated following the method described by Hoover and Berkshire (1969). The multiplier on k was significantly different from 1 (up to 1.97) in July and August 2012. Chemical enhancement refers to the direct hydration of CO_2 with hydroxyl ions. This occurs mostly when hydroxyl ions are abundant (high pH) and more so at high temperatures and under low wind conditions.

The GHG bubble flux (ebullition) was measured 4 times in 2011 and 5 times in 2012, with 4 to 9 submersible inverted funnels located either in the pelagic or in the littoral zone of the lake for 4-24h depending on flux rates. The collected bubbles were sampled in triplicate vials (5.9 ml Exetainer), and analyzed via gas chromatography as described above. The bubble flux was calculated as: $Flux_e = C\ V_g\ V_m^{-1}\ A^{-1}$, where C is the concentration of a given gas in the syringe, V_g is the total volume of the gas in the syringe, V_m is the gas molar volume at ambient temperature obtained from the meteorological station, and A is the area of the funnel.

The GHG fluxes calculated for the pelagic zone (site 1 in Bartosiewicz et al. 2015, in addition to sites 2 & 3 in the present study) and for the littoral zone were used to calculate the open water global warming potential (GWP) of gases evading from Lake Jacques. This was done by multiplying fluxes by 1 for CO_2, 105 for CH_4 and 298 for N_2O in order to get values in CO_2-equivalent units. These multiplications factors correspond to estimations of the climate forcing by CH_4 over a 20-year horizon and by N_2O over a 100-year horizon (Schindel et al. 2009).

2.2.3 Results

Surface meteorology and lake mixing

The weather in 2012 was hotter and drier than in 2011, particularly during the summer heat wave which lasted from DOY 178 to 257 (for details on surface meteorology in Lake Jacques during the study, see Bartosiewicz et al. 2015). Despite its shallowness, the lake was stratified in both years, but hot and dry weather resulted in stronger stratification. Buoyancy frequencies (N) were persistently high (50 cph) during summer 2012, indicative of reduced vertical exchanges within the water column. In contrast, there was more intensive mixing of the water column in summer 2011 as N often dropped to 30 or 40 cph after a rainstorm. Assuming mixing efficiency, defined as the ratio of potential energy gain to dissipation, at $\Gamma = 0.2$ and kinetic energy dissipation rate $\varepsilon = 10^{-7}$ m^2 s^{-3}, we calculated vertical distribution of eddy diffusivity as $Kz = \Gamma\ \varepsilon / N^2$. This parameter was used to calculate the time scales of mixing, given by $\tau = L^2/ Kz$, where L is the mixing depth and Kz is the vertical distribution of eddy diffusivity. It averaged 4 hours to mix 0.4 m of water column in 2011, whereas it was taking 15 hours for same amount of mixing in 2012 (0.4 m was used as the depth required to mix down through the thermocline or below). The mixed layer depth (MLD) was on average 30% shallower in 2012 than in 2011 (0.46 m versus 0.31 m in 2011).

Oxygen stratification

Persistent oxygen stratification was observed in both years, but the extent and hypoxic volume was much larger in the heat wave year (on average 28339 m^3) than in 2011 (7878 m^3, $p=0.001$, Student's t-test). During the heat wave, the oxygen was depleted in the hypolimnion beginning from May 2012 and continuing up to the mixing event in mid-September (Fig. 2.2.2). The sporadic rainfall events in July and August 2012 did not disturb this oxygen stratification. Interestingly, macrophytes did not result in any apparent oxygen increase in the lower water colmn of the littoral zone during the summer. There was significantly less oxygen in the hypolimnion during the summer heat wave than during the previous summer (4.77 and 8.16 mg L^{-1}, respectively; paired Student's t-test, $p<0.001$). However, surface oxygen concentration was higher in 2012 than in 2011 ($p<0.01$), with maxima in the pelagic zone reaching about 13 mg L^{-1}. In 2012, surface oxygen levels were also higher in the macrophyte-free pelagic zone of the lake than in its littoral zone ($p=0.045$).

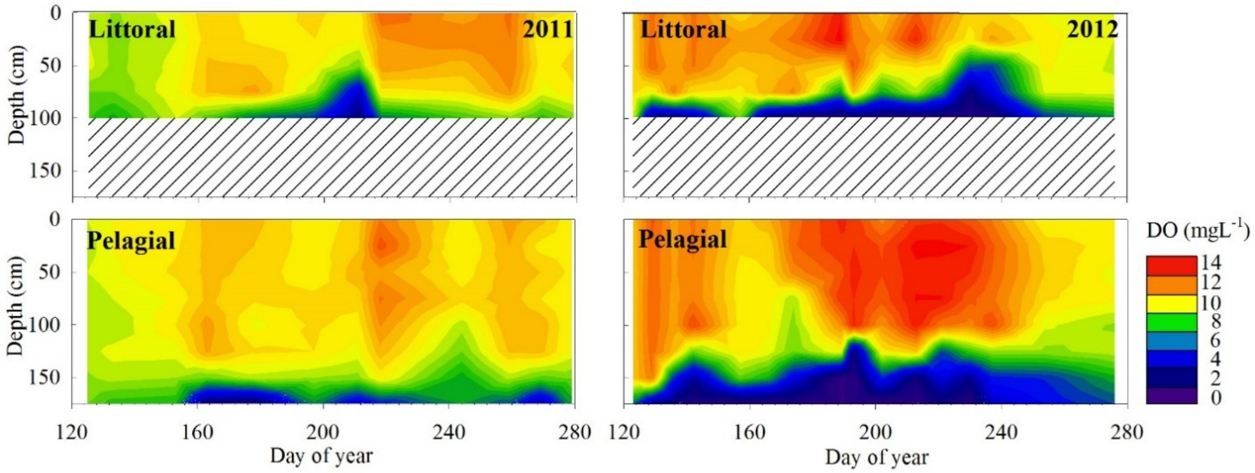

Figure 2.2.2 Temporal evolution of dissolved oxygen (DO) stratification in the littoral (upper) and pelagic (lower panels) zone of Lake Jacques in 2011 (left) and during the heat wave of 2012 (right panels).

Nutrients and phytoplankton biomass

Seasonal changes in nutrient concentrations appear to be related to differences in the frequency of heavy rain events (>10 mm per day) and extent of water column mixing between years (Fig. 2.2.3, Table 2.2.1).

Year	2011			2012		
Variable	Temp	MLD	Oxy	Water T	MLD	Oxy
TP	**0.571**	**0.562**	-0.290	0.148	-0.001	-0.259
TN	**-0.562**	0.121	**0.502**	-0.144	-0.146	-0.228
SRP	0.092	**0.390**	0.145	**-0.603**	0.116	**0.500**
NO_3^-	**-0.389**	0.207	**0.536**	**-0.509**	**0.420**	**0.569**
Chl-*a*	**0.715**	**0.459**	**-0.562**	**0.530**	-0.159	**-0.629**

Table 2.2.1 Pearson's correlation coefficients obtained between surface water temperature (Temp), mixed layer depth (MLD), oxycline depth (Oxy), and total phosphorus (TP), total nitrogen (TN), soluble reactive phosphorus (SRP), nitrates (NO_3^-) as well as chlorophyll-a concentration (Chl-*a*) during 2011 and 2012 summer heat wave. All data were normalised taking the napierian logarithm.

In 2011, TP concentrations followed a similar pattern in the littoral and pelagic zones and were related to temperature and MLD. Concentrations were higher in summer 2012 (24 compared to 17 µg L^{-1} in 2011, $p = 0.03$, Mann-Whitney test) and no significant correlations with vertical structure were found. The TP patterns were less variable in the pelagic zone in 2012 (CV = 33%) than in 2011 (47%) and more variable in the littoral zone (CV = 54% in 2012 compared to 46% in 2011). Seasonal changes in dissolved P and N were markedly different between years and did not clearly follow the rainstorms. In summer 2011, SRP concentrations reached maximum values of 4 or 5 µg L^{-1} (in the pelagic and littoral zone, respectively) and were never depleted below 0.3 µg L^{-1}. In 2012, SRP peaked in the pelagic zone after the initial springtime mixing, and then decreased markedly (below the detection limit) in both littoral and pelagic zones. Nitrate was considerably higher and more variable in 2011 as compared to 2012 (Fig. 2.2.3 G and H; 0.29 mg L^{-1} versus 0.09 mg L^{-1}, CV = 105% versus 68% in 2012). Periods of depletion were observed in both years but more so in 2012.

In summer 2011, there was no major phytoplankton accumulation in surface waters (< 25 µg Chl-*a* L^{-1}), and no trends were observed between the pelagic and littoral zone. On the contrary, phytoplankton biomass increased steadily after the onset of stratification in June 2012, reaching 40 and 60 µg Chl-*a* L^{-1} (pelagic and littoral zones respectively) during the mid-summer algal bloom. Thus, significantly more Chl-*a* was found in surface waters of the lake in 2012 than in 2011 (on average 25.6 compared to 10.2 µg L^{-1}, $p=0.005$, Mann-Whitney U-test). Moreover, bloom-forming cyanobacterial species such as *Anabaena* sp. and *Aphanizomenon* sp. were regularly observed during the hotter summer (up to 40% of the total phytoplankton biovolume, data not shown).

Greenhouse gas concentrations and the global warming potential

Patterns in the deviation from saturation (Δ) for all gases differed between the two years, with the most striking difference being the peak in saturation of all three gases at overturn in 2012 in both littoral and pelagic zones representing a hot-moment (Fig. 2.2.4). Indeed the values of 172 µM, 11.5 µM and 26.6 nM for ΔCO_2, ΔCH_4 and ΔN_2O are among the highest surface water concentrations to be observed. Overall the average ΔCO_2 of surface waters were significantly higher in 2011 than in 2012 (24.9

and 17.5 µM respectively, p=0.001, Mann-Whitney U-test, Fig. 2.2.4). The lake was continuously supersaturated in CO_2 in 2011, but was persistently undersaturated during the summer heat wave of 2012.

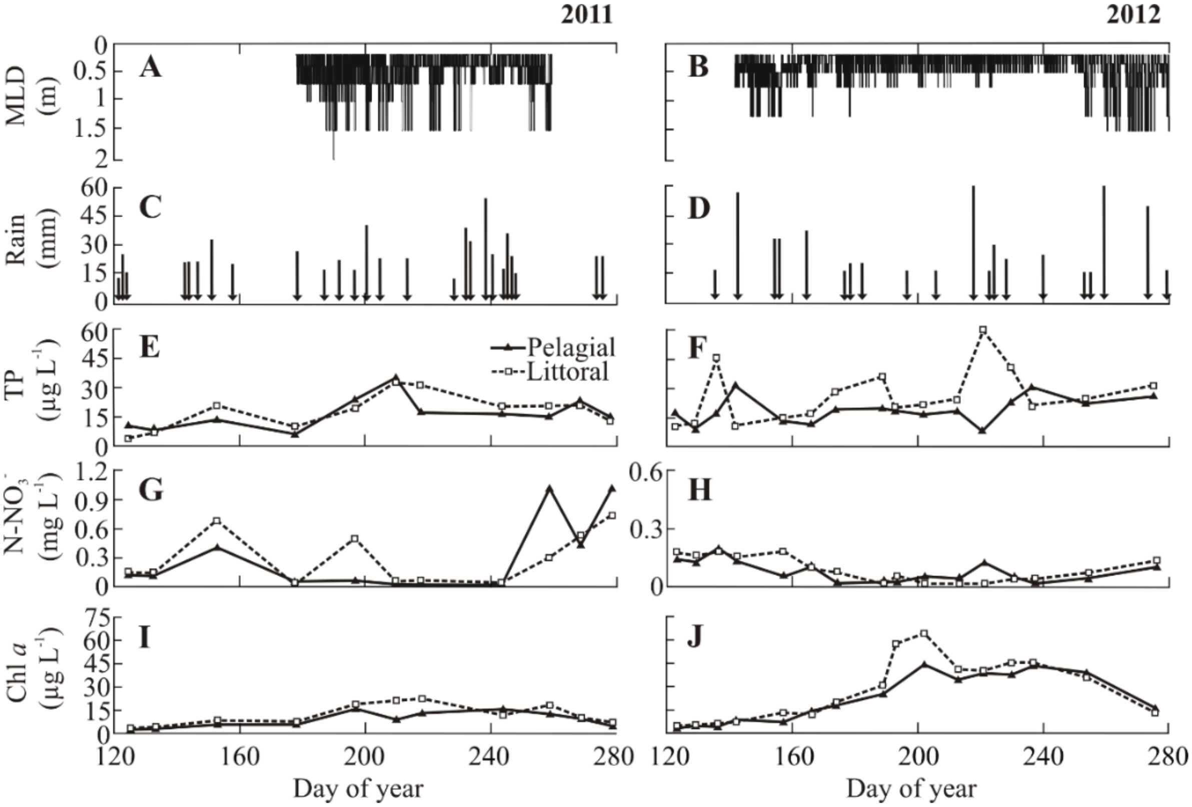

Figure 2.2.3 Seasonal dynamics of mixed layer depth (MLD), rainfall events (arrows indicate rainfall above 10 mm d^{-1}), surface water nutrients and phytoplankton biomass (Chl-*a*) in the littoral (dotted lines) and pelagic (solid lines) zones of Lake Jacques in 2011 and during the heat wave of 2012.

This undersaturation event coincided with strong thermal stratification and high chlorophyll a concentrations (Fig. 2.2.3 B and D; Fig. 2.2.S1). In both years, ΔCO_2 was negatively correlated to ΔO_2, but the relationship was noticeably stronger during the heat wave (Fig. 2.2.5). On average ΔCO_2 was higher in the littoral zone (littoral =29.5 µM, pelagic = 5.9 µM) and patterns were more synchronous between zones during the heat wave year (Fig. 2.2. 4 A and B). Departure from saturation for dissolved CH_4 (ΔCH_4) at the lake surface was on average significantly higher in 2012 than in 2011 (1.61 µM versus 0.74 µM, p=0.014, Mann-Whitney U-test Fig. 2.2.4) but this was due primarily to the much higher concentrations measured at autumnal overturn.

Concentrations were also higher in the littoral than in the pelagic zone on both years ($p < 0.001$). The highest dissolved ΔCH_4 concentrations were recorded by the end of summer 2012. In both years, ΔCH4 was negatively related to NO_3^-. Interestingly in 2011, ΔCH4 was negatively related to TP, but in 2012 was positively related to chlorophyll *a* (Fig. 2.2.S1). Moreover, in 2012 but not in 2011, ΔCH_4 positively correlated to ΔO_2 (Fig. 2.2.5).

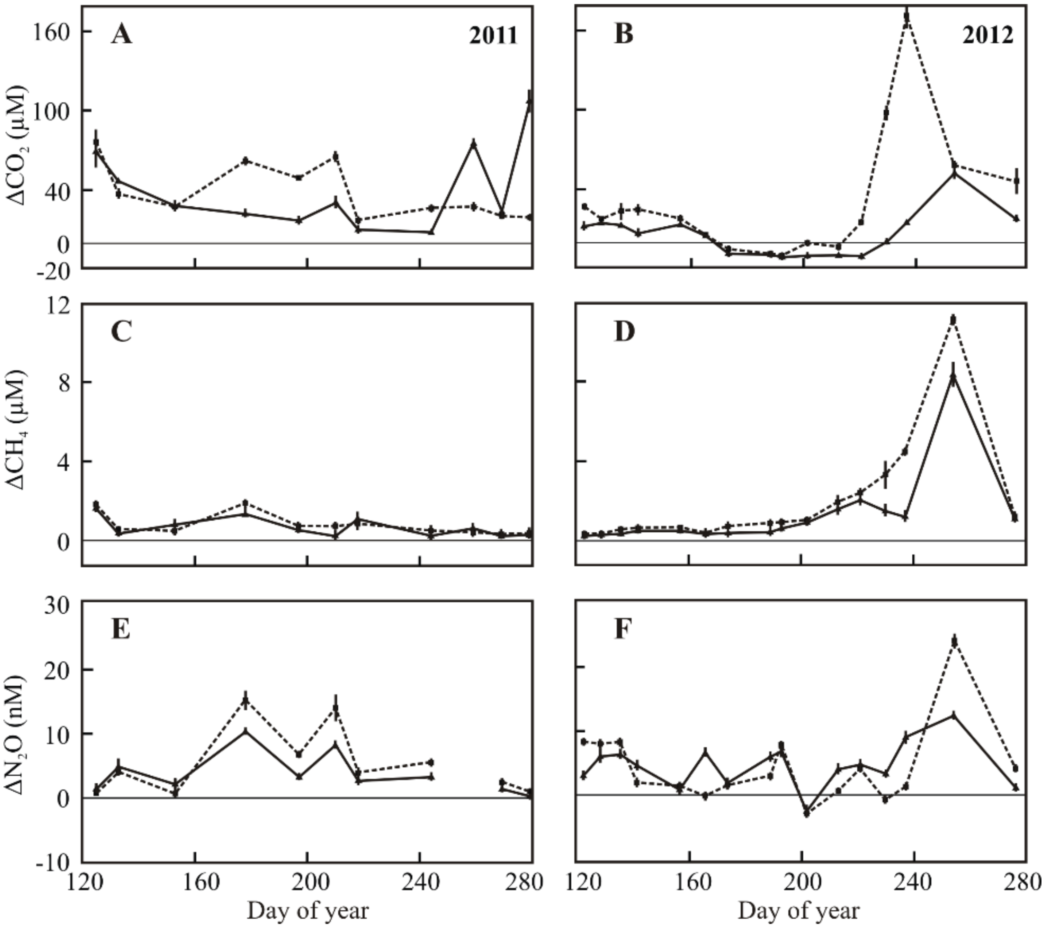

Figure 2.2.4 Departure from saturation (Δ) of CO_2, CH_4 and N_2O at the lake surface in the littoral (dotted lines) and pelagic (solid lines) zone of Lake Jacques in 2011 and during the heat wave of 2012.

The porewater sampling revealed that during the heat wave, CH_4 was produced at high rates in both littoral and pelagic shallow sediments (mainly between 0 and 4 cm depth). CH_4 was also partly consumed in situ at the littoral zone, whereas at the pelagic zone it evaded without being oxidized (Fig. 2.2.S3, Table 2.2.S1). There was significantly more CH_4 in the littoral than pelagic sediments, and there was a significant increase in the sedimentary CH_4 concentrations between sampling dates, but only in the pelagic zone (p=0.001, Paired t-test).

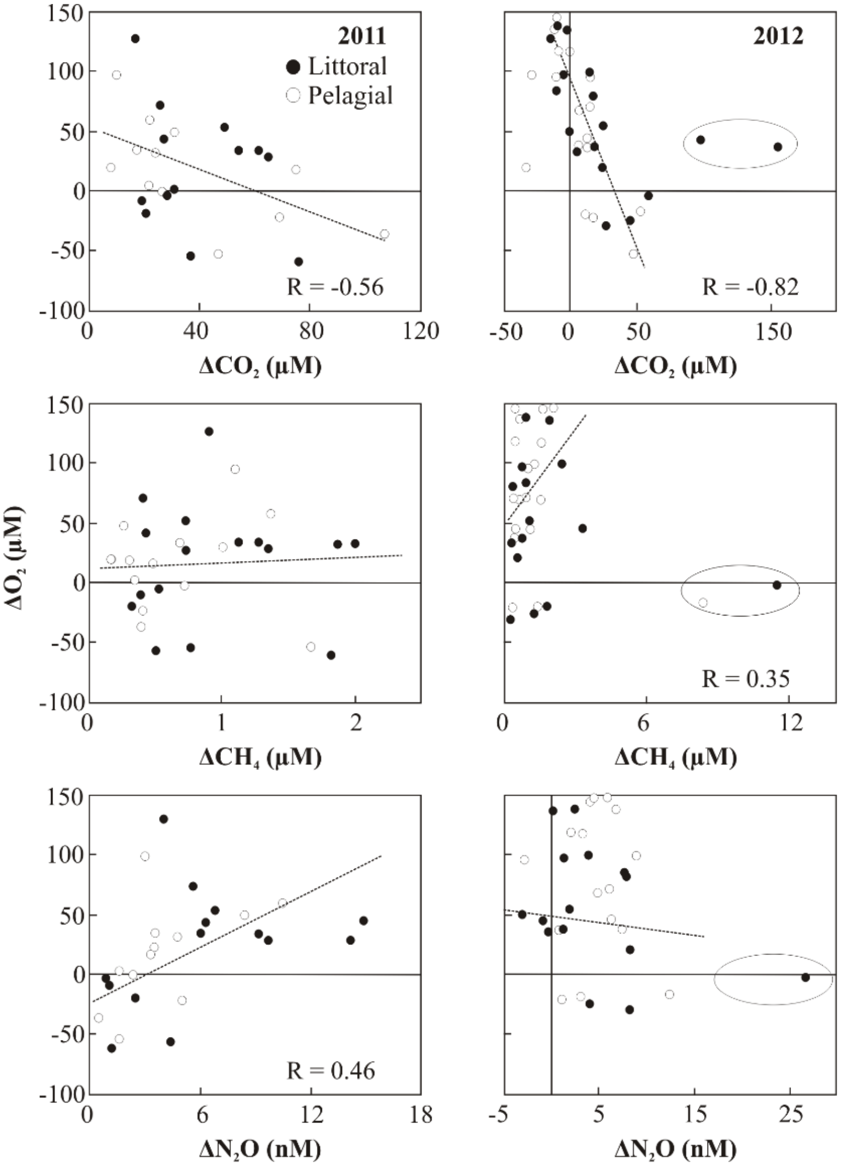

Figure 2.2.5. Correlations between oxygen and GHG (CO_2, CH_4 and N_2O) in the littoral (black) and pelagial (white circles) zones of Lake Jacques in 2011 (left) and 2012 (right panels). P<0.01 when R is indicated.

For ΔN_2O, no significant differences in overall concentrations were observed between years (p=0.09). However, during the heat wave, N_2O concentrations were typically lower and on occasion surface waters were undersaturated. While in 2011, ΔN_2O decreased with increasing NO_3^- concentrations and increased with surface temperature (Fig. 2.2.S1), no significant relationships with environmental variables were observed in 2012.

Reference	Location	CH_4 production ($pmol\ cm^{-2}\ s^{-1}$)	Method
Sivan *et al.*, 2007	Marine sediments	0.001–0.002	Numerical model (DIC)
Beer *et al.*, 2008	Peatland	1.1–3.0	Inverse modelling (CH_4)
Segarra *et al.*, 2013	Brackish wetland	1.5	Incubation experiments
	Freshwater wetland	1.1	Incubation experiments
Norði *et al.*, 2013	Lake Ørn, Denmark	0.3	Inverse modelling (CH_4)
Lofton *et al.*, 2014	Alaskan lakes	0.03–0.2	Incubation experiments
Adler *et al.*, 2011	Lake Kinneret, Israel	4.0	Numerical model (DIC)
This study	Lake Jacques, Canada	1.4–4.7	Inverse modelling (CH_4)

Table 2.2.S1 Comparison of CH_4 production rates in various aquatic environments.

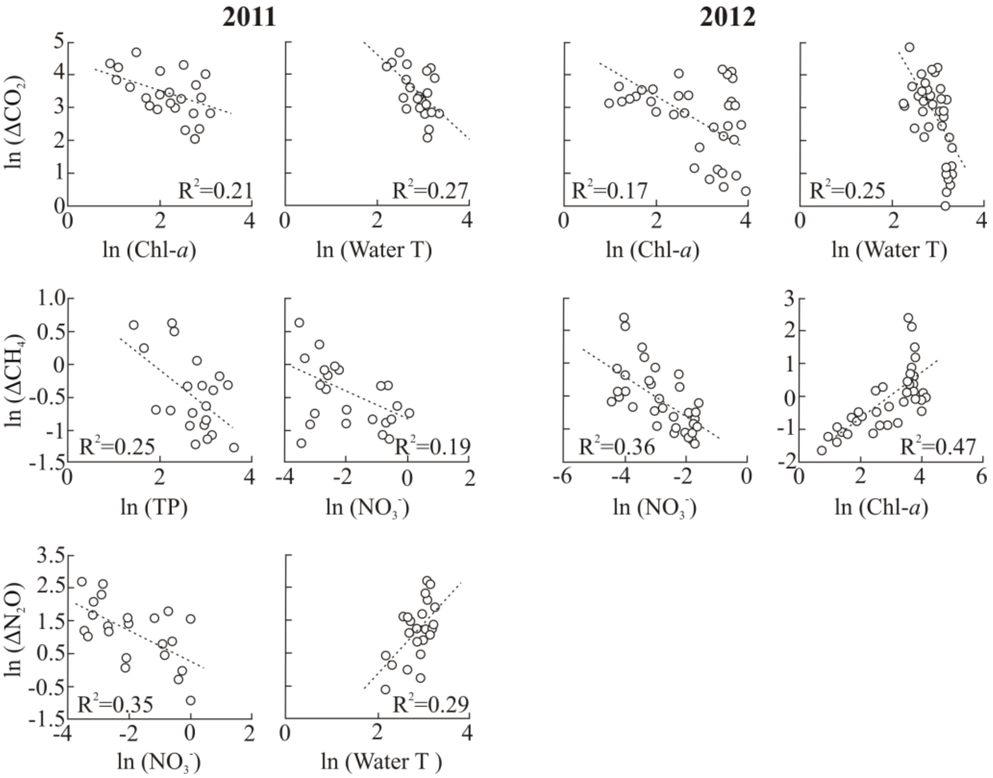

Figure 2.2.S1 Relationships between GHG concentrations and some limnological characteristics at the surface of Lake Jacques in 2011 (left) and during the summer heat wave of 2012 (right panels). Data were normalised taking the napierian logarithm. P < 0.05 for all regression lines.

When all GHG diffusive fluxes were normalized to their GWP, seasonal patterns were more similar in 2011 (CV = 57 and 59% for littoral and pelagic zones, respectively) than in 2012 (CV = 141 and 180%) (Fig. 2.2.6). Indeed, the heat wave resulted in a major hot moment during fall overturn in both the pelagic and littoral zones of the lake where GWP of diffusing gases increased by up to a factor of 4 compared to rest of that year.

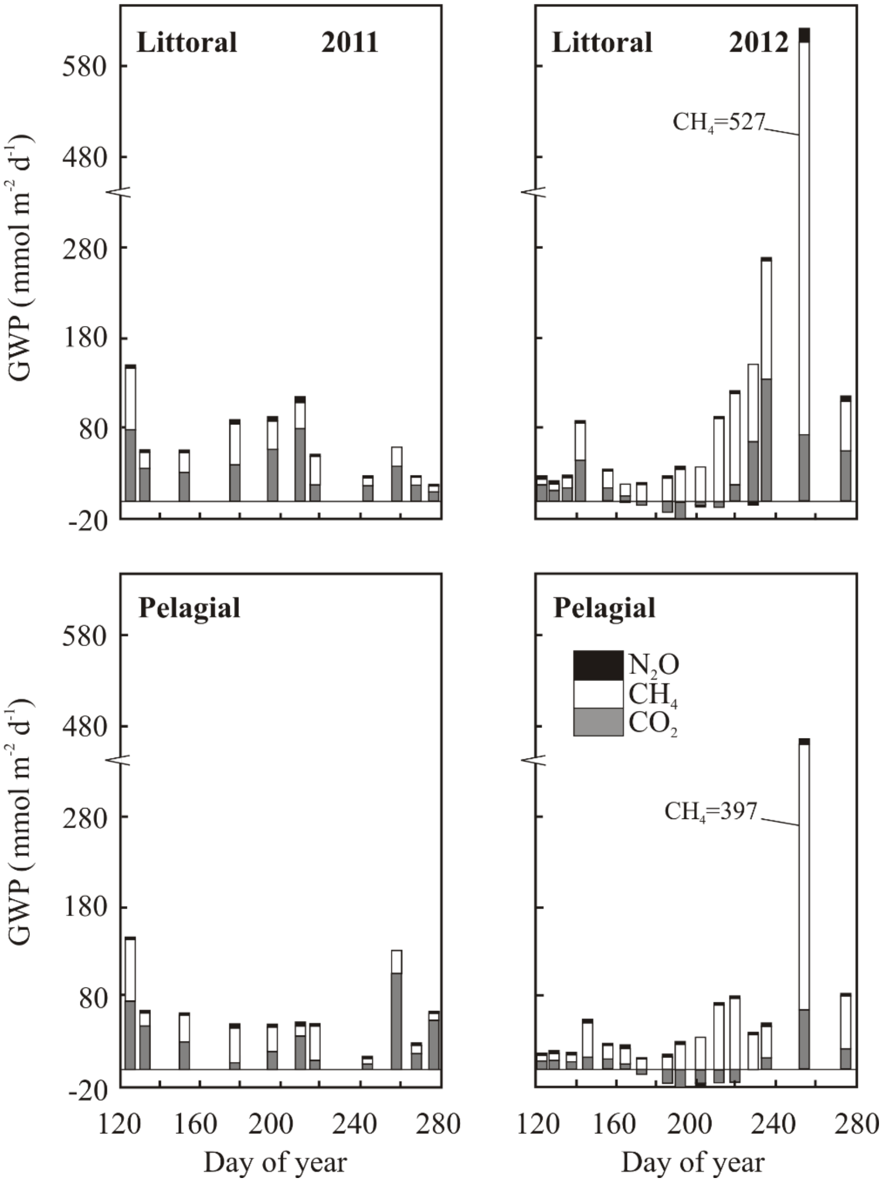

Figure 2.2.6 Open water global warming potential (GWP, given as moles of CO_2-equivalent per day) of diffused CO_2, CH_4 and N_2O evading from the littoral (upper) and pelagic (lower panels) zone of Lake Jacques 2011 (left) and 2012 (right panels).

When comparing years, cumulative peak values during the hot moment in the pelagial zone were also approximately 4 times higher (468 and 125 mmol CO_2 m^{-2} d^{-1} in the pelagial of the lake in 2012 and 2011, respectively). The C gases were the most relevant gases in GWP in both years, however throughout the wet year, CO_2, with the exception of one date, contributed more to GWP (up to 80%) then diffusive CH_4, whereas during the heat wave, the importance was reversed, where CH_4 contributed up 98%. N_2O contributed minimally to GWP in both years, ranging from 0.1 to 11%. In order to estimate overall annual GWP of the lake during the open water period, we needed to consider GHGs emissions during ebullition. On average bubble flux of CH_4 and N_2O was significantly higher from the pelagic zone in both years (Fig. 2.2.S2) and overall much higher from both zones in the lake in 2011 (Table 2; $p = 0.01$, Student's t test; up to 4 and 7 times higher in littoral and pelagic zone, respectively). Bubbles contained up to 51% of CH_4 and up to 17 ppm of N_2O, while CO_2 partial pressure was negligible. This resulted in a significant proportion of emissions of both of these gases through ebullition (Table 2.2.2).

GWP (mmol CO_2 m^{-2} d^{-1})	2011		2012	
	Littoral	Pelagial	Littoral	Pelagial
dCO_2	37.7 ±22.6	37.3 ±29.4	26.0± 39.7	3.7 ±20.9
dCH_4	27.3 ±17.3	24.7 ±17.1	76.3± 138.8	55.4 ±90.7
dN_2O	1.6±1.2	1.1 ± 0.8	1.3± 2.7	1.4 ±1.2
eCH_4	93.5 ±35.3	151.4 ±45.7	25.5± 8.2	22.4 ±11.2
eN_2O	2.0 ±0.6	2.2 ±0.6	1.0 ±0.1	1.0 ±0.1
Total	**162.1**	**216.7**	**130.0**	**83.9**

Table 2.2.2 The average open water global warming potential (GWP) of GHG emitted by diffusion (*d*) and by ebullition (*e*) from the littoral and pelagial zones of Lake Jacques in 2011 and 2012. The littoral zone represents 39% of the total lake area.

Although the sum of CH_4 emissions was contributing most to GWP during both years, striking differences in net fluxes and emission pathways were observed. Firstly, overall emissions were approximately two times higher in the wet year, 2011. The dominant CH_4 emission pathway was through ebullition in 2011 (64% of total emissions), whereas diffusive fluxes were more important in 2012 (61%). Diffusive CO_2 flux contributed more than diffusive CH_4 flux to GWP in the wet year (21% and 14%, respectively), but less in the heat wave year (14%). Neither diffusive nor ebullitive N_2O flux contributed significantly to the GWP of the lake (>5%). However, the amount of N_2O released via ebullition represented a significant fraction of the total emitted in both years, around 40% in 2012, and 60% in 2011. The GWP differences between years were largely due to higher CH_4 ebullition from both the littoral (4 times higher) and pelagic zones (8 times higher) in 2011 versus 2012. When ebullitive CH_4 flux was accounted for, the effect of summer heat wave did not result in a significant increase in the GWP of this small and shallow lake.

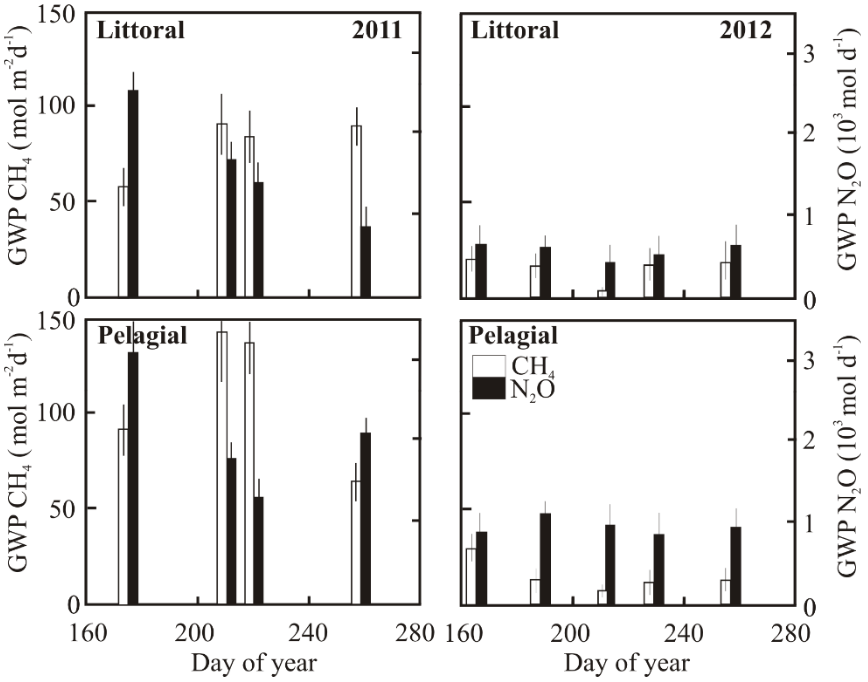

Figure 2.2.S2 Global warming potential (given as moles of CO_2-equivalent per day) of CH_4 (values on left axis) and N_2O (values on right axis) released by ebullition from the littoral (upper) and pelagic (lower panels) zone of Lake Jacques in 2011 (left) and 2012 (right panels). The littoral zone represents 39% of the total lake area.

2.2.4 Discussion

This study clearly shows the strong influence of a summer heat wave on the physical structuring and subsequent functioning of a small shallow lake ecosystem. The pronounced effects of climate forcing on oxygen and temperature stratification resulted in differential GHG speciation, temporal patterns and emission pathways. Contrary to what has been suggested in the literature the warmer temperatures brought on by the summer heat wave did not result in an overall increase in GWP for the present study lake (Moss et al., 2011; Yvon-Durocher et al., 2014; Marotta et al., 2014). Indeed the overall GWP of the lake was almost double during the wetter, more average year. The assumption that systems will emit more GHGs when conditions are warmer is primarily based on the impact of increased temperatures on the different microbial metabolic processes that result in enhanced gas production. This is expected to impact GWP even more strongly since it is anticipated that more CH_4 will be produced relative to CO_2 given the higher activation energy of methanogenesis relative to heterotrophic respiration (Yvon-Durocher et al., 2011). However what occurred during the heat wave was a multi-fold effect on emissions where increased water column stability resulted in significantly lower overall hypolimnetic temperatures likely limiting GHG production, decreased rates of ebullition and enhanced phytoplankton production resulting in periods of undersaturation of both CO_2 and N_2O, thus reducing total emissions.

Overall CH_4 was the most important contributor to the GWP of the lake during both wet and dry summers, representing 78 and 83% of total GWP respectively, which is typical for small and shallow lakes (e.g. Casper et al., 2000). However, the differences in the water column mixing regime and oxygen levels between summers resulted not only in a change in the total amount of GHG emitted but also in a shift in the dominant CH_4 emission pathway, from relatively more via ebullition in 2011 to more via diffusion in 2012. The total amount of CH_4 emitted via ebullition was over 5 times higher in 2011. This was due to the greater volume of bubbles emitted since the proportion of CH_4 in the bubbles was higher during the heat wave year (on average 40%, compared to 30% in 2011). On the other hand, diffusive CH_4 (*d*CH_4) loss during the open water season was nearly 2.5 times higher during the heat wave. Concurrently to this shift in dCH_4 loss, the relative importance of diffusive CO_2 (dCO_2) flux was inversed. Interestingly, the total GWP from diffusive fluxes was similar between years at 10.5 and 11.1 × 10^3 mol CO_2 d^{-1} in 2011 and 2012 respectively, but the ratios of the GWP of open water dCH_4:dCO_2 were widely different at 0.5 and 3.3. Ratios reported in the literature are typically less than 1 (e.g. Kling et al., 1992; López Bellido et al., 2009), but these are most often applied to incubation estimates and not entire seasonal averages. While during the entire open water season of 2011 the lake acted as a source of CO_2 to the atmosphere, net emissions of CO_2 were the case only at the overturn period in 2012. The lake was a CO_2 sink during the summer algal bloom arising under the warm and more stratified conditions. Indeed this represented a shift from net heterotrophy to net autotrophy as a function of meteorological forcing (Carpenter and Pace, 1997; Staehr et al., 2010).

Factors that influence CH_4 ebullition were perhaps the most critical at nearly doubling the GWP during the non heat wave year. These include increases in energy inputs to the sediments (i.e. higher temperature) as raised by Wik et al. (2014), and reductions in pressure, either through changes in water table (Chanton et al., 1989) or atmospheric pressure (Mattson and Likens, 1990). It was reported in small shallow Mirror Lake that CH_4 ebullition rates increased 3 fold with an increase in hypolimnetic temperature of approximately 5°C (Mattson and Likens, 1990; Winter and Likens, 2009). Furthermore, a remarkable synchrony between CH_4 bubbling rates and small (1-3%) changes in air pressure was documented by Mattson and Likens (1990). The positive relationship between decreasing atmospheric pressure and CH_4 ebullition has also been reported from temporarily submersed peatlands (Fechner-Levy and Hemond, 1996) and boreal wetlands (Bubier et al., 1993). Our work supports these findings. The hypolimnetic temperatures were on average higher by 5°C during the rainy summer in Lake Jacques (Bartosiewicz et al., 2015), resulting from enhanced mixing associated to more frequent shifts between warm and cold fronts and rainfalls. During these mixing periods, warmer waters are moved from the warmer surface down to the hypolimnion, likely stimulating the production of CH_4 through enhanced methanogenesis (Zeikus and Winfrey, 1976; Nusslein et al., 2003) and triggering bubble release, either via increase energy input and/or changes in pressure. By comparison, during the heat wave, increased solar energy inputs to surface waters, decreased heat losses as a function of prevalent warm fronts, and efficient absorption in surface layers by high phytoplankton biomass lead to stronger stratification with reduced mixed layer thickness and lower hypolimnetic temperatures. This likely resulted in lower rates of methanogenesis in the sediments and lower rates of CH_4 ebullition.

During the heat wave year diffusive CH_4 was the most important source of GHG from the lake. Higher emissions via diffusive flux have been previously observed under strongly stratified conditions in a small shallow lake (Strayer and Tiedje, 1978). In contrast, the majority of studies have shown that ebullition is the main CH_4 emission pathway from lakes and reservoirs (Casper et al., 2000; Bastviken et al., 2004; Del Sontro et al., 2010; Schubert et al., 2012). In the case of Lake Jacques, *d*CH_4 fluxes represented almost 60% of total GWP in 2012, similar to the proportion emitted as bubbles in 2011. However, almost 70% of that diffusive flux was released during fall overturn. Indeed, accumulation of all three GHG in the hypolimnion during the summer heat wave lead to hot moments of fluxes during the fall, with concentrations of *d*CH_4 reaching some of the highest values ever reported in the surface waters of temperate lakes (e.g., Huttunen et al., 2003). This hot moment is consistent with other studies that have showed that the fraction of stored CH_4 lost via diffusion during the fall overturn period may be substantially larger than that lost during spring mixing and summer stratification (Encinas-Fernandez et al., 2014). Our results suggest that greater temporal variability in the emission of all GHGs will occur with warming climate, thus detailed assessments of seasonal variability in gas emissions from small lakes may be required to fully evaluate their GWP.

Strong thermal stratification during the summer heat wave resulted in persistent hypolimnetic anoxia in Lake Jacques, with little oxygen introduced below the thermocline throughout the entire summer. This was accompanied by a much higher phytoplankton production and oxygen supersaturation in surface waters. Overall, these extreme conditions lowered the GWP of the lake. Diffuse CH_4 was stored in the hypolimnion during the heat wave period, likely allowing a certain fraction to be oxidized to CO_2 by methanotrophs and possibly resulting in a greater contribution of CH_4-derived carbon to the lake food web through grazing (Bastviken et al., 2003).

Based on CH_4 profiles measured in the sediments of Lake Jacques during the heat wave (Fig. 2.2.S3) and on the one-dimensional mass balance equation of Boudreau (1997), we roughly estimated the sedimentary diffusive flux of CH_4 as varying between 2 and 3 mmol m^{-2} d^{-1}. Given the average surface diffusive flux (prior to the overturn period) of about 1 to 1.5 mmol m^{-2} d^{-1}, CH_4 oxidation in the water column would represent around 40-60% of the produced CH_4. These values appear relatively low in comparison to estimated 74% reported by Utsumi et al. (1998), 80% by Kankaala et al. (2006) or 75% by Schubert et al (2012). However, Lake Jacques is shallower than above studied lakes, with an oxygenated water column of only about one meter in 2012, potentially constraining aerobic methanotrophy. Additional CH_4 production in the hypolimnion within anoxic microzones (Oremland, 1979; Schulz et al., 2001) or in the oxic epilimnion (within phytoplankton agglomerates, Tang et al., 2014) would also raise the estimated amounts of CH_4 produced by the lake, and therefore increase the fraction consumed. Direct measurements of methanotrophy and the isotopic composition of plankton and GHG are needed to further understand the mechanisms responsible for the lower GWP observed during the heat wave.

There were stark differences in overall phytoplankton biomass production between years, where peak biomass levels were 3 times higher during the heat wave year. The variability in surface water CO_2 was partly explained by changes in the phytoplankton biomass where enhanced rates of photosynthesis can explain the negative relationship observed between ΔCO_2 and Chl-*a* (Fig. 2.2.S1). However the high biomass in 2012 resulted in greater variability in this relationship along with periods of CO_2 undersaturation. We also observed a positive relationship between ΔCH_4 and Chl-*a* during the heat-wave year (Fig. 2.2.S1) although the causality of that relation is not clear. It is possible that some of the CH_4 was produced in the water column though the consumption of phytoplankton-excreted acetate as discussed in Bogard et al. (2014). Other pathways of CH_4 production exist, such as syntrophic interactions between fermentative bacteria and acetoclastic methanogens (Stams and Plugge, 2009) or demethylation of organic matter released by cyanobacteria (Beversdorf et al., 2010), but the relevance of these pathways in nature is unknown. However, even if these processes are occurring in Lake Jacques, their contribution is likely to be secondary to the relative amount of CH_4 produced in the anoxic sediments (Bogard et al., 2014). Therefore, during the summer heat wave, the stimulating effect of high phytoplankton biomass on increased CH_4 was most likely due to increased deposition of organic matter to sediments and it subsequent decomposition (Schwarz et al., 2008; Fig. 2.2.S3).

Similar to other reports, we found that N_2O emissions did not contribute significantly to overall GWP of the lake (Huttunen et al., 2003; Hirota et al., 2007), however this gas did show remarkable variability between years, seasons and surprisingly emission pathways.

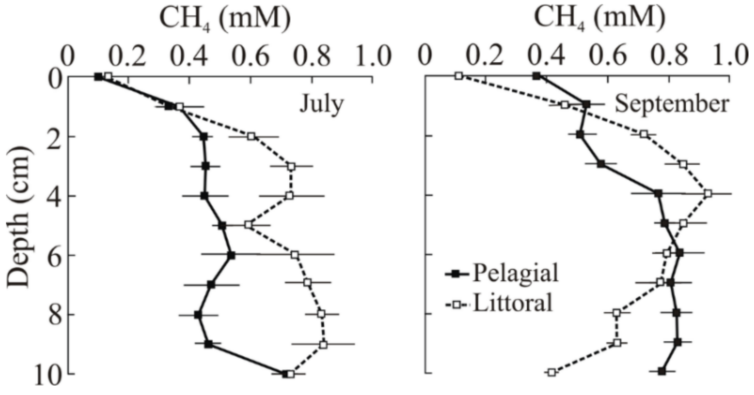

Figure 2.2.S3 Distribution of CH_4 concentrations (±SD) in the sediments of Lake Jacques at the beginning (DOY 189-202, July) and by the end of the summer heat wave in 2012 (DOY 234-254, September), by in situ dialysis using acrylic peepers.

In 2011, there was no obvious pattern in concentrations other than constant supersaturation, whereas during the heat wave year, the lake surface became periodically undersaturated during the peak phytoplankton bloom, and highly supersaturated at fall overturn. Although denitrification can produce N_2O, it can also consume it, particularly under conditions where NO_3^- is limiting and conditions are anoxic (Seitzinger et al., 1988). The great stability of the water column, low nitrate and anoxic hypolimnion in this shallow lake provided ideal conditions for N_2O consumption, helping to explain extended periods of undersaturation or near equilibrium during the heat wave. A similar pattern has previously been observed in a temperate reservoir (Beaulieu et al., 2014). Seasonal patterns in ΔN_2O reported in the literature seem to differ significantly among ecosystems where some studies have reported peaks in supersaturation during the summer (Cole and Caraco, 2001; Harrison and Matson, 2003), at fall overturn (Mengis et al., 1997), or even during the winter (Baulch et al., 2011). One study that followed the seasonal fluxes of N_2O in 12 streams over multiple years found no season pattern, but suggested that production varied more as a function of moments of point source N loading (Beaulieu et al. 2009). In our study, physical forcing seemed to be the main driver. Although the overall amount of N_2O emitted by diffusion was similar between the years, like the C gases, fall overturn was the hot moment for emissions during the heat wave year, whereas emissions were relatively constant during the non heat wave year. However, one unexpected result was the total amount of N_2O emitted by ebullition. In both years, this represented a significant proportion of total N_2O emissions (68 and 42% of the total flux in 2011 and 2012, respectively). Previous studies indicated that ebullition contributed little to N_2O flux in lakes (Sturm et al., 2014), while it was shown to be low (Clough et al., 2006) or negligible in rivers and streams (Baulch et al., 2011). To our knowledge this study is the first to show that N_2O emissions via ebullition can contribute such a significant amount of the total N_2O flux in a small lake.

Although there were distinctive stratification patterns between the rainy and dry summers, how these patterns played out between the littoral and pelagic zones of the lake also differed somewhat. The variability and concentration of nutrients and phytoplankton were typically greater in the littoral zone, with the exception of nitrate slightly more pronounced in the pelagic zone during the heat wave year. Temporal patterns in GHG concentrations, especially CH_4 and N_2O, were similar between zones, but again concentrations tended to be slightly higher in the shallower areas. In contrast, overall GHG ebullition was lower in the littoral than in the pelagial zone, specifically during the non heat wave year. It is possible that this lower rate of ebullition was a function of increased oxygenation to the root zone by macrophytes resulting in reduced CH_4 production (Brix, 1997; Maltais-Landry et al., 2009), or alternatively a result of degassing that occurs predominantly through the plant stems (Van Der Nat and Middelburg, 1998). Regardless, we did not find the littoral zone to be a consistent hot spot for *d*GHG emissions during the open water season, which is in contrast to other studies that report higher CH_4 and N_2O fluxes from shallow areas (Wang et al., 2006a; 2006b; Hofmann, 2013). In the case of Lake Jacques, the littoral zone was more strongly supersaturated in *d*GHG at the overturn period during the heat wave, and did emit nearly 2 times more as compared to the pelagic zone per unit area. Thus macrophyte effect on *d*GHG during the heat wave year only became apparent when the lake was mixing and plants started to senesce.

In conclusions, our study shows that increased variability in meteorological forcing under a warmer climate may lead to changes in GHG speciation and emission pathways in small lakes, and result in stronger temporal and spatial heterogeneity in their emissions. However, contrary to the generally accepted view, warmer climate may not necessarily result in higher GWP of small shallow lakes. If stratification is stronger and established more early in the season bottom waters remain colder throughout the summer likely slowing the microbial processing of organic matter and the subsequent production of GHGs. Direct estimations of GHG production and consumption rates are needed to decipher the combined effects of warming and stratification on GHG microbial cycling and storage in the hypolimnion. The stable conditions brought on by heat waves may reduce the amount of GHG produced and the GWP of a lake by a reduction in ebullition. Furthermore, stable conditions may favour methanotrophy. Accurate estimations of GWP from lakes used for future predictions require assessments of gas fluxes via all emission pathways, and need to account for interannual, seasonal and diurnal variability and hot moments of production. The interplay between temperature and water column stability will indeed influence total GHG emissions from shallow lakes, but current models fail to accurately predict not only pathways but overall emissions.

Acknowledgments

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2.3 Effects of planktonic food web structure and algal blooms on greenhouse gases in a hypereutrophic lake

Effet de la structure du réseau alimentaire planctonique et des fleurs d'eau d'algues sur les émissions de gaz à effet de serre par un lac hypereutrophe

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Les données présentées dans cet article nous permettent de comprendre les effets des fleurs d'eau d'algues sur les autres composantes de la chaine trophique planctonique et sur la saturation et l’accumulation des GES dans la colonne d’eau d’un lac hypereutrophe. Les données limnologiques ont été collectées et analysées par M. Bartosiewicz, les données de gaz ont été collectées et analysées par M. Bartosiewicz sous la supervision d’Isabelle Laurion. Les données météorologiques ont été collectées et analysées par M. Bartosiewicz. Le manuscrit a été écrit par M. Bartosiewicz et révisé par I. Laurion. Les 2 autres co-auteurs feront leur contribution dans les semaines à venir.

Résumé

1. Les efflorescences de phytoplancton sont fréquentes dans les lacs autour de la planète. Les fleurs d'eau peuvent mener à des plus hauts niveaux de saturation en CH_4 et en N_2O comparativement au CO_2. Le CH_4 et le N_2O sont des gaz à effet de serre respectivement 27 et 298 fois plus puissants que le CO_2. Le réchauffement global et l’eutrophisation stimulent tous deux les fleurs d'eau, et peuvent donc générer une rétroaction positive sur le climat en augmentant les proportions de CH_4 et N_2O émises par les lacs.

2. Les taux d’absorption de CO_2 par les efflorescences d’algues et la transformation de la matière organique en GES dépend de la biomasse planctonique et de la structure du réseau trophique. Le carbone fixé par le phytoplancton est transféré vers les sédiments soit directement par sédimentation, ou indirectement à travers le broutage du zooplancton. L’occurrence et la composition des espèces phytoplanctoniques peuvent affecter les taux de photosynthèse, de sédimentation du plancton, de broutage et de décomposition de la matière organique.

3. Pour évaluer le lien potentiel entre les fleurs d'eau, la composition du réseau trophique planctonique et la dynamique des GES, nous avons suivi le niveau de saturation du CO_2, CH_4 et N_2O dans un lac hypereutrophe subissant des fleurs d'eau récurrentes et principalement constituées de *Glenodinium* sp., *Anabaena* sp. ou *Asterionella* sp. Nous avons reliés la météorologie de surface, les caractéristiques limnologiques (oxygène, pH et nutriments) et les changements saisonniers de biomasse du picoplancton et du zooplancton aux niveaux de saturation des GES.

4. La surface du lac était soit super ou sous-saturée en CO_2 (jusqu’à 62.6 µM et aussi peu que -8.5 µM) et en N_2O (jusqu'à 15.2 nM et aussi peu que -0.8 nM) dépendamment des conditions météorologiques et de l’abondance du plancton. Le lac était super-saturé en CH_4 pendant toute la durée de l’étude (jusqu’à 2.7 µM). Les profils verticaux révèlent que le stockage de CO_2 dans la colonne d'eau était en moyenne 20% plus faible pendant l'été caniculaire présentant des fleurs d'eau de cyanobacteries, alors que le stockage de CH_4 et de N_2O était en moyenne 15% supérieur. Les résultats montrent une relation significative entre la concentration en GES à la surface et la vitesse du

vent, la température de l’air, le pH, l'oxygène et les nutriments. Ils indiquent aussi une corrélation avec la structure du réseau trophique planctonique. Les fleurs d'eau de dinophycés et de cyanobactéries étaient associées à la diminution du CO_2 et à l’augmentation du CH_4, alors que l’augmentation de la biomasse de bacterioplancton était accompagnée d’une augmentation du CH_4 et d'une diminution du N_2O.

5. La prolifération des fleurs d'eau d'algues sur la planète peut causer une libération accrue de CH_4 par les lacs au détriment des émissions de CO_2. L’occurrence des fleurs d'eau affectera le niveau de saturation des GES dans les lacs de façon directe, via leur consommation de CO_2, et indirecte, via leurs effets sur la disponibilité en oxygène et le fonctionnement du réseau trophique planctonique. La structure du réseau trophique aquatique affectée par le réchauffement du climat et l’eutrophisation peuvent donc contrôler le rôle des lacs dans le cycle global du carbone.

Summary

1. Phytoplankton blooms are a common feature of many lakes around the globe. Blooms may lead to higher CH_4 and N_2O saturation levels relative to CO_2 in lake water. Methane and N_2O are respectively 27 and 298 times more potent than CO_2 as heat-trapping gases. Thus, global warming and eutrophication, both of which promote phytoplankton blooms, may generate positive feedbacks on climate by raising the proportion of CH_4 and N_2O emitted from lakes.

2. Rates of CO_2 absorption by blooming algae and the transformation of organic matter into greenhouse gases (GHG) depend on plankton biomass and food web structure. The carbon fixed by phytoplankton is transferred to the sediments either directly through sedimentation or indirectly through zooplankton grazing. The occurrence and species composition of phytoplankton blooms can affect rates of photosynthesis, plankton settling, grazing and organic matter decomposition.

3. To evaluate the potential link between phytoplankton blooms, planktonic food web composition and GHG dynamics, we followed the saturation levels of CO_2, CH_4 and N_2O in a hyper-eutrophic lake suffering from recurrent blooms mainly consisting of *Glenodinium* sp., *Anabaena* sp. or *Asterionella* sp. We related the surface meteorology, limnological characteristics (oxygen, pH and nutrients) and seasonal changes in bacterioplankton and zooplankton biomass to GHG saturation levels.

4. The lake surface was either super- or under-saturated in CO_2 (as high as 62.6 µM and as low as -8.5 µM) and N_2O (as high as 15.2 nM and as low as -0.8 nM) depending on surface meteorology and plankton abundance. The lake was supersaturated in CH_4 during the entire study period (up to 2.7 µM). Vertical profiling revealed that CO_2 storage was on average 20% lower during the warmer summer presenting cyanobacterial blooms, while storage of CH_4 and N_2O was on average 15% higher. The results show significant relationships between surface GHG concentrations and wind speed, air temperature, pH, oxygen, and nutrients. They also indicate a correlation with phytoplankton community structure. Dinophyceae and Cyanobacteria blooms were related to decreasing CO_2 and increasing CH_4 levels, while increasing bacterioplankton biomass was accompanied by increasing CH_4 and decreasing N_2O levels.

5. Proliferation of phytoplankton blooms around the globe may result in higher rates of CH_4 releases from lakes at the expense of CO_2. The occurrence of algal blooms will affect GHG saturation levels in lakes, directly via their consumption of CO_2, and indirectly via their effects on oxygen stratification and food web functioning. The aquatic food web structure affected by warming and eutrophication will thus control the role of lakes in the global carbon cycle.

Key words: eutrophication, algal blooms, greenhouse gases, plankton, food web, global warming, cyanobacteria, undersaturation, autotrophy

2.3.1 Introduction

Many lakes which receive excessive nutrients suffer from noxious phytoplankton blooms (Schindler, 2006). These blooms affect the functioning of the entire ecosystem with cascading effects through the food web (Havens, 2008). Recently, Moss et al. (2011) pointed out a possible positive feedback mechanism between eutrophication and climate warming, as algal blooms developing in warmer, nutrient-rich lakes can lead to higher production and emissions of potent greenhouse gases such as CH_4 and N_2O. Additionally, Yvon-Durocher et al. (2011) reported that the warming of lakes may result in higher proportions of C released as CH_4 than CO_2 due to the stimulation of methanogens without concomitant increases in methanotrophs. Lakes have already been recognized as globally important sources of greenhouse gases (GHG) to the atmosphere (Tranvik et al., 2009) and reactors that transform and store allochtonous organic matter (Cole et al., 2007). Warmer and more eutrophic lakes may act as converters of atmospheric CO_2 and allochtonous organic matter into CH_4 and N_2O. Changes in ecosystem functioning and food web structure associated with increased occurrences of phytoplankton blooms may require special attention when considering the role of lakes in the global carbon budget.

Phytoplankton blooms directly affect CO_2 concentrations in the water column through photosynthesis and respiration (del Giorgio and Peters, 1994). In the early phase of blooms, intensive photosynthesis results in decreased CO_2 (Alin and Johnson, 2007) and increased alkalinity (Schindler et al., 1985). The species compositions of blooms may affect CO_2 uptake rates, and conversely CO_2 saturation levels may affect the composition of phytoplankton communities. For instance, Caraco and Miller (1997) calculated the CO_2 concentrations which result in the reduction of maximum growth by half (Beardall, 1985); approximately 1 μM for cyanobacterium as compared to 15 μM for other phytoplankton species. Stable and warm weather conditions likely favour bloom forming phytoplankton species and their high densities at lake surfaces may inhibit other phytoplankton through shading (Ibelings and Maberly, 1998). Many bloom forming phytoplankton species including cyanobacteria possess a carbon-concentrating mechanism that elevates the concentration of CO_2, around the photosynthetic carboxylation enzyme (Badger et al. 1994). Thus CO_2 can be severely depleted in the surface waters during algal blooms (Talling 1976; Maberly 1996). In contrast, net-phytoplankton may contribute up to 60% of the total community respiration (CO_2 production) in eutrophic lakes (del Giorgio and Peters 1993). An abundance of phytoplankton may also directly contribute to the aerobic production of CH_4 (Grossart et al., 2011) and indirectly stimulate anaerobic CH_4 production (Schwarz et al., 2008). Information on how phytoplankton species composition affects the rate of aerobic CH_4 production is lacking, though Shulz and Condrad (1995) demonstrated that anaerobic CH_4 production depends on the quality of phytoplankton material, and that green algae had higher rates of production than cyanobacteria. There is also evidence of direct N_2O production (Florez-Leiva et al., 2010; Fagerstone et al., 2011; Guieysse et al., 2013) or consumption (Farias et al., 2013) in phytoplankton blooms. The production of N_2O can be indirectly stimulated by phytoplankton blooms through the formation of hypoxic or suboxic zones in the water column (Mengis et al., 1997). Phytoplankton blooms may also indirectly affect GHG in lakes via their effects on other components of the planktonic food web, namely pico- and zooplankton.

Phytoplankton blooms may have a negative (Doucette, 1995) or stimulating (Brussaard et al., 1995) effect on picoplankton depending on the stage of the bloom. Toxic bloom exudates would have negative effects, while the provision of labile organic matter would positively stimulate picoplankton. Indirectly, blooms may stimulate picoplankton by negative effects on grazing zooplankton (Paerl and Pinckney 1996). Picoplankton are responsible for the production and consumption of GHG in the water column. For example, photosynthetic picoplankton absorb CO_2 and may contribute to aerobic CH_4 production through the release of substrates for methanogenesis (Karl et al., 2008; Grossart et al., 2011). Moreover, picoplanktonic methanotrophs facilitate decreases in CH_4 and increases in CO_2 concentrations in lakes. Bastiviken et al. (2003) showed that methanotrophic bacteria contributed from 0.5–10% of the total heterotrophic bacterioplankton biomass during summer stratification, while during winter and overturn periods, they contributed from 68 to 100%. Bacteria are also responsible for changes in N_2O through maintaining nitrification and denitrification processes in the water column of the lake (Mengis et al., 1997). This latter process may act both as a sink or source of

N_2O to the water column depending on the thermal and oxygen stratification (Beaulieu et al. 2014).

Phytoplankton blooms, particularly those containing cyanobacteria, are known to negatively affect zooplankton (DeMott et al., 2001; Ghadouani et al., 2003). Changes in zooplankton communities may directly influence GHG saturation levels, particularly CO_2 via individual respiration (Urabe et al., 2010), the production of CH_4 (De Angelis and Lee 1994) and possibly N_2O in their digestive tracts. Zooplankton can also indirectly influence GHG concentrations in lakes through several distinct mechanisms. Firstly, planktonic herbivores can transfer carbon and nutrients from deep to shallow water layers during vertical migrations (Steinberg et al., 2002; Haupt et al., 2010), which affects phytoplankton (CO_2) and bacterial production (CO_2, CH_4 and N_2O). Secondly, abundant herbivorous zooplankton may feed selectively on epilimnetic phytoplankton and sestonic organic carbon (Bloesch and Burgi, 1989; Sarnelle, 1999), leading to increased sedimentation rates (Reinertsen et al., 1990; Elser et al., 1995) or the emergence of inedible/toxic phytoplankton taxa (Benndorf et al., 2002; Scharf, 2008). Finally, zooplankton exert an effect on GHG through grazing on picoplankton, and specifically on methanotrophic bacteria (Kankaala et al., 2007).

The main source of organic matter for the production of GHG in the hypolimnion and lake sediments is material settling from the water column (Molongoski and Klug 1980). The seasonal changes of the planktonic food web structure and occurrence of algal blooms affects the sestonic composition and sedimentation processes (Rychła et al., 2012) potentially leading to differential cycling of organic matter in lakes and differences in GHG saturation (at the lake surface) and storage (in the entire lake volume). In the present study, we investigated the coupling of the biomass and structure of the planktonic food web with the GHG in an eutrophic lake. We tested if the occurrence of phytoplankton blooms had a cascading effect on GHG, and whether the strength of this effect varied in relation to the density and composition of the bloom. Specifically, we hypothesized that phytoplankton blooms is associated with an undersaturation of CO_2 at the lake surface and increased levels of CH_4 and N_2O. Considering the importance of allochthonous processes on carbon and nutrient cycling in shallow areas, we hypothesized that GHG would be more strongly coupled with plankton in the pelagial than in the littoral zone of the lake.

2.3.2 Methods

Study site

Lake St. Augustin (46 42’N, 71 22’W, Fig. 2.3.1) is a relatively small (0.62 km^2) urban lake located on the outskirts of Quebec City, along the north shore of the St. Lawrence River. The lake is supplied with water from two small surface inflows and an underground source, with a water renewal time of about 6 months (Roberge et al., 2002). Lake St. Augustin can be classified as eutrophic to hypereutrophic in terms of nutrients (TP = 20-160 $\mu g\ L^{-1}$) and Chl-*a* (summer values from 10 to 60 $\mu g\ L^{-1}$), and there is a relatively high amount of organic matter stored in the surface sediments (LOI = 18 ± 7%, n = 10). During the past two centuries, the lake has been greatly impacted by anthropogenic development and urbanisation (Pienitz et al., 2006). This resulted in a severe eutrophication by the mid twentieth century, and persistent summer algal blooms from 1960 onward. The blooms consist of an alternation between planktonic cyanobacteria, dinoflagellates and diatoms (Deshpande et al., 2014). Despite the abundant phytoplankton, the shallower zones (less than 2 m deep) of the lake are occupied by dense submerged macrophyte beds (e.g. *Myriophyllum spicatum*).

Physicochemistry and plankton

The lake was sampled during the open water season (3 May – 13 October) in 2011 and 2012 (22 April – 18 October). During both years hourly air temperature, relative humidity, wind speed and precipitation data were obtained from the meteorological station of Environment Canada located 1 km from the lake (anemometer threshold 1 $m\ s^{-1}$). Vertical profiles of temperature, conductivity, pH and dissolved oxygen (DO) were assessed fortnightly during the open water season with a 600R multiparametric probe (Yellow Spring Instruments). Two thermistor chains collected water temperatures from June to October 2012 from the littoral and pelagial areas of the lake. For the littoral sampling site, six temperature loggers (Onset Tidbit v2; accuracy 0.2°C, resolution 0.2°C, response time 5 min) were deployed at 0, 0.2, 0.4, 0.7, 1.0 and 1.3 m, and acquired data every 8 minutes. For the pelagic zone the loggers were deployed at 0, 0.2, 0.4, 0.8, 1.2 2.0, 2.5, 3.0, 3.5 and 4.0 m and acquired data every 8 minutes. Isotherms were calculated using linear interpolation.

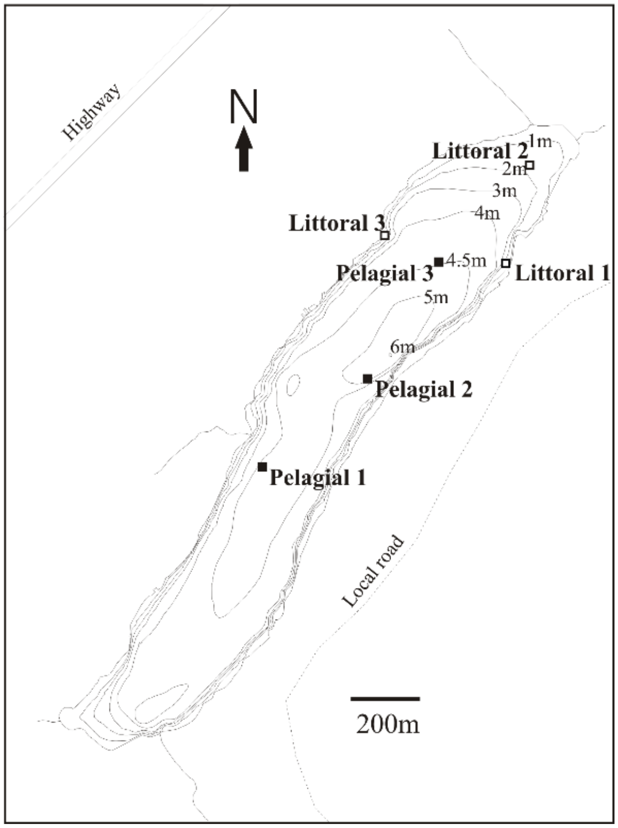

Figure 2.3.1 Location of the sampling stations on Lake St. Augustin. Gas samples were taken at sites 1-2 (2011) and 1-3 (2012) for both the littoral and pelagial zones of the lake.

On each sampling date, water samples collected at the surface of the littoral and pelagic sites (Fig. 2.3.1) were filtered through pre-rinsed cellulose acetate filters (0.2-µm pore size; Advantec Micro Filtration Systems, MFS) to measure soluble reactive phosphorus (SRP, persulfate digestion after Stainton et al., 1977) and dissolved nitrogen (NO^{3-}, alkaline persulfate digestion). Non-filtered water was acidified with H_2SO_4 (0.15% final concentration) and stored at 4°C until samples were analysed for total phosphorus (TP, spectrophotometry after persulfate digestion) and total nitrogen (TN, colorimetric method after potassium persulfate digestion in alkaline conditions as nitrate-nitrite by reduction with hydrazine). Additional surface water samples (100–500 ml) were filtered onto GF/F glass fibre filters (0.7 µm nominal mesh size; Advantec MFS Inc.) for the determination of Chl-a concentration using a UV–Vis spectrophotometer at 750 and 665 nm (Wintermans and De Mots, 1965).

Total bacterioplankton (both hetero- and phototrophic) enumeration was carried out on surface water samples (4 ml) fixed with glutaraldehyde (1% final concentration) and frozen at -80°C upon collection, using a FACScalibur flow cytometer (Becton Dickinson) equipped with an argon laser. Samples were processed at the lowest flow rate (12 µL min^{-1}), using 1 µm yellow-green microspheres (Polysciences) solution as an internal standard. Bead concentration was controlled using Truecount Absolute counting tubes (BD Biosciences). Samples were stained with the nucleic acid dye SYBR Green I (40 µL mL^{-1} final concentration) for 10 min in the dark. Stained cells were then discriminated on the basis of their green fluorescence at 530 nm and their 90° light scatter signal when excited at 488 nm. Data were analyzed using the CellQuest Pro software. The biomass was calcuated assuming spherical shape of a bacterial cell and an average diameter of 1.5 µm. Cells were assumed to have water density of 1 g cm^{-3}.

Picophytoplankton (picocyanobacteria and picoeukaryotes) concentrations were also analyzed on unstained surface water samples. Detection of the two groups was performed by the comparison of flow cytograms where cells were discriminated based on their side scatter signals (SSC) and both red (FL3) and orange fluorescence (FL2) as well as FL3 vs. FL2. The cytograms were analyzed using the Cell Quest Pro software, with manual gating to discriminate the different populations. For the picophytoplankton biomass estimates, the diameters of 10 cells of each group were measured under epifluorescence microscopy at 1000 × magnification, and were then converted to spherical biovolumes. The measured cell diameters (±SD) were 0.9 ± 0.1 µm for picocyanobacteria and 2.0 ± 0.6 µm for picoeukaryotes.

For the phytoplankton, lake water was collected at 0.25 m below the surface, transported to the laboratory in 1L dark polyethylene bottles, and fixed immediately with Lugol's iodine (5% final concentration). Samples were then transferred to sedimentation columns (Utermohl, 1958) for the enumeration of cyanobacteria and other phytoplankton cells with an inverted microscope (Zeiss Axiovert 200). For each phytoplankton species, at least 20 individual organisms were measured, and the biovolume was calculated following Hillebrand et al. (1999).

Zooplankton were collected with a conical plankton net (mesh size of 63 µm; mouth diameter of 30 cm) towed through the entire water column 3 or 4 times depending on the quantity of animals. Zooplankton were immediately preserved in 4% sugar-buffered formaldehyde (Prepas, 1978). Identifications, including rotifers, were carried out according to Wilson (1959), Pennak (1989) and Haney et al. (2013). The number of individuals counted per sample ranged from 150 to 600. For each species, at least 10 individual organisms per sample were measured, and biomass estimations were based on the standard operating procedure for zooplankton analysis (U.S. EPA 2003) developed from Gannon (1971), Stemberger (1979) and Evans et al. (1982).

Greenhouse gas saturation

The profiling of GHG (CO_2, CH_4 and N_2O) concentrations in the water column was performed at the littoral and pelagial sites of the lake (at 1m intervals) using the headspace method developed by Hesslein et al. (1991). Briefly, 2 L of lake water was equilibrated with 20 ml of ambient air for 3 min. The headspace volume was injected into vials (Exetainers, Labco) previously evacuated and flushed with helium. Gas samples were analyzed with a chromatograph equipped with an FID or ECD detector. Dissolved GHG concentrations at the lake surface were calculated according to Henry's law (details in Laurion et al., 2010). Global values of atmospheric partial pressures (IPCC 2007; 380 ppm of CO_2 and 1.8 ppm of CH_4) were used to determine the CO_2 and CH_4 saturation levels, while measured values of atmospheric partial pressures were used to determine N_2O. To assess the GHG storage (the amount of gas in the entire water column) in Lake St. Augustin, the mass of CO_2, CH_4 and N_2O was calculated by multiplying the average concentration over a depth interval by the water volume within that depth interval, and summing over depths of the lake (Rudd and Hamilton, 1978).

Greenhouse gas flux estimations

In 2011 and 2012, diffusive gas fluxes were calcuated as: $Flux_d = k\ (C_{sur} – C_{eq})$, where k is the gas transfer coefficient (cm h^{-1}), C_{sur} is the gas concentration in surface water (mmol L^{-1}), and C_{eq} is the gas concentration in equilibrium with the atmosphere. The gas transfer coefficient was taken from Cole and Caraco (1998) wind based model as $k_{600} = 2.07 + 2.15\ (u_{10})^{1.7}$. Ebulitive fluxes were assessed in 2012 using 6 inverted funnels located in the littoral (3) and pelagial zone (3) of the lake approximately 0.2 m below the surface of the water. The collected gas was sampled in triplicate vials (5.9 ml Exetainer), and analyzed by gas chromatography. The ebullition flux was calculated as: $Flux_e = C\ V_g\ V_m^{-1}\ A^{-1}$, where C is the concentration of a given gas in the syringe, V_g is the total volume of the gas in the syringe, V_m is the gas molar volume at ambient temperature obtained from the meteorological station, and A is the area of the funnel.

Statistical analyses

Prior to statistical analyses, environmental data were normalized using the naperian logarithm transformation. After normalization, GHG saturation levels were related to environmental variables and planktonic food web structure using Pearson's correlation analysis. The coupling between phytoplankton biomass (Chl-*a*) and GHG in the littoral and pelagial zones were compared using Fisher R-to-z transformation. Subsequently, only data for plankton components in the pelagic zone were used to investigate effects on GHG. Pearson's correlation was also used to analyze autocorrelations between environmental variables. Mutual relationships among variables were investigated by a principal component analysis. Stepwise multiple linear regression analysis was used to correlate saturation levels of the three GHG with the principal components (Camdevyren et al., 2005; Wang et al., 2006). The analyses were carried out using Sigma Plot 11 and Past 3.06 software.

2.3.3 Results

Meteorology and physicochemistry

The meteorological conditions differed between 2011 and 2012. The summer 2011 was cooler (average or AVG = 15°C) and windier (AVG = 3.6 m s^{-1}) than summer 2012 (16°C and 3.0 m s^{-1}). However, the most striking difference was for the daily rainfall (Fig. 2.3.2) that was 25% lower in 2012 than in 2011. During the study period, the maximum air temperature (31°C) was recorded during the warm front of mid-July 2011, and the coolest conditions (-1.2°C) occurred on the night of 6 October 2011. The daily rainfall (average for each month) did not fall below 3 mm in 2011 (June), with 12 rainstorm events (classified when total rainfall was > 20 mm, max. of 64.3 mm recorded on 28 August), and not below 2.4 mm in 2012 (July and August), with 9 rainstorm events.

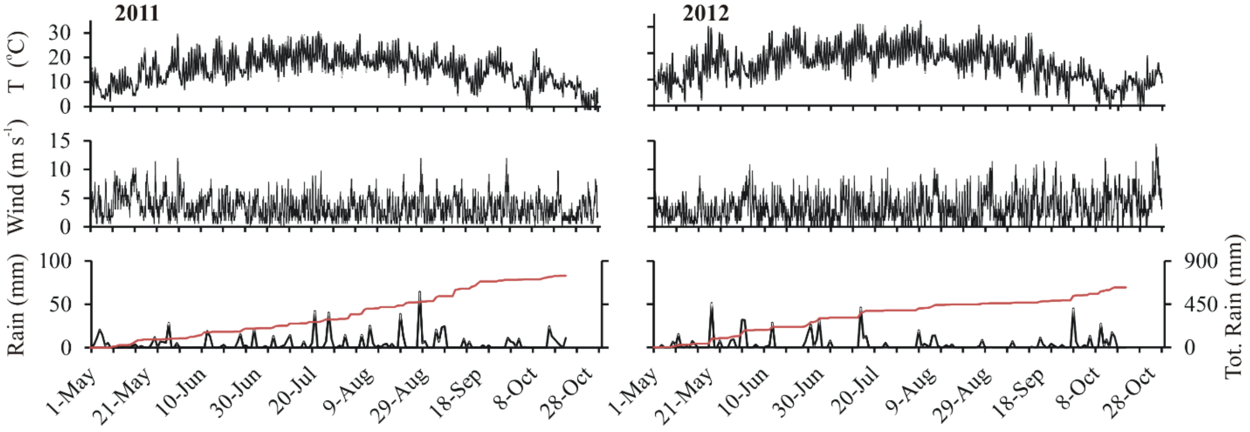

Figure 2.3.2 Meteorological conditions including air temperatures (T), wind speed at 10 m (Wind), daily rainfall (Rain), and cumulative rainfall (Tot. Rain) in 2011 and 2012.

These meteorological conditions resulted in differing limnological characteristics of Lake St. Augustin between years (Table 2.3.1). The average surface water temperature was lower in 2011 (17.9°C) than in 2012 (19.2°C, p=0.01, Student's t test) in the pelagial zone, as well as in the littoral zone of the lake (18.2 °C and 19.5 °C in 2011 and 2012, respectively). Surface waters were supersaturated with oxygen in both years (11.6 mg L^{-1} compared to 10.4 mg L^{-1} in 2011) while bottom waters became suboxic in the summer, and more so in 2012 (3.2 mg L^{-1} compared to 4.5 mg L^{-1}). Lake St. Augustin is polymictic, with short (up to 5 days) periods of thermal stratification (Fig. 2.3.3). The temperature difference between surface and bottom waters in the pelagic zone reached a maximum of 6°C, but was only around 4°C for most of the open water period in 2012. Winds from the NE and SW above 9 m s^{-1} lead to a complete mixing of the water column, as observed multiple times in 2012. The water column in the littoral zone was virtually unstratified for most of the open water period, with temperature differences between surface and bottom waters rarely exceeding 1°C. The lake had higher total nutrient levels in the warmer year (69 µg P L^{-1} and 0.44 mg N L^{-1}, compared to 38 µg P L^{-1} and 0.35 mg N L^{-1} in 2011). In contrast, concentrations of soluble nutrients were on average higher in the cooler year (4.5 µg SRP L^{-1} and 0.15 mg NO_3 L^{-1}, compared to 4.0 µg SRP L^{-1} and 0.12 mg NO_3 L^{-1} in 2012).

Variables	Littoral		Pelagial	
	2011	2012	2011	2012
Temp (°C)	8.0-25.5	6.1-27.4	7.5-24.3	6.4-28.2
pH	7.9-8.8	7.8-8.7	8.0-8.9	8.0-8.9
O_2 surf (mg L^{-1})	9.6-12.6	9.0-14.2	8.6-12.6	9.5-14.6
O_2 bott (mg L^{-1})	7.0-9.6	0.9-10.8	2.1-8.0	0.5-9.8
TP (µg L^{-1})	5-96	27-170	16-72	19-166
TN (mg L^{-1})	0.12-0.64	0.32-1.11	0.25-0.45	0.21-0.80
SRP (µg L^{-1})	*bd*-9.0	*bd*-12.5	0.5-14.6	*bd*-17.2
NO_3^- (mg L^{-1})	0.06-0.51	0.01-0.98	0.05-0.25	0.04-0.34
Chl-*a* (µg L^{-1})	4.9-55.1	6.3-50.2	4.8-45.2	4.3-45.8

Table 2.3.1 Limnological characteristics in Lake St. Augustin during the studied period (May-October 2011 and 2012) including range of surface water temperature (Temp), pH, average surface and bottom oxygen concentration, nutrients (TP, TN, SRP, NO^{3-}), and chlorophyll-*a* (Chl-*a*). *bd* = below detection limit.

The littoral zone was richer in total nutrients (TP, TN) and poorer in soluble nutrients (SRP) in both years. Chl-a levels were higher in the warmer year (on average 20 µg L^{-1}, compared to 14 µg L^{-1} in 2011) and overall higher in the littoral zones (Table 2.3.1).

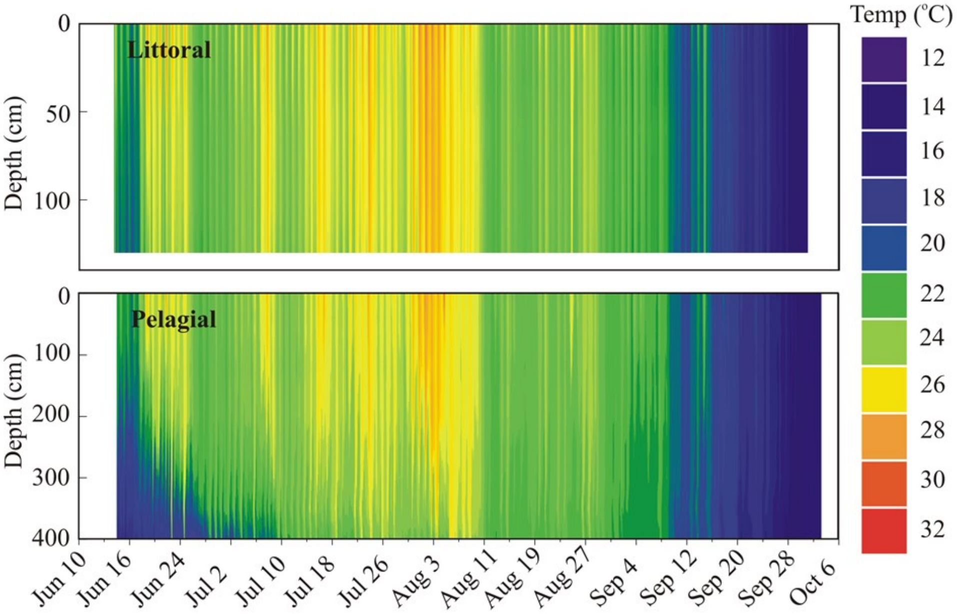

Figure 2.3.3 Thermal structure of Lake St. Augustin in the littoral (upper) and pelagic zone (lower panel) during 2012.

The pH increased during the summers of both years as a consequence of growing phytoplankton, reaching 8.9 early in autumn 2011 or by the end of summer 2012, and was on average higher in the warmer year (8.5, compared to 8.3 in 2011).

Plankton abundance and food web structure

The plankton of Lake St. Augustin showed dynamic seasonal changes, alternating between phytoplankton blooms dominated by different taxa, rises in bacterioplankton biomass, and phases with abundant zooplankton (Fig. 2.3.4).

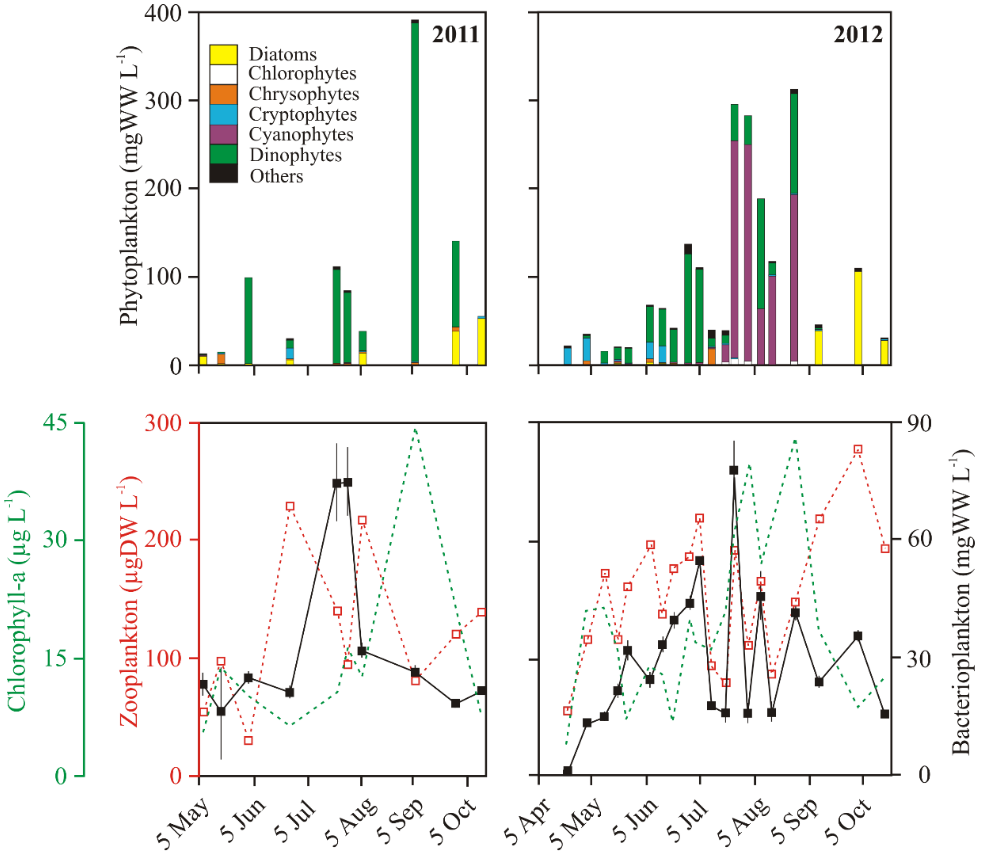

Figure 2.3.4 Seasonal changes in the biomass of planktonic food web components in the pelagial zone of Lake St. Augustin, including phytoplankton (7 main taxonomic groups), total bacterioplankton, and zooplankton (sum of cladocerans, copepods and rotifers) in 2011 and 2012. Picocyanobacteria and picoeukaryotes did not contribute significantly to photoautrotrophic biomass.

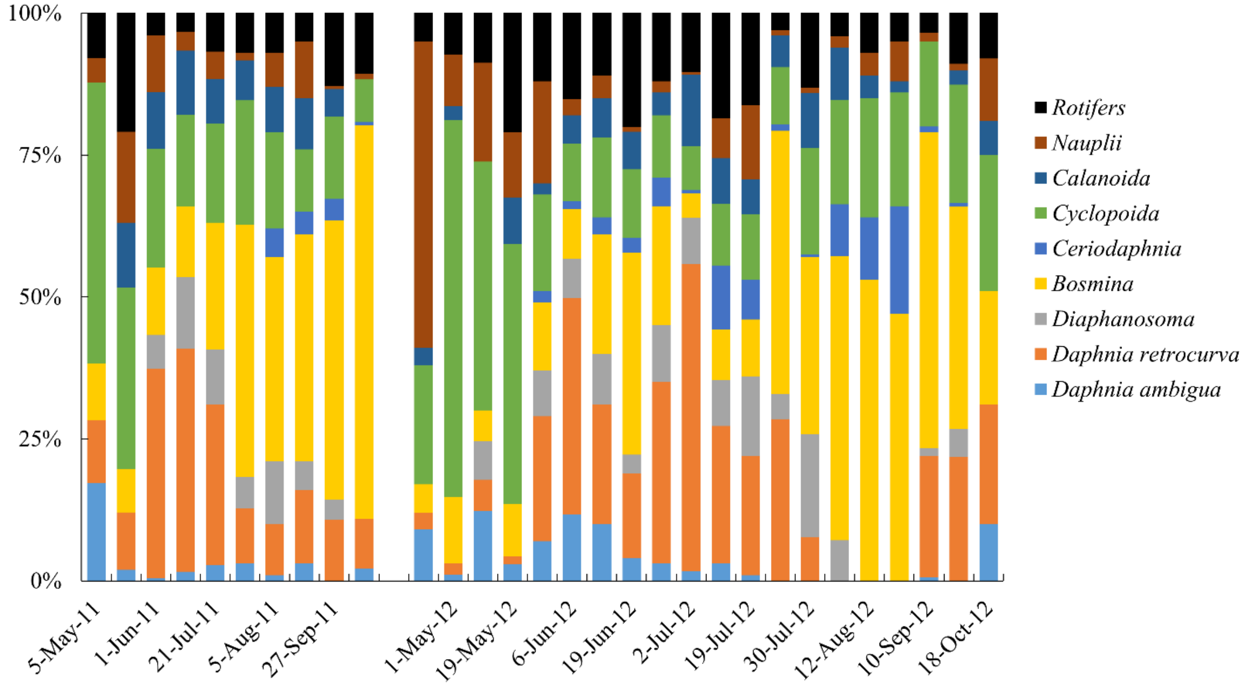

Figure 2.3.S1. Zooplankton community composition in Lake St. Augustin from May to October 2011 and from April to October 2012.

The algal blooms consisted of either Dinophyes (*Glenodinium* sp.), Cyanobacteria (*Anabaena* sp., *Aphanizomenon* sp.) or Diatoms (*Asterionella* sp.). In 2011, Dinophyceae dominated the phytoplankton community starting from June when the biomass reached 96 mg wet weight L^{-1} (mg WW L^{-1}), equivalent to 15 µg Chl-*a* L^{-1}. This increase in phytoplankton biomass was followed by an increase in zooplankton (mainly small cladocerans). By the end of June, the phytoplankton biomass decreased to 30 mg WW L^{-1} and consisted of a mixture of species from all main taxonomic groups. Later that year, the dominance of Dinophytes increased again, with a culmination in September when biomass reached the extremely high value of 395 mg WW L^{-1} during a massive algal bloom (47 µg Chl-*a* L^{-1}). Later in September and October, Diatoms became increasingly abundant, with phytoplankton biomass reaching up to 105 mg WW L^{-1} (22 µg Chl-*a* L^{-1}). In 2011, the bacterioplankton biomass varied from 16 to 75 mg WW L^{-1}, with maximum values following heavy rainfalls, and apparently synchronized with the decrease in zooplankton biomass by the end of July.

In 2012, the phytoplankton was dominated by Cryptophytes and Diatoms early in the season (May). Dinophyceae increased later in May and June, with phytoplankton biomass reaching a maximum of 125 mg WW L^{-1}, accompanied by an increase in zooplankton biomass (mainly Copepods). Later in the summer, the phytoplankton were dominated by Cyanobacteria, with biomass reaching 250 mg WW L^{-1}. During the same period there was a striking decrease in zooplankton biomass and changes in community structure (shift toward small bodied cladocerans e.g., *Bosmina* sp. and Rotifers, Fig. 2.3.S1).

Again, a bloom of Bacillariophyceae developed in the fall (August and September). The bacterioplankton biomass varied from 2 to 76 mg WW L^{-1} in 2012, with the maximum value reached in early July, along with a bloom of Cyanobacteria. The phytoplankton biomass was correlated with environmental and limnological conditions (Temp, O_2, pH, TP; $R > 0.5$, $p < 0.001$), and with bacterioplankton biomass ($R = 0.35$, $p = 0.01$). The relationship with zooplankton was not significant, suggesting a possible decoupling of primary production from grazing during the bloom. The bacterioplankton biomass was correlated with the temperature ($R = 0.63$; $p = 0.01$) and the O_2 concentration at the bottom of the lake ($R = -0.60$; $p = 0.01$).

Greenhouse gases saturation and storage

The saturation levels of GHG in surface waters of Lake St. Augustin varied both seasonally and interannually (Fig. 2.3.5). There were also differences in GHG saturation levels between the littoral and pelagial zones of the lake. On average surface waters were less saturated in CO_2 in the warmer year (departure from saturation 1.5, compared to 6.6 μM in 2011), and this difference was even stronger in the littoral zone (respectively 1.1 and 6.8 μM, $p = 0.001$). In 2011, the lake became highly supersaturated in CO_2 (up to 28 μM in July) and was undersaturated only for a short period in September (-8.5 μM). In contrast, the lake surface was undersaturated in CO_2 for most of the summer of 2012 (down to -5.3 μM), and became supersaturated only during the fall phytoplankton bloom (up to 35 μM). The temporal pattern in CO_2 did not differ between zones. Surface waters were more saturated in CH_4 during the warmer year (0.36 compared to 0.27 μM in 2011, $p = 0.01$), a difference mainly due to the high levels of CH_4 by the end of summer. Methane saturation levels varied greatly between zones and more so in 2012 (CV = 51% and 117% respectively for the littoral and pelagic zones, compared to 45% and 66% in 2011), when the littoral zone had the highest concentrations (2.7 μM) observed for the entire study period (Fig. 2.3.5). Surface waters were significantly less saturated in N_2O during the warmer year (0.8, compared to 1.2 nM in 2011, $p < 0.001$), but this difference was largely due to exceptionally high supersaturation levels in the littoral zone. The patterns in N_2O differed between years, the lake more often acting as a sink in the warmer year (down to -0.6 nM). Peak supersaturation of N_2O occurred by the end of September 2011 (15.2 nM) and by the end of April 2012 (6.5 nM).

Greenhouse fluxes at the lake surface

The CO_2 diffusive fluxes estimated with wind based model varied from -10.7 to 19 mmol m^{-2} d^{-1} in the pelagial zone, and from -12.3 to 32.2 $mmol^{-2}$ d^{-1} in the littoral zone of the lake in 2011. The CO_2 fluxes were lower on average in 2012 (p=0.01) and varied from -7.6 to 62.6 $mmol^{-2}$ d^{-1} in the pelagial zone, and from -10.2 to 45.0 $mmol^{-2}$ d^{-1} in the littoral zone. The CH_4 fluxes in 2011 varied from 0.01 to 0.88 $mmol^{-2}$ d^{-1} in the pelagial zone, and from 0.03 to 1.13 $mmol^{-2}$ d^{-1} in the littoral zone of the lake; fluxes were higher in 2012 (p=0.01) varying from 0.12 to 1.14 $mmol^{-2}$ d^{-1} and from 0.11 to 4.18 $mmol^{-2}$ d^{-1}, respectively in both zones. The N_2O fluxes in 2011 varied from -0.15 to 5.19 μmol m^{-2} d^{-1} in the pelagial zone, and from -1.53 to 17.34 μmol m^{-2} d^{-1} in the littoral zone; fluxes were lower in 2012 (p=0.01) varying from -1.31 to 2.25 μmol m^{-2} d^{-1} and from -0.66 to 3.97 μmol m^{-2} d^{-1} respectively in both zones. The bubbles collected in the pelagial zone of the lake (2012) contained from 2 to 17% of CH_4 and those in the littoral zone from 16 to 45%. Ebullitive flux of CH_4 varied from 0.01 (early July) to 0.11 mmol m^{-2} d^{-1} (late August) in the pelagial zone, and from 0.1 to 0.88 mmol m^{-2} d^{-1} in the littoral zone. Ebullition contributed respectively up to 18% and up to 53% of the total CH_4 efflux from the pelagial and littoral zone, respectively. Concentrations of CO_2 and N_2O in bubbles were negligible, thus ebullition did not contribute significantly to the efflux of these gases from Lake St. Augustin.

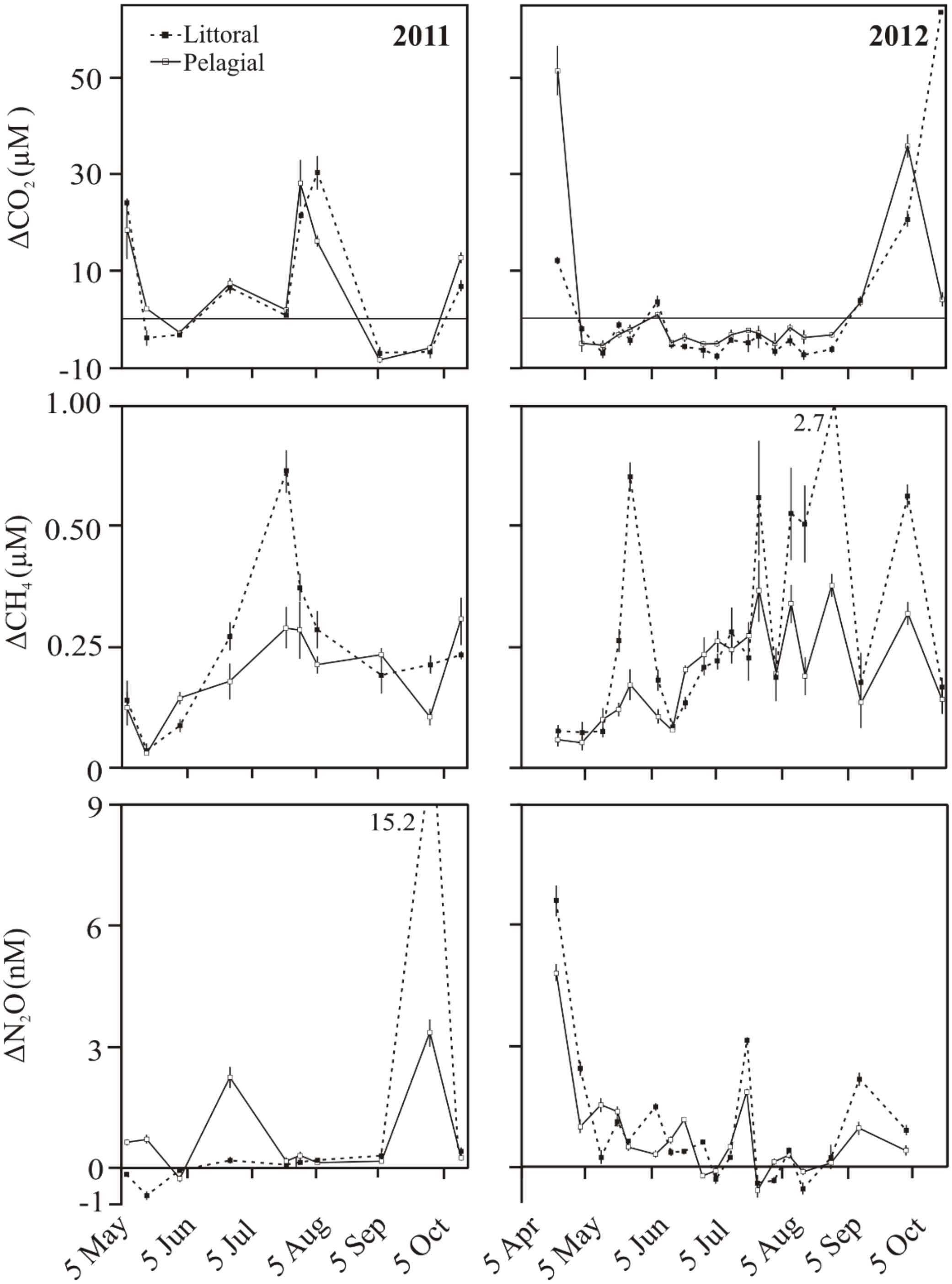

Figure 2.3.5 Saturation levels of CO_2, CH_4 and N_2O in the littoral (dotted) and pelagial zone (solid lines) of Lake St. Augustin in 2011 (left) and 2012 (right panels).

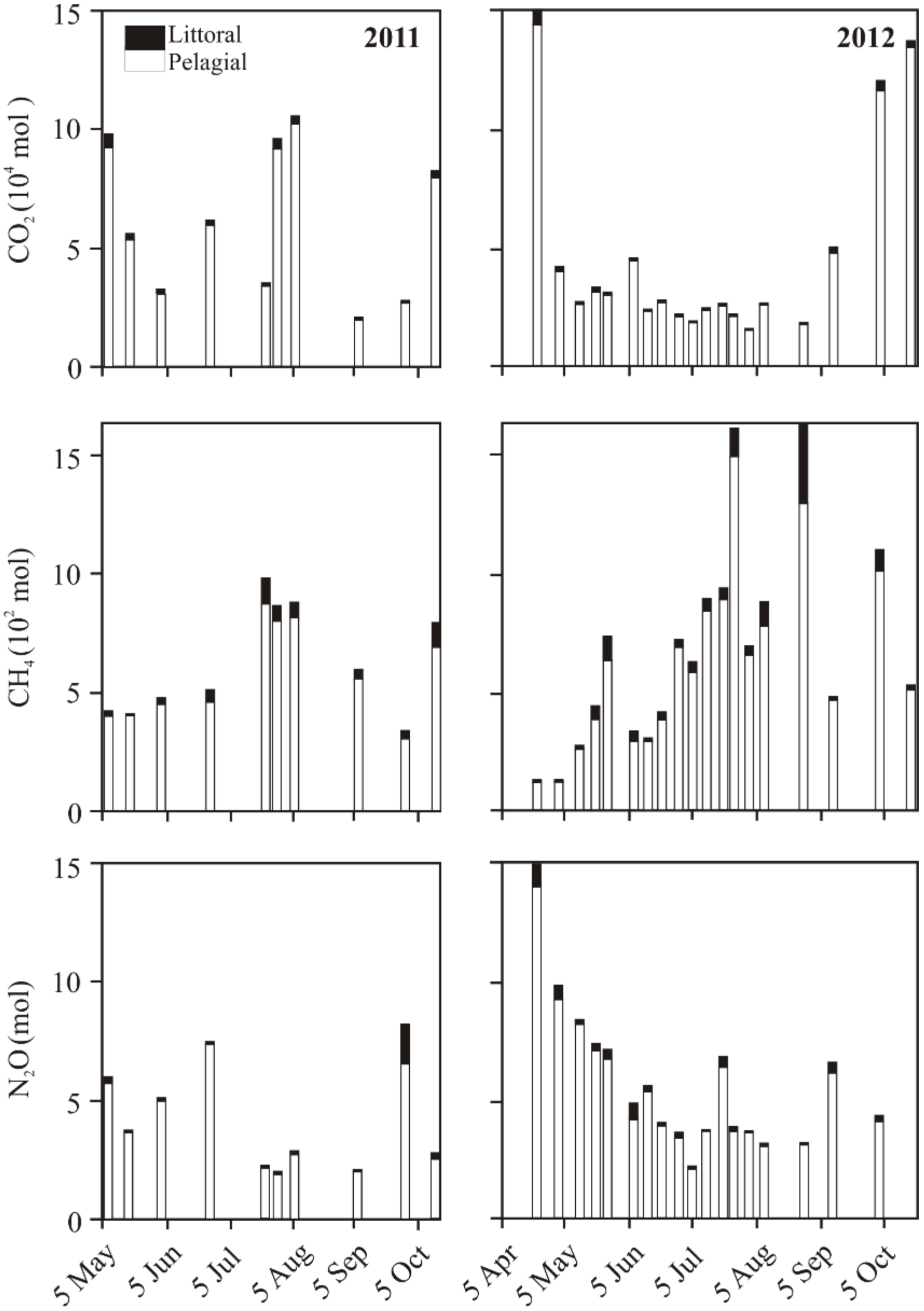

Figure 2.3.6 Water column storage of GHG (CO_2, CH_4 and N_2O) for the entire littoral and pelagial zones of Lake St. Augustin in 2011 and 2012.

Even though the lake is polymictic, it stored large quantities of GHG both in 2011 and 2012, with periodical accumulation in bottom waters (Fig. 2.3.S2).

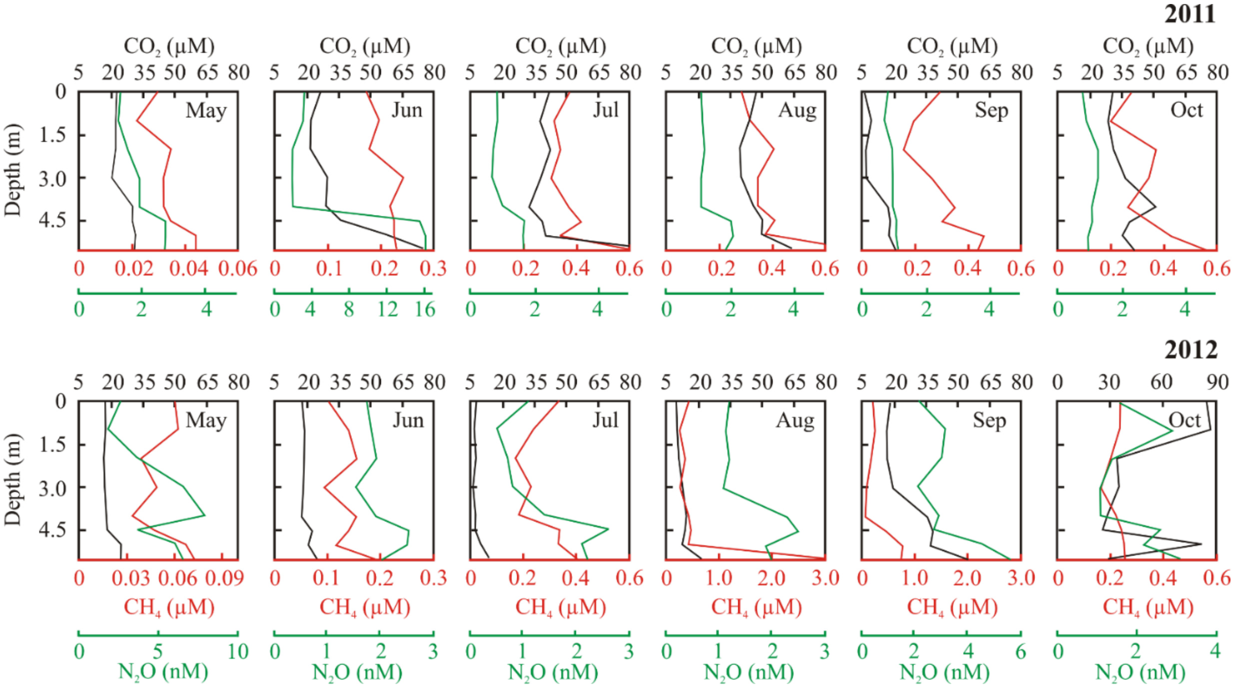

Figure 2.3.S2 Changes in the vertical distribution of GHG (CO_2, CH_4 and N_2O) in the pelagial of Lake St. Augustin.

The seasonal and interannual changes in GHG storage followed patterns of gas saturation in the surface waters for CO_2 and CH_4 but not for N_2O (Fig. 2.3.6). The lake stored less CO_2 (on average 45 × 10^3 moles, compared to 62 × 10^3 moles in 2011, $p = 0.02$) and more CH_4 (0.62 × 10^3 moles, compared to 0.56 × 10^3 moles in 2011, $p = 0.01$) during the warmer summer. Unlike observations for surface waters, the entire water column of the lake (storage) contained more N_2O during the warmer year (5.9 moles, compared to 4.2 moles in 2011, $p = 0.01$; Fig. 2.3.6). The littoral zone, representing less than 10% of the total lake surface, stored about 15% of the total GHG. The amount of CO_2 stored in lake waters decreased in both years during the algal blooms. The maximum CH_4 storage was recorded in August 2012 (1.66 × 10^3 moles) during the cyanobacteria bloom. Interestingly, the maximum N_2O storage was observed just after the spring melt in April 2012 (17.5 moles), and was not in association with the phytoplankton bloom.

Effects of environment characteristics and food web structure on greenhouse gases

GHG saturation levels were correlated with meteorological and limnological characteristics of Lake St. Augustin (Table 2.3.S1) such as wind speed (averaged over previous day), surface water temperature, pH, O_2, nutrients and Chl-*a*. Correlations between environmental factors and GHG were stronger (and more consistent between gases) in the pelagic zone of the lake.

Variables	Littoral			Pelagial		
	CO_2	CH_4	N_2O	CO_2	CH_4	N_2O
	(µM)		(nM)	(µM)		(nM)
Temp (°C)	**-0.495**	**0.522**	-0.175	**-0.496**	**0.681**	**-0.436**
Rain (mm)	0.104	0.146	-0.131	0.172	0.098	-0.124
Wind (m s^{-1})	**-0.458**	**0.426**	0.079	**0.556**	**-0.381**	0.261
pH	**-0.379**	**0.386**	-0.096	**-0.704**	0.274	-0.276
O_2 Surf (mg L^{-1})	**-0.394**	0.293	-0.264	**-0.419**	0.160	-0.052
O_2 Bot (mg L^{-1})	0.188	**-0.678**	0.123	**0.448**	**-0.776**	**0.421**
TP (µg L^{-1})	-0.079	**0.380**	0.282	-0.187	0.234	-0.240
TN (mg L^{-1})	-0.141	0.308	-0.032	0.059	-0.169	0.005
SRP (µg L^{-1})	0.253	0.316	-0.028	**0.447**	0.210	0.065
NO_3^-(mg L^{-1})	0.309	-0.226	**0.418**	0.097	**-0.424**	**0.467**
Chl-*a* (µg L^{-1})	**-0.453**	**0.306**	0.068	**-0.712**	**0.488**	**-0.351**

Table 2.3.S1 Result of Pearson's correlation analysis between meteorological and limnological characteristics, including surface water temperature (Temp), rainfall (averaged over the 3 days prior to sampling), wind speed (averaged over one day prior to sampling), pH, surface and bottom (10 cm above the sediments) oxygen concentrations, nutrients (TP, TN, SRP, NO^{3-}), chlorophyll-*a* (Chl-*a*), and greenhouse gas concentrations (CO_2, CH_4 and N_2O) given as the level above saturation in the littoral and pelagial zones of Lake St. Augustin.

The correlation analysis among gases revealed a negative relationship between CH_4 and N_2O ($R = -0.48$, $p = 0.01$). Noticeably, all three GHG were correlated with Chl-*a* (except N_2O in the littoral zone). The relationship between CO_2 and Chl-*a* was stronger in the pelagial zone of the lake (Fisher R to Z transformation, $p = 0.02$).

The principal component analysis (PCA) on the pelagic data set resulted in three components with eigenvalues above 1, explaining 71% of the total variance (Table 2.3.2, Fig. 2.3.S3).

Variables	Littoral				Pelagial		
	PC 1	PC 2	PC 3	PC 4	PC 1	PC 2	PC 3
Temp	**0.737**	**0.491**	-0.049	-0.215	**0.465**	-0.300	-0.204
Rain	-0.154	0.196	**0.740**	-0.006	-0.070	-0.316	**0.739**
Wind	**-0.710**	-0.036	0.380	-0.120	-0.372	-0.014	0.183
Chl-*a*	**0.760**	-0.058	0.180	**0.483**	**0.492**	0.140	0.125
pH	**0.638**	0.425	-0.048	0.165	0.364	0.099	-0.093
O_2 surf	**0.765**	-0.079	-0.271	-0.034	0.313	0.228	-0.191
O_2 bott	-0.387	0.295	-0.308	**0.646**	**-0.496**	0.212	-0.322
TP	0.340	**-0.533**	**0.493**	**0.474**	0.269	**0.484**	0.420
TN	0.368	**-0.691**	-0.125	-0.238	0.0437	**0.548**	0.187
Bact	**0.642**	0.334	0.382	-0.176	0.281	**-0.483**	-0.030

Table 2.3.2 Results of Principal Component Analysis (PCA) for the meteorological conditions and limnological variables in the littoral and pelagial zones of Lake St.Augustin. All variables were normalized with naperian logarithm. Bact = Bacterioplankton abundance.

Stepwise multiple linear regression analysis revealed CO_2 saturation was best explained ($R^2 = 0.51$) by variations in surface temperature, wind speed and Chl-*a* (PC1). Methane saturation was best explained ($R^2 = 0.60$) by variations in surface temperature, wind speed and Chl-a (PC1), as well as by bacterioplankton and nutrients (PC2). Nitrous oxide saturation was best explained ($R^2 = 0.20$, $p = 0.01$) by variations in surface temperature and O_2 concentrations (PC1). In the littoral zone, the PCA resulted in 4 components with eigenvalues above 1, explaining 75% of the total variance. Again, CO_2 was best explained by PC1 ($R^2 = 0.25$, $p = 0.01$) related to surface temperature, wind speed and Chl-*a*. CH_4 was best explained ($R^2 = 0.75$) by variations in wind speed, Chl-*a*, rainfall and bottom O_2 concentrations (PC1 and PC3). N_2O was only related to PC4 ($R^2 = 0.15$, $p = 0.03$) representing variability in O_2 concentrations and TN.

Given the more consistent relationships between environmental conditions (including Chl-a) and GHG in the pelagic zone, only this data set was used to explore the relationships between components of planktonic food web and GHG saturation levels using correlation analyses (Fig. 2.3.7). There were positive effects of Diatoms and zooplankton biomass on CO_2, as well as negative effects of Dinophyceae and picocyanobacterial biomass. For CH_4, there were positive effects of bloom-forming Chlorophytes, Cyanobacteria and Dinophytes, and a negative effect of bacterioplankton biomass. Methane and zooplankton biomass were not significantly related. Finally, there was a positive effect of the Cryptophytes on N_2O, and a negative effect of bloom-forming cyanobacteria and bacterioplankton.

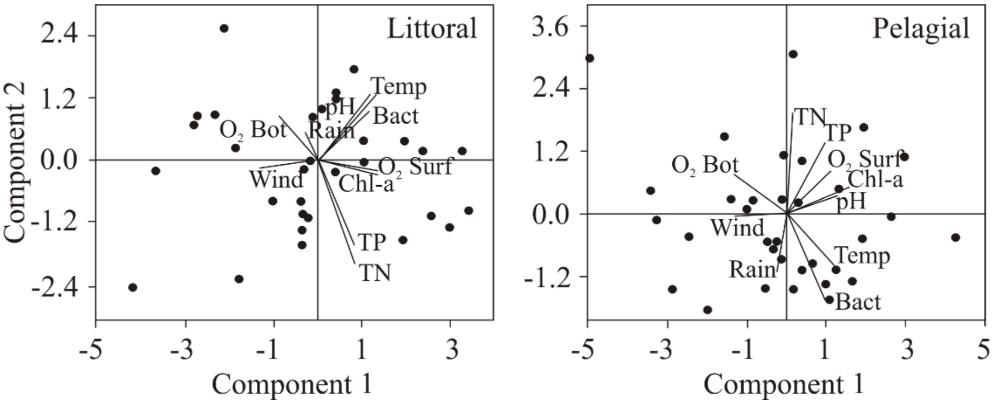

Figure 2.3.S3 The results of the principal component analysis (PCA) for environmental characteristics of the water in the littoral and pelagial zones of Lake St. Augustin.

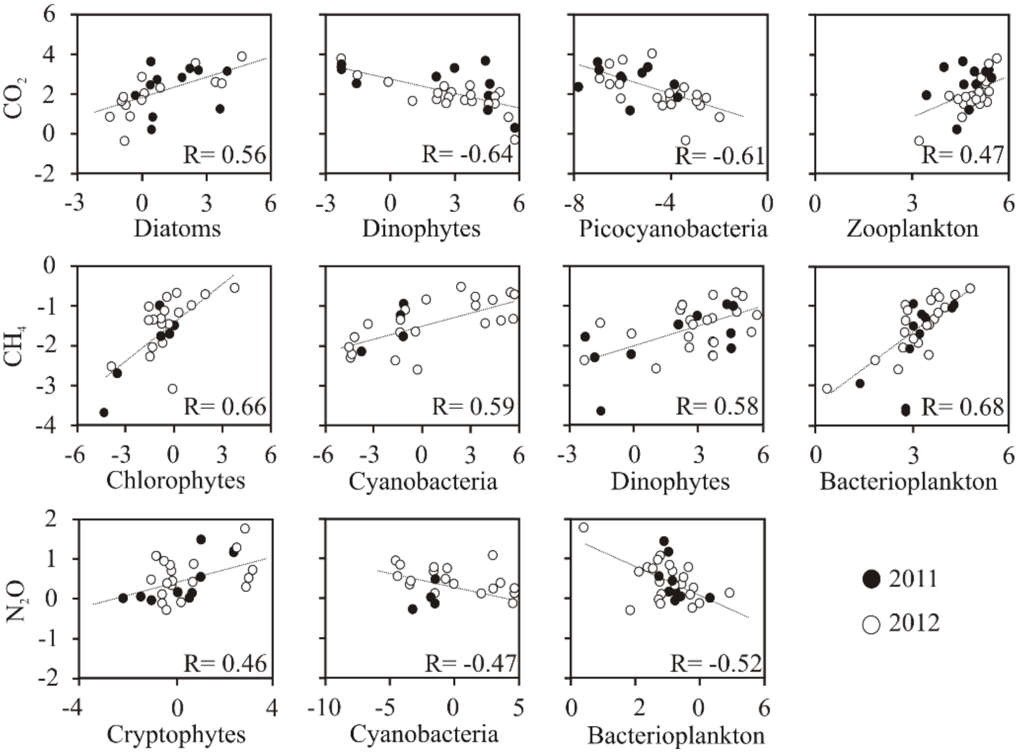

Figure 2.3.7 Relationships between components of the planktonic food web and GHG saturation levels in surface waters of Lake St. Augustin. All variables were normalized by naperian logarithm.

2.3.4 Discussion

Lake St. Augustin is a productive, eutrophic lake where phytoplankton biomass (expressed as Chl-a) is high throught the entire open water season (~20 µg L^{-1}). The blooms, when phytoplankton concentrations are 2-3 times higher than average summer values, occur either mid-summer or early in autumn. The food web approach taken in this study allowed us to investigate effects of phytoplankton bloom on other components of the food web, and examine the coupling between planktonic biomass and dissolved GHG.. In addition to the significant effects of blooming phytoplankton on GHG saturation levels at the lake surface, our results support earlier studies showing relationships between GHG and meteorological conditions as well as some limnological characteristics (Liikanen et al. 2002; Bartosiewicz et al. 2015). The potential positive feedback mechanism between recurrent phytoplankton blooms and higher GHG emissions from lakes depends on the interplay between physical and biological components of lake ecosystems. Phytoplankton bloom density and composition is partly controlled by meteorological and limnological conditions (wind speed, inputs of nutrients), thus they may not only affect food web structure but also the strength of the interaction between plankton and GHG in the lake. In a warmer and drier climate, eutrophic and hypereutrophic lakes suffering from noxious phytoplankton blooms may absorb more atmospheric CO_2 and release back more CH_4, which has a global warming potential 27 times greater than CO_2.

The surface of the lake was periodically or persistently undersaturated in CO_2 during algal bloom in both 2011 and 2012. The majority of lakes around the globe are presented as significant sources of CO_2 (Cole et al. 2007). For instance, Cole et al. (1994) reported CO_2 supersaturation in 87% of the 1835 studied lakes with CO_2 concentrations ranging from 1087 µatm in average for North American lakes, 1469 µatm for boreal lakes, 1520 µatm for South American lakes, and 2269 µatm for African lakes. However, for 88% of the studied lakes, the data were collected during the overturn period. Sobek et al. (2005, 4902 lakes) using mostly data from Cole et al. 1994 (1676 lakes) and Swedish National Lake Monitoring (2851 lakes sampled in 2000) also concluded that majority of global lakes is supersaturated in CO_2. Noticeably, the sampling for the Swedish Lake Monitoring program in 2000 took place between September 2000 and January 2001. This episodic sampling may have precluded observation of CO_2 undersaturation during peak of phytoplankton biomass, which at least for dimictic lakes generally occurs in summer months. Recurrent CO_2 supersaturation in lakes reported by Cole et al. (1994) and Sobek et al. (2005) may have also resulted from increased deposition, and subsequent respiration, of terrestrial carbon arriving to with runoff that is usually higher in rainy seasons (Vachon and del Giorgio, 2014). Using these data to upscale estimates on the global importance of lakes to the carbon cycle and climate might be biased if the datasets were skewed toward a specific season, time of the day, mixing regime or trophic status.

Lazzarino et al. (2009) observed that majority of meso- to eutrophic lakes in Florida (88%) were supersaturated in CO_2 during both rainy and dry season. In this case however, the average Chl-*a* concentration for all studied lakes remained below 20 µg L^{-1}. Further, the majority of these lakes received large inflows of allochtonous carbon through the entire season, masking the CO_2 uptake by phytoplankton. In Lake St. Augustin, surface water CO_2 concentrations fell below saturation during the phytoplankton blooms of both years. This undersaturation persisted also during 24h sampling with only slight increase in surface CO_2 levels at night (data not shown). These results stand in line with recent findings by Balmer and Downing (2011) who reported CO_2 undersaturation during the summer in 60% of the studied productive lakes. We postulate that small eutrophic lakes suffering from phytoplankton blooms may often represent a sink of CO_2, changing the alkalinity buffering reactions during daylight hours. There are however remarkably few studies addressing the global importance of carbon uptake by phytoplankton blooms and its subsequent release as CH_4 (e.g., Heathcote and Downing 2012).

The model by Den Heyer and Kalff (1998) showed that the contribution from sediment respiration to ecosystem CO_2 production is higher for lakes with low primary production. In this study we were unable to validate the applicability of this model, as sediment respiration was not assessed. However, CO_2 levels fell below saturation following the emergence of phytoplankton blooms, indicating that despite likely high rates of carbon mineralization in the sediments, most of the CO_2 produced was fixed through photosynthesis. Moreover, the deposition of algal carbon to the sediments may have resulted in incomplete degradation of organic matter (Condrad et al., 2010) and in a stimulation of methanogenesis. Microbial communities may preferentially utilize more labile algal carbon than recalcitrant terrestrial carbon (Kritzberg et al., 2004; 2005). Kankaala et al. (2003) and West et al. (2006) showed that methane production is stimulated more extensively by algal than macrophyte or terrestrial detritus. Schwarz et al. (2008) simulated the sedimentation associated to a *Peridinium gatunenese* bloom onto intact lake sediments, and results indeed indicated enhanced CH_4 production and increased concentrations of propionate and acetate within the upper sediment layers. These results support a positive relationship between phytoplankton bloom and CH_4 in lakes. The mechanism in which phytoplankton bloom stimulated CH_4 production was not assessed directly in the present study. However, we show that CH_4 was more strongly correlated with Chlorophytes ($R = 0.66$) than bloom-forming cyanobacteria or Dinophyceae ($R = 0.59$). There are two possible explanations for this difference: i) anaerobic decomposition of Chlorophyceae provided better and more readily available substrates for methanogenesis, and/or ii) the rates of aerobic CH_4 production (Grossart et al., 2011) were higher in association with Chlorophytes than with Cyanobacteria. This latter effect may be associated to the production of cyanotoxins in species abundant in Lake St. Augustin (e.g., *Anabaena* sp.).

The CO_2 was negatively correlated to planktonic picocyanobacteria. This negative relationship, may be attributed to the photosynthetic uptake (Raven, 1998). The much stronger negative relationship obtained between total bacterioplankton and N_2O may suggest the biological uptake or more efficient N_2O reduction in the presence of more abundant bacteria. Nitrous oxide is produced in oxic epilimnia, in anoxic hypolimnia or at oxic-anoxic boundaries, but is consumed in completely anoxic layers (Mengis et al., 1997). It is possible that the presence of anoxic microhabitats in the epilimnion of Lake St Augustin allowed higher rates of N_2O consumption. Apparently, N_2O undersaturation is more common in shallow productive lakes than previously assumed (Lemon and Lemon 1981). For example, Lehnherr et al. (2012) reported high levels of N_2O undersaturation in Arctic wetland ponds. Jacinthe et al. (2012) observed mid-summer N_2O undersaturation at the surface of a productive reservoir. Recently, Bartosiewicz et al. (2015) showed instances of N_2O undersaturation in a small shallow lake suffering from summer phytoplankton blooms. Lake St. Augustin acted also as a sink for atmospheric N_2O but only during the summer with persistent phytoplankton blooms.

On the contrary to CO_2 and N_2O, CH_4 was strongly positively related to bacterioplankton. This may suggest that increased bacterial activity in the epilimnion was followed by higher rates of organic matter settling to the sediments and changes in benthic community. However, CH_4 may have also been produced within the microzones of settling organic matter (lake snow, Schulz et al., 2001) or aerobically in association with autotrophic bacterioplankton (Grossart et al., 2011). Because we did not assess the relative contribution from free-living or particle-attached bacteria, we cannot exclude either possibility. The positive relationship between bacteria and CH_4 also indicates relatively little contribution from methanotrophs to the total bacterioplankton biomass in the epilimnion of Lake St. Augustin.

Our comparison between the littoral and pelagial zones of the lake supports that shallow areas may at times act as hot spots for CH_4 and N_2O emissions in lakes (Wang et al., 2006; Hofmann, 2013). However, in the case of Lake St.Augustin, surface waters of the littoral zone were sometimes more undersaturated in both CO_2 and N_2O than the pelagial zone. This suggest that macrophytes, abundant despite recurrent phytoplankton blooms, were responsible for some CO_2 uptake and stimulated N_2O removal from the water column (Tall et al., 2011). Further, in agreement with our hypothesis, the link between plankton biomass and GHG was stronger in the pelagic than in the littoral zone of the lake. This may be because of the relatively greater effect of allochtonous organic matter on the GHG production in the inshore areas (e.g., Huttunen et al., 2003). On the whole lake scale, however, littoral zones contained relatively little GHG during the entire study period.

There are few studies that have sought to quantify GHG saturation and storage in small eutrophic and hypereutrophic lakes (Duarte and Prairie, 2005; Downing et al., 2008; Lazzarino et al., 2009). These systems being a subject of unprecedented attention in terms of eutrophication and occurrence of noxious alga bloom have not yet been properly accounted for in the global carbon cycle. Here we present a dataset showing the clear effects of phytoplankton bloom characteristics on GHG saturation levels of a hypereutrophic lake. The occurrence of persistent summer bloom, related to hot and dry weather conditions, resulted in changes of the entire planktonic food web, and lead to greater rates of CH_4 emissions, on the expense of CO_2. This relationship may be of crucial importance in understanding the impact of lakes on global climate, and provide arguments for a more efficient restoration of polluted and anthropogenically eutrophicated lakes. As climate warms and eutrophication progresses, more lakes around the globe are likely to suffer from noxious blooms. The composition of the blooms and associated response from the aquatic food web may have a significant effect on the global warming potential of lakes.

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PARTIE 3

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PARTIE 4 ANNEXE

4.1 Surface renewal model: calculation details

Following Macintyre et al. (2002), the momentum sensible heat and latent heat fluxes necessary to use surface renewal model were calculated using the bulk aerodynamic formula: $\tau = \rho_A C_D U_Z^2$, $H = \rho_A C_{Pa} C_H U_Z (T_s - T_z)$, $E = \rho_A L_V C_E U_z (q_s - q_z)$

where τ is the shear stress of the air at the surface interface (N m^{-2}), ρ_A is the density of the air at the air-water interface (kg m^{-3}), C_D is the drag coefficient, U_z is the wind speed at height z above the water surface (m s^{-1}), H is the sensible heat transfer (W m^{-2}), C_{Pa} and C_{Pw} are the specific heats of air and water (J kg^{-1} °C^{-1}), C_H is sensible heat transfer coefficient, T_S is the water surface temperature (°C), T_Z is the air temperature (°C) at height z, E is the latent heat transfer (W m^{-2}), L_V is the latent heat of vaporization (J kg^{-1}), C_E is the latent heat transfer coefficient, q_s is the specific humidity at saturation pressure at T_s, and q_z is the specific humidity of the air at z.

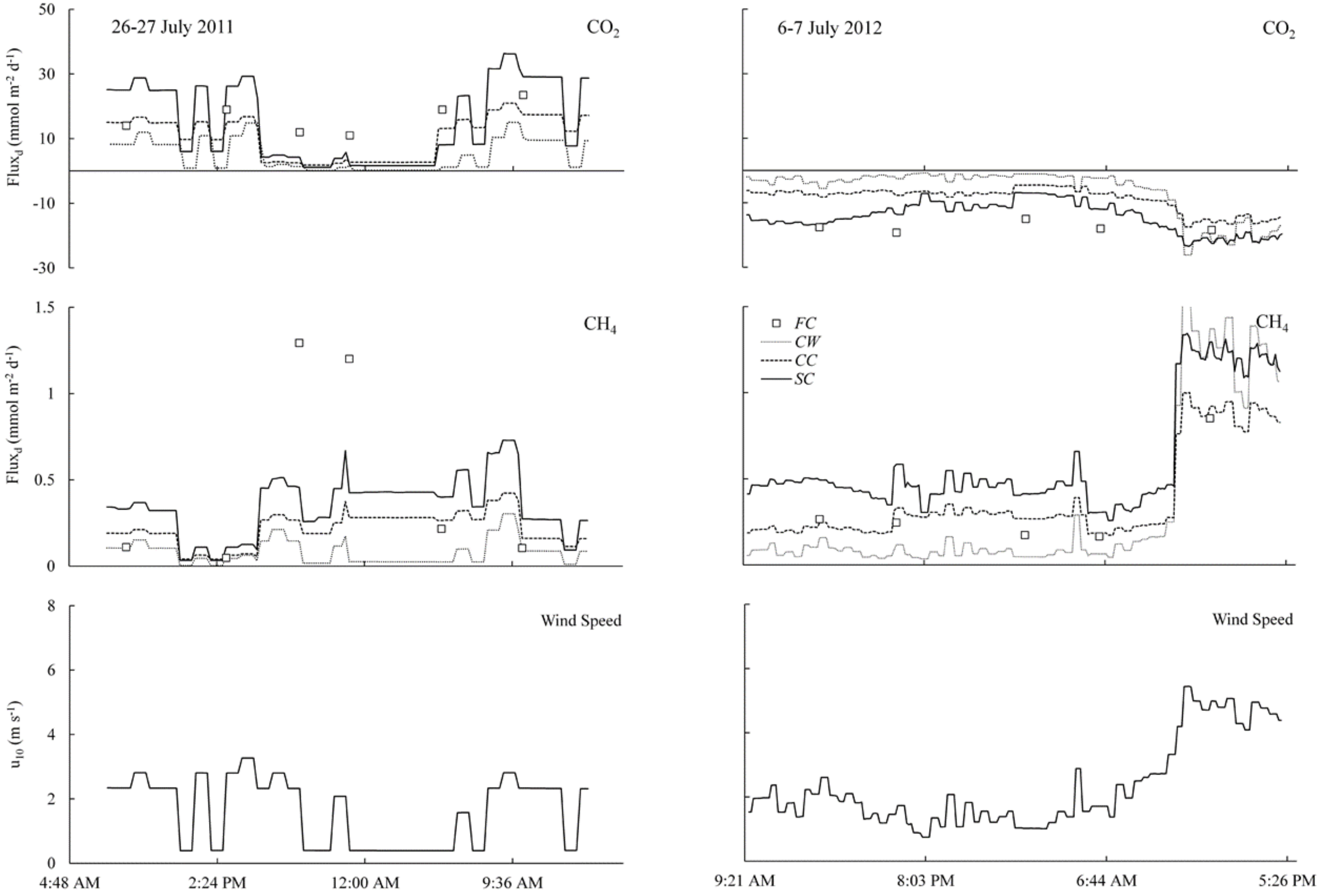
The methods of Imberger (1985) were used to assess the diurnal mixed layer dynamics, and the effective surface heat flux H (W m^{-2}) was calculated according to Kim (1976) as: $H = S + q(0) + q(h) - 2/h\ (\int_0^h q(z)dz)$, $S = \rho_0 C_{PW} [w' T'(0)]$, where S is the surface heat flux, h is the depth of the surface layer (m), q(0) is the shortwave radiation at the surface (W m^{-2}), and q(h) is the shortwave radiation at the bottom of the surface layer (W m^{-2}). Following Imberger (1985) and MacIntyre et al. (2002, 2009) the buoyancy flux was computed as: $\beta = g\alpha H\ (C_{Pw}\rho_0)^{-1}$, where α is the thermal expansion coefficient and ρ_0 is the density of water. According to Deardoff (1970) and MacIntyre et al. (2002), the penetrative convection velocity w* and the surface water shear velocity u_{*w} were calculated as: $w^* = (\beta h)^{0.5}$ and $u_{*w} = \tau\ \rho_0^{-1}$, respectively.

Interestingly, assuming equivalence of shear stress on both sides of the air-water interface, $u_{*w} = \rho a/\rho w\ C_d\ u^2$, where ρa and ρw are density of air and water respectively. Assuming a neutral drag coefficient C_d at 10 m of 0.0013, this expression simplifies to $u_{*w} = 0.001u_{10}$. Following equation from MacIntyre et al. (1995) $k = a1\ (\varepsilon/v)^{1/4} Sc^{-1/2}$, where v the kinematic viscosity, assuming neutral atmosphere stability, and converting to cm hr^{-1}, $k = (2.64u_{10})^{3/4}$. Thus the equation for the surface renewal model using similarity can be expressed simply, and the resulting curve can be seen as near linear. Contrasting it with Cole and Caraco (1998), k_{600SC} and k_{600CC}, respectively, have values of 2.6 cm hr^{-1} and 2 cm hr^{-1} for 1 m s^{-1} winds and values of 10 cm hr^{-1} versus 6 cm hr^{-1} at 6 m s^{-1} winds. Thus, k_{600SC} will, in this specific conditions, give higher estimates of fluxes. K_{600SC} provides similar values to the equation under heating in MacIntyre et al. (2010, SE), $k_{600SE} = 1.74\ (\pm 0.63)\ u_{10} - 0.15\ (\pm 2.43)$ which has only a slight offset from the formulation for k_{600} in Crusius and Wanninkhof (2003). The similarity of k_{600} following Tedford et al. (2014), MacIntyre et al. (2010), and Crucius and Wanninkhof (2003, linear case) under heating all provide support for this equation. Thus, for neutral atmospheric stability, k_{600SC} and k_{600CC} provide likely envelope for anticipated values of k. The window should be wider at low wind speeds. The drag coefficient increases when the atmosphere is unstable and decreases when it is stable, with the effect largest for winds less than ~ 2 m s^{-1} and tapering as winds increase. For winds less than 2 m s^{-1}, and substituting in the estimated drag coefficients, 0.0024 and 0.0005, the multiplier on $(u_{10})^{3/4}$ is up to 5 for the unstable case and 1.6 for the stable case. Thus, variability in atmospheric stability (factor not included to this thesis) may also contribute to variability in k with the largest effect at low wind speeds.

4.2 Supplementary information for Article 1

Diurnal variability in GHG was followed every 4-5 hours for 25 h on 26-27th July 2011 and 6-7th July 2012 (Fig. 4.2.1). The two 24 hour cycles highlight the difficulties in determining an exact model for computing fluxes. Some times fluxes measured with the chambers were equal to those computed using the surface renewal model; at other times they were equal to those computed using the wind based model. In some instances, spatial or temporal variability in surface gas concentrations may have contributed to the observed differences. For example, before the diurnal sampling in 2011, winds were above 5 m s^{-1} causing the thermocline to downwell and, on its relaxation, to upwell. These motions and associated mixing may have contributed to the larger measured than modeled fluxes near midnight. Spatial variability in CO_2 concentrations may have caused errors in modeled results as they rely on accurate gas concentrations at the water surface. During the period when winds were ~1 m s^{-1} in 2012, modeled CO_2 fluxes were larger than measured whereas modeled CH_4 fluxes were similar to measured. Again, processes within the water column may have led to the differences. For example, during the morning and early afternoon on the 7 July, an increase in CH_4 concentration was accompanied by a decrease in CO_2 concentration. Alternatively, since winds were low at night and neither meteorological station obtained measurements within the atmospheric boundary layer over the lake, input terms for both models may not have the required accuracy. Other sources of error, such as differences in controls on evasion of the two gases, cannot be addressed with this limited data set.

Supplemetary Figure 4.2.1 Diurnal variability in CO_2 and CH_4 diffusive fluxes during rainy night in 2011 and hot night in 2012 estimated using high frequency k (at 0.2 h intervals) from wind based models (CW dotted line, CC dashed line), surface renewal model (SC solid line) or directly measured using floating chambers (FC, open squares).

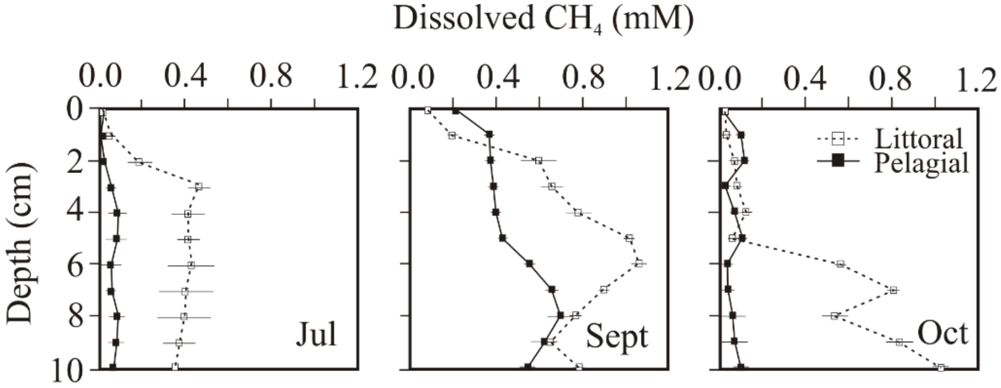


4.3 Supplemetary information for Article 3

The sediment pore water was collected in Lake St. Augustin using acrylic peepers as above. The peepers were deployed to one littoral and one pelagial site on 29 June and 4 September 2012, corresponding to the beginning and the end of the summer stratification period in this lake. The peepers were left 2-5 weeks to equilibrate as above.

The CH_4 concentrations in the porewater of Lake St. Augustin varied spatiotemporarily in 2012. For the entire studied period, porewater CH_4 concentrations were significantly higher in the littoral than in the pelagic zone of the lake (Fig. 4.3.1 $p<0.001$, Paired t-test). It increased significantly at both the littoral and pelagic zones after the collapse of the summer cyanobacterial bloom. The highest concentrations were recorded at a depth of 4-6 cm in the sediments in September (0.96 and 1.1 mM) and at 10 cm in October (1.0 mM).

Littoral sediments are hot spots for CH_4 production in Lake St. Augustin, when compared to pelagic sediments. This possibly a result of higher organic matter content of the littoral sediments and overall higher bottom temperatures stimulating CH_4 production. This difference may also result from a positive 'priming effects' described by Guenet et al. (2010). Moreover, during summer 2012 the CH_4 concentrations increased significantly in both littoral and pelagic sediments. This may have also resulted from higher rates of OM decomposition with increasing bottom temperatures. However, other possible explanation is that increased deposition of highly labile organic matter from phytoplankton bloom stimulated CH_4 production in the first 10 cm of the sediments.



Supplementary Figure 4.3.1 The profiles of CH_4 concentrations in the surface sediments of littoral (dotted) and pelagic zones (solid line) of Lake St. Augustin during (July) and after (October) the occurrence of algal blooms in 2012.

4.4 The greenhouse gas emissions during decomposition of algal and macrophyte detritus

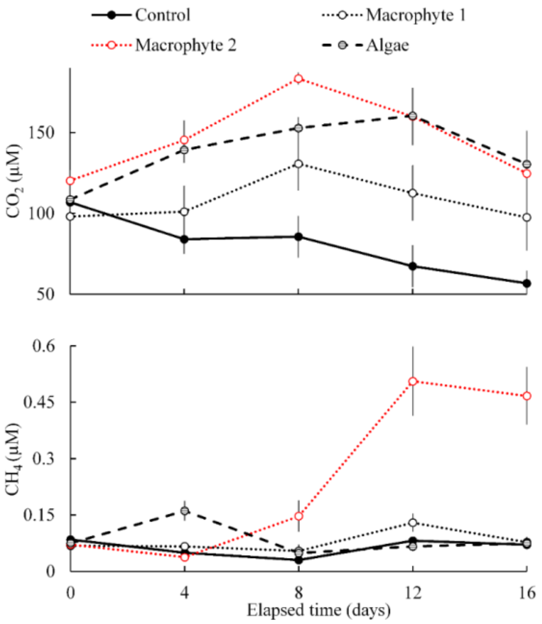
A decomposition experiment was conducted in 16 dark PVC mesocosms filled with 70L of lake water collected in the pelagial zone of Lake St. Augustin in September 2013, prefiltered through a zooplankton net (120 µm mesh). Prior to the experiment, 500 ml of homogenized surface sediments (0-10 cm) was added to each mesocosms. Subsequently, they were kept for 7 days in the experimental conditions (dark room with controlled temperature of 21°C) for acclimation. At the beginning of the experiment, four replicate mesocosms were supplemented with a solution of phytoplankton (*Nannochloropsis limnetica*), four with detritus of watermilfoil (named macrophyte 1*, Myriophyllum spicatum*), and four with detritus of water lilies (named macrophyte *2, Nuphar lutea*). The solutions were prepared adding 4 grams of organic matter powder to 300 ml of deionized water. The organic matter was prepared as follow: phytoplankton and macrophyte material was centrifuged, rinsed in deionized water, freeze dried and grinded to a fine powder. In addition to these 12 treatment mesocosms, 4 mesocosms were left undisturbed to represent the control conditions. Changes in GHG (CO_2 and CH_4) concentration in each mesocosms were followed every four days using the head space method (see section 1.4 for details).

The addition of macrophyte and phytoplankton detritus resulted in significantly different CO_2 concentration the experimental mesocosms (Fig. 4.4.1; Table 4.4.1). The addition of water lilies detritus resulted in the highest increase of the CO_2 concentrations by comparison to control ($p<0.001$, Tukey test). The effect of detritus addition on CH_4 concentrations overall was not significant ($p=0.326$). However, significantly more CH_4 was produced in the mesocosms supplemented with water lilies. The initial peak in CH_4 concentration in the mesocosms supplemented with phytoplankton was observed at day 4 of the experiment, while it was observed at day 12 and 16 for the mesocosms supplemented with macrophytes 1 and 2 respectively. This suggests that the phytoplankton biomass was more readily available to the methanogenic community as compared to the macrophyte biomass.

Water lilies seems to be the organic matter leading to the largest net production of GHG. This contradict result of Kaankala et al. (2003) who showed higher rates of GHG production from phytoplankton than macrophyte detritus. In our study we have indeed observed initially higher CH_4 production in treatment with phytoplankton detritus but only until day 8 of the experiment. Phytoplankton material is more bioavailable than recalcitrant macrophyte detritus. However, our mesocosms also contained surface sediments from Lake St. Augustin what likely resulted in 'priming effect' on the decomposition of macrophyte detritus. As a result we observed much higher GHG production in the second phase of the experiment in treatment containing plant detritus and particularly water lilies.

Table 4.4.1 Result of One-way ANOVA testing for the effect of detritus addition on CO_2 concentrations in experimental mesocosms.

Source of Variation in CO_2	DF	MS	F	P
Between Groups	3	4634.9	11.2	0.001
Residual	16	414.5		



Supplementary Figure 4.4.1 Changes in GHG concentrations in experimental mesocoms during OM decomposition.

PARTIE 5 -
5.1 Premier article publié

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PRIMARY RESEARCH PAPER

Greenhouse gas emission and storage in a small shallow lake

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Abstract Small lakes are likely to show considerable temporal variability in greenhouse gas emissions given their transient stratification and short residence time. To determine the extent that CO_2 and CH_4 emission and storage depends on surface meteorology, we studied a shallow lake during 2 years with contrasting rainfall and thermal stratification. Gas fluxes were estimated with wind-based and surface renewal models and compared to direct measurements obtained with floating chambers. The assessment of greenhouse gases storage revealed that the lake gained CO_2 in association with rainfall in both the rainier (2011) and drier summer (2012). In 2011, stratification was less extensive and disrupted frequently. The lake was a source of CO_2 and CH_4, and ebullition exceeded diffusive fluxes of CH_4. In 2012, stratification was more persistent, the lake was a sink for CO_2 during dry periods, CO_2 and CH_4 accumulated in the hypolimnion later in the summer when rainfall increased, diffusive fluxes of CH_4 were similar to those in 2011 mid-summer and over four times higher during overturn. Ebullition was lower in the drier summer. Fluxes measured with chambers were closer to estimations from the surface renewal model and about two times values estimated with wind-based models.

Keywords Small lake · Weather variability · Thermal stratification · Greenhouse gases · Storage · Diffusive flux · Ebullition · Gas transfer modeling

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Introduction

Small and shallow lakes and impoundments are abundant in the landscape (Downing et al., 2006; Downing, 2010) and emit substantial quantities of atmospheric greenhouse gases (GHG) (Casper et al., 2000; Repo et al., 2007). They are active reactors where carbon derived from the catchment, atmosphere, and ground waters can be stored, utilized, chemically altered, or released as CO_2 and CH_4 (Kling et al., 1992; Cole et al., 2007). The rate and direction of CO_2 transport from lakes depends on the ratio of primary production to ecosystem respiration which in turn are driven by inputs of inorganic nutrients and allochthonous and autochthonous organic material

(Kortelainen et al., 2006). The rate of CH_4 transport to the atmosphere depends on the rates of methanogenesis (Zeikus & Winfrey, 1976) and methanotrophy (Rudd & Hamilton, 1975; Whiticar et al., 1986). Methane can diffuse from the lake surface or be released via gas bubbles formed in the sediments (Bastviken et al., 2011; Wik et al., 2013). The amount of gas released to the atmosphere in these two modes of transport depends on water depth and temperature (Hofmann et al., 2010), on the composition, temperature, and chemical characteristics of the sediments (Martinez & Anderson, 2013; Wik et al., 2014), and on turbulent mixing as it causes fluxes within the water column and across the air–water interface.

Estimations of GHG flux from inland waters are increasing in accuracy due to improvements in modeling of the gas transfer coefficient k, more extensive data on dissolved gas concentrations (Raymond et al., 2013), and more detailed description of seasonal dynamics in GHG storage (Fernández et al., 2014). Further improvement requires understanding how weather and its effects on stratification and turbulence control the production, emission, and storage of GHG in lakes of different morphometries. For example, GHG emissions from a dimictic lake were shown to increase during cold fronts when water from below the mixed layer was entrained (Aberg et al., 2010), and during fall as summer stratification breaks down (Vesala et al., 2006). The study by Ojala et al. (2011) illustrates between year variability in a dimictic lake due to intrusions from rainfall. Few studies have been undertaken in shallow polymictic lakes in which diel stratification and mixing would be expected to lead to similar GHG storage and fluxes in summer and fall.

In shallow polymictic lakes, water temperature and oxygen concentrations may change rapidly with changing weather (Wilhelm & Adrian, 2008). Hence, the seasonal and inter-annual variability in GHG storage and fluxes, both diffusive and ebullitive, may be markedly different depending on weather conditions (Natchimuthu et al., 2014). These conditions affect GHG through their effect on sediment re-suspension (Bussmann, 2005; Hofmann et al., 2010) and on inflows of ground and stream waters, which can be considerable given the short residence time of such small lakes. They also affect the water column thermal structure and the intensity of turbulent mixing controlling gas diffusion at the air–water interface (Jahne et al., 1987; MacIntyre, 1993). Diffusive gas fluxes at the lake surface can be assessed using wind-based models (e.g. Cole & Caraco, 1998), surface renewal models (MacIntyre et al., 2010; Tedford et al., 2014), or measured directly using floating chambers (Soja et al., 2014). Depending on the heterogeneity in surface water GHG concentrations and on factors controlling turbulent mixing, the choice of calculation method can introduce biases. The amount of energy incorporated into the water column due to wind stress or heating/cooling of the water column and sediments has also been shown to affect ebullitive fluxes (Wik et al., 2014).

In this study, we describe the inter-annual differences in stratification and surface meteorology between a cool rainy summer and a dry hot summer for a small 2 m deep lake. We hypothesized that when exposed to contrasting weather conditions, the lake will alternatively be polymictic or stably stratified during the summer, with subsequent effect on GHG emission and storage. We compare GHG diffusive fluxes calculated with wind-based and surface renewal models, and evaluate these against floating chamber measurements. We tested if stable stratification as opposed to conditions with more frequent mixing events results in a delayed release of diffused GHG until autumnal overturn and globally higher CH_4 emissions. We also hypothesized that the amount and composition of the gas released via ebullition will depend on water column stratification and hypolimnetic temperature.

Methods

Study site

Lake Jacques is a small ($0.18\ km^2$), shallow lake (maximum depth = 1.9 m, mean depth = 0.75 m) located 30 km north of Quebec City (QC, Canada), and supplied with water from two creeks in the eastern part of the lake, as well as from a ground water spring in its southern part. The lake can be classified as meso-eutrophic in terms of total phosphorus (mean summer value of $21\ \mu g\ l^{-1}$), total nitrogen ($0.7\ mg\ l^{-1}$), and Chl *a* concentrations ($17\ \mu g\ l^{-1}$). Approximately, 50% percent of the lake surface area is covered with the macrophyte *Brasenia schreberi* in summer. The upper 10 cm of the sediments has high organic carbon content (loss on ignition = $38 \pm 5\%$, $n = 10$). The water retention time averages 14–20 days in summer (R. Tremblay, *pers. comm.*). The concentration of dissolved organic carbon ranged between 2.2 and 3.0 mg l^{-1} during the study and an algal bloom formed on the surface in 2012.

Meteorological and physicochemical characteristics

In 2011, hourly data on air temperature, relative humidity, wind speed, wind direction, dew point, and precipitation were obtained from the meteorological station of Environment Canada located 7 km from the lake (anemometer threshold 1 m s^{-1}). In 2012, a meteorological station WeatherHawk 511 (anemometer threshold 0.6 m s^{-1}) mounted 25 m from the lake shore and 7 m above the water level, recorded air temperature, relative humidity, wind speed and wind direction every 30 min. Between 21 July and 6 August 2012, the on-site anemometer had to undergo maintenance and wind speed data were replaced by those from the Environment Canada station. To correct the wind speed data from the Environment Canada meteorological station, we performed a linear regression analysis when both sensors were operational in 2012. We used the resulting equation, $u_1 = 0.442 + 0.791u_2$ ($R^2 = 0.65$, $P < 0.001$), to correct wind speeds (u) from 2011 and those from 21 July and 6 August in 2012. Downwelling hourly solar irradiance data (short wave and long wave) for both years were obtained from high spatial resolution (0.125–0.125°) surface meteorological forcing model from NASA (Giovanni Interactive Tool for Data Visualization and Analysis). Upwelling short wave and long wave were computed as in MacIntyre et al. (2014). Diffuse attenuation coefficient for downward visible light (LI-COR spherical quantum sensor LI-193) was calculated as the slope of the linear regression of $\ln(E_z/E_0)$ versus depth, where E_Z is the irradiance at depth z and E_0 is the surface irradiance, and used in surface renewal model computing. The value was on average equal to 2.4 m^{-1}.

In 2011, surface temperature, conductivity, dissolved oxygen (DO), and pH were assessed fortnightly with a 600R multiparametric probe (Yellow Springs Instrument). The oxygen probe was calibrated at the beginning of each sampling day in water-saturated air. Surface water samples (100–500 ml) were filtered on GF/F glass fiber filters (0.7 μm nominal mesh size; Advantec MFS Inc.) for the determination of chlorophyll-a concentration (Chl *a*) using a UV–Vis spectrophotometer at 750 and 665 nm (Wintermans & De Mots, 1965). In 2012, all measurements were performed weekly from May until the end of August, and then fortnightly until the ice cover was formed. The lake properties were always assessed between 10:00 a.m. and 2:00 p.m. Time series of water temperature were obtained with a thermistor chain installed from June to September 2011 and from May to October 2012. In 2011, seven temperature loggers (Onset Tidbit v2; accuracy 0.2°C, resolution 0.2°C, response time 5 min) were deployed at 0, 0.2, 0.4, 0.7, 1.0, 1.5, and 2.0 m, and acquired data every 15 min (sampling site 1.9 m deep at time of deployment, Fig. 1). In 2012, the loggers were deployed at the same site (depth of 1.7 m at time of deployment) at 0, 0.2, 0.35, 0.5, 0.75, 1.2, and 1.5 m, and acquired data every 8 min. Isotherms were calculated using linear interpolation. The surface energy budget and computation of dissipation rate of turbulent kinetic energy used in gas transfer coefficient calculations were computed following MacIntyre et al. (1995, 2002) and Tedford et al. (2014).

Indirect estimations of *k* with wind-based and surface renewal models

The wind speed normalized at 10 m (u_{10}), according to the logarithmic wind profile relationship including atmosphere stability effects (Smith, 1988), was used to calculate the gas transfer coefficients (k_{600}) standardized to a Schmidt number (*Sc*) of 600 following Wanninkhof (1992). Using the equations of Cole & Caraco (1998; hereafter *CC*), we calculated

$$k_{600} = 2.07 + 0.215u_{10}^{1.7} \qquad (1)$$

and the equation of Crusius & Wanninkhof (2003; hereafter *CW*)

$$k_{600} = 0.168 + 0.228u_{10}^{2.2} \qquad (2)$$

The equation in Cole & Caraco (1998) applies for wind speeds up to 9 m s^{-1} whereas that of Crucius & Wanninkhof (2003) to winds up to 6 m s^{-1}.

We also computed the gas transfer coefficient with the surface renewal model (MacIntyre et al., 1995; Zappa et al., 2007, hereafter *SC*):

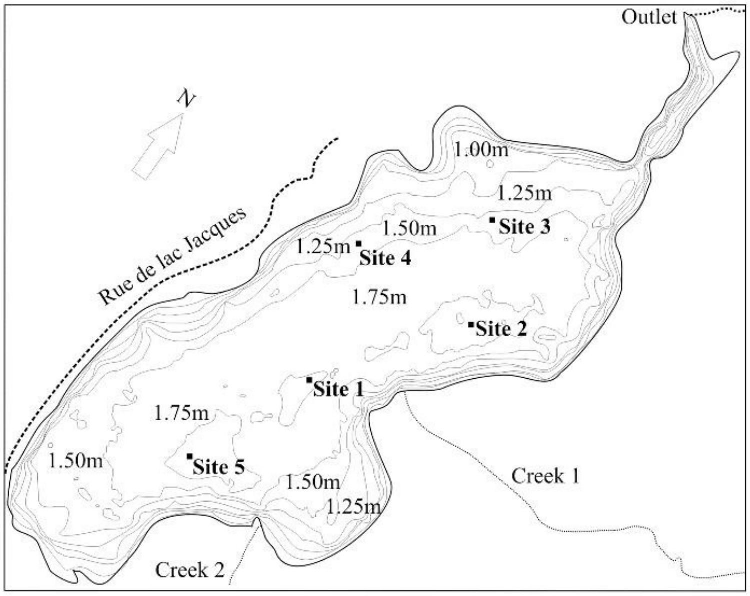

Fig. 1 The morphometry and bathymetry of Lake Jacques. The thermistor chain was positioned at *site* 1 and ebullition funnels at *sites* 1–4 (2011) and 1–5 (2012)

$$k_{600} = a_1(\varepsilon/v)^{1/4} Sc^{-1/2}, \tag{3}$$

where a_1 is an experimental constant (assumed as 0.5 in this study), ε is the kinetic energy dissipation rate, and the kinematic viscosity. The kinetic energy dissipation rate, describing turbulence in the surface mixing layer where mixing is directly energized by wind shear and convection (Imberger, 1998), was calculated during heating as $\varepsilon = 0.6\,u^3_{*\mathrm{w}}/kz$ and during cooling as $\varepsilon = 0.56\,u^3_{*\mathrm{w}}/kz + 0.77\beta$, where $u_{*\mathrm{w}}$ is the water friction velocity computed from wind shear stress, k is the von Karman constant, z is the depth equal to 15 cm, and β is the surface buoyancy flux (Tedford et al., 2014). For details on computing $u_{*\mathrm{w}}$ and β, see MacIntyre et al. (2002, 2014). The first equation implies wind as the dominant factor responsible for turbulence at the lake surface, while the second one includes cooling as an additional factor that generates turbulence.

Dissolved GHG measurements and flux estimations

Aqueous concentrations of CO_2 and CH_4 were determined fortnightly from June to mid-September in 2011, and weekly from May to the end of August in 2012, then fortnightly until the ice cover was formed. Gas sampling was performed by equilibrating 2 l of lake water with 20 ml of ambient air, shaking for 3 min, and then injecting the headspace gases into a 5.9 ml Exetainer (Labco Scientific) previously flushed with helium and vacuumed (Hesslein et al., 1991). The procedure was always repeated three times and yielded CV on average less than 10%. Gas samples were taken within 5 min after collecting the lake water, and were kept in the dark at 4°C until analyzed by gas chromatography (Varian 3800 with a COMBI PAL head space injection system). In addition, vertical profiles of CO_2 and CH_4 (resolution 0.25 m) were taken monthly or fortnightly from June to September in 2011 and from May to October in 2012 following the procedure described above. To assess gas storage, the total mass of CO_2 and CH_4 above (or below) saturation was calculated by multiplying the average concentration over a depth interval by the water volume within that depth interval, and summing over the depth of the lake (Rudd & Hamilton, 1978).

Diffusive CO_2 and CH_4 flux ($Flux_{\mathrm{d}}$) was calculated using the gas transfer coefficients for a given gas k (cm h^{-1}) estimated with either *CC*, *CW*, or *SC* models as:

$$Flux_{\mathrm{d}} = k\left(C_{\mathrm{sur}} - C_{\mathrm{eq}}\right), \tag{4}$$

where C_{sur} is the gas concentration in surface water (mmol l^{-1}) and C_{eq} is the gas concentration in the water at equilibrium with the atmosphere. Global values of atmospheric partial pressures (IPCC, 2007) were used to determine C_{eq}. The gas transfer coefficient was calculated as:

$$k = k_{600}(Sc/600)^{c}, \tag{5}$$

where c equals -0.5 for rough surfaces (Csanady, 1990).

Synchronized with the determination of aqueous CO_2 and CH_4 concentrations, a floating chamber (circular, 23.4 l, hereafter *FC*), made of 10 mm thick PVC plastic with floaters distributed evenly on the sidewall, extending 4 cm into the water, was coupled with an infra-red gas analyser (EGM-4, PP-Systems), and deployed 2 m from the boat on the surface of the lake to directly assess CO_2 flux ($Flux_{\mathrm{c}}$) according to

$$Flux_{\mathrm{c}} = S\,M_{\mathrm{w}}V_{\mathrm{ch}}(V_{\mathrm{m}}A)^{-1}, \tag{6}$$

where S is the slope of the linear regression of gas concentration in the chamber versus time (measurements taken for a maximum of 20 min depending on flux rate), Mw the gas molecular weight, V_{ch} the volume of the chamber, V_{m} the gas molar volume at ambient temperature, and A the area of the chamber. Atmospheric and surface water dissolved CO_2 together with direct measurements of CO_2 flux were used to estimate the floating chamber gas transfer coefficients using the equation:

$$k = Flux_{\mathrm{c}}/\left(C_{\mathrm{sur}} - C_{\mathrm{eq}}\right) \tag{7}$$

k was standardized to k_{600} using Eq. 5. Gas transfer coefficients obtained from the *FC* were used to calculate the chamber CH_4 flux following Eq. 4.

Ebullition measurements

The rate of gas ebullition was measured with four (2011) or five (2012) submersible inverted funnels installed monthly from June to September 2011 ($n = 14$ measurements, only two funnels worked in August), and 2012 ($n = 20$ measurements). The funnels consisted of a soft PVC cone-shaped body mounted on a metal frame, with a plastic syringe and luer-lock valve installed on top, and maintained with floaters at approximately 20 cm below the water surface. At each sampling date, the funnels were submerged in the lake at one of the four or five pelagic sites (Fig. 1) and moored with three weights deployed at least 2 m away from the funnel to avoid the collection of bubbles escaping at installation and to keep the funnel upright in the same position for 4–24 h depending on the flux rate. The collected bubbles were sampled in triplicate vials, and the samples diluted 100 times with helium before they were analyzed by gas chromatography as described above. The ebullitive flux was calculated as:

$$Flux_{\mathrm{e}} = C\,V_{\mathrm{g}}V_{\mathrm{m}}^{-1}A^{-1}, \tag{8}$$

where C is the concentration of a given gas in the syringe, V_{g} is the total volume of the gas in the syringe, V_{m} is the gas molar volume at ambient temperature obtained from meteorological station, and A is the area of the funnel.

Finally, the total global warming potential over a 100 year period (GWP) was calculated as the sum of diffusive and ebullitive GHG fluxes with CH_4 having 34 times higher GWP than CO_2 (nominal 1; Myhre et al., 2013).

Results

Meteorological conditions and mixing of the lake

Air temperatures varied on seasonal and diel time scales. Maxima in summer were near 30°C; shifts between warm and cold fronts caused 5°–10°C changes in air temperatures, similar to variations over the course of a day (Fig. 2). Three to four day long cold fronts prevailed in 2011 starting from the end of June. In early July 2011, they were associated with rainfall and wind speeds reaching 5 m s^{-1}. Warm fronts later in July caused the lake surface to warm to 26°C, brought heavy rains and winds up to 8 m s^{-1} during the daytime. Winds often declined at night below the anemometer's threshold (1 m s^{-1}). Cold fronts with air temperature falling to 15°C and rainfall dominated the weather of early August 2011, but the winds at this period did not exceed 5 m s^{-1}. Early in September, cold fronts were associated with heavy rains, but the highest winds, 6 m s^{-1}, were recorded later in the month during a warm front. The rate of

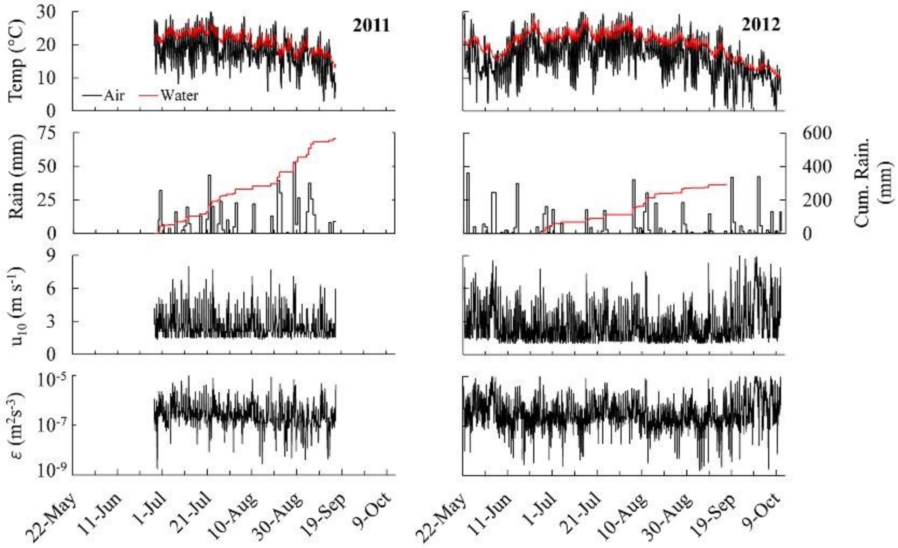

Fig. 2 Meteorological conditions including air and surface water temperatures, daily and cumulative rainfall, wind speed at 10 m (u_{10} when winds were above 1 m s^{-1}), as well as kinetic energy dissipation rate (ε) following Tedford et al. (2014)

dissipation of turbulent kinetic energy computed at 15 cm depth following Tedford et al. (2014) indicated that ε ranged between 10^{-8} and 10^{-6} m^2 s^{-3}, that is, the upper water column was moderately turbulent (MacIntyre et al., 2009).

In 2012, warm fronts prevailed, starting from May when air temperatures reached 28°C and afternoon winds remained above 6 m s^{-1}. As in 2011, winds were low at night. In early June cooler air masses were associated with rain and daytime winds above 6 m s^{-1}. At this time, the lake was cooling also during the day and buoyancy contributed to the turbulence at the surface. The weather in July and August 2012 was dominated by warm fronts with little rain particularly in July. On average, 2011 was cooler and windier than 2012 and had almost two times higher rainfall (Table 1).

Table 1 Meteorological conditions, including air temperature (air *T*), cumulative precipitation (cum. rain), daily precipitation (daily rain), wind speed (wind), downwelling short wave (SW) and long wave (LW) irradiation, relative humidity (RH), and the dissipation rate of turbulent kinetic energy (ε) as a mixing index at Lake Jacques in summer 2011 and 2012

Meteorology	2011	2012
Air *T* (°C)	17.5	18.2
Cum. rain (mm)	563	292
Daily rain (mm day^{-1})	8.4	4.3
Wind (m s^{-1})	2.9	2.4
SW (W m^{-2})	213	222
LW (W m^{-2})	349	365
RH (%)	75.6	73.5
$\varepsilon \times 10^8$ (m^2 s^{-3})	62.4	49.6

The comparison of wind speeds and dissipation rates in the actively mixing layer was done for winds exceeding 1 m s^{-1}

Thermal stratification, pronounced in both years despite the lake's shallow depth, was greater in 2012 than 2011 (Fig. 3). For example, the temperature difference from top to bottom was 8°C in mid-July 2011 and 13°C in mid-July 2012. The strong near surface heating is a result of the relatively high diffuse attenuation coefficient in this lake, 2.4 m^{-1}. In both summers, shallow diurnal thermoclines formed in the upper 20–60 cm. In 2011, the afternoon winds caused episodes of apparent mixed layer deepening. These patterns result from winds tilting the thermocline; on relaxation, the warm water was found at shallower depths and cooler water upwelled. Similar events occurred in 2012, but the penetration of warm water

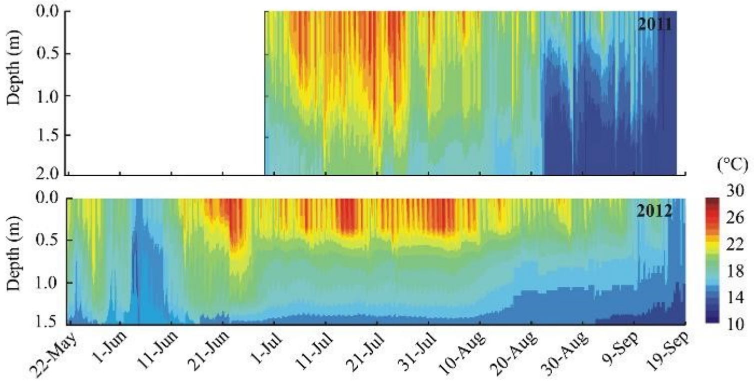

Fig. 3 Thermal structure of the lake during rainy, cooler summer 2011 and dry, hotter summer 2012

was restricted to shallower depths. Intervals with warm temperatures in the upper meter were interspersed with ones with cooler due to the passage of cold fronts during which near surface temperatures dropped by ~10°C. In consequence, the temperature differences within the water column were reduced to 2°C in 2011 and 7°C in 2012 (Fig. 3).

During cold fronts, temperatures often decreased in the lower water column. The large decreases in temperature in the lower water column in 2011 co-occurred with rain events and may be the signature of cold inflows of stream water. The transport could also be associated with differential cooling between pelagic and littoral zones. For example, at the pelagic site, temperatures cooled from 20 to 22 July in the thermocline between 40 and 80 cm but not at one meter depth. As inflows from rain were low at that time, the mid-water column cooling, with temperatures similar to the inshore site, is indicative of offshore flows from differential cooling. Even in 2012, cold fronts tended to be associated with rainfall, so cool water intrusions may have been due to the combination of increased stream/ground water inflows and/or differential cooling.

Stratification, quantified as buoyancy frequency, $N^2 = g\,\rho^{-1}\,\mathrm{d}\rho/\mathrm{d}z$, where g is the acceleration constant due to gravity, ρ is the water density, and $\mathrm{d}\rho/\mathrm{d}z$ is the vertical density gradient, showed layering with features typical of seasonally stratified lakes. The upper water column stratified in the day and mixed at night, and comprised the epilimnion. For the first half of summer 2011, the metalimnion, with more persistent stratification, began at 0.8 or 1 m and extended nearly to the bottom with N having maximal values of 60 cycles per hour (60 cph = 0.1 rad s^{-1}). To provide context, oceanographers consider water to be strongly stratified when $N > 20$ cph. For most of summer 2012, the metalimnion began between 0.3 and 0.6 m and extended to 0.6–0.8 m with maximal values of N near 100 cph. Below the metalimnion, buoyancy frequencies were higher than in 2011. Importantly, in 2011, the stratification across the metalimnion intermittently weakened throughout its depth, with values dropping to 30 or 40 cph. In contrast, this weakening did not occur in 2012. The weakening is indicative of periods with increased mixing, which reduce the stratification and enable transport of dissolved gases across the metalimnion.

In both years, increased rainfall after mid-August weakened the stratification in the lower water column, with N dropping below 30 cph in 2011 and below 50 cph in 2012. During this latter period in 2011, there were intervals when buoyancy frequencies became nearly uniform throughout the water column. In 2012, buoyancy frequencies only became near uniform with values below 20 cph in concert with the large rainfall event on 16 September (Fig. 2). Thus, rainfall contributed to weakening of vertical stratification. We infer that the persistent rainfall in summer 2011 enabled frequent exchanges between the upper and lower water column. The persistently high metalimnetic values of N in 2012 (50 cph), until the rainfall events later in the summer, imply reduced vertical fluxes between the metalimnion and epilimnion.

Variability in GHG vertical distribution and departure from saturation

The differences in stratification and rainfall between years led to differences in vertical distribution of dissolved gases. The oxycline tended to be located between 0.5 and 1.5 m in both years, with anoxia prevalent below 1.5 m in 2012 (Fig. 4). Concentrations of dissolved gases differed between sampling periods in both years, with large increases prevalent either near the bottom or in mid-water intrusions (Fig. 4). In 2011, increases of CO_2 were associated with the higher frequency of rainstorms. For example, the large increase of CO_2 in the hypolimnetic waters on 6 August followed heavy rains on 5 August. Dissolved CH_4 did not accumulate in the lower water column during summer 2011. The higher temperature and oxygen levels in the lower water column in 2011 relative to 2012 provide evidence for larger vertical exchanges in 2011. Were CH_4 produced in 2011, it would have been oxidized quickly. In 2012, high concentrations of CO_2 and CH_4 did not occur in bottom waters during July but increased in August, and de-gassing occurred when stratification was eroded in September. Increases in gas concentrations in August 2012 co-occurred with increased rainfall, decreases in near-bottom temperatures, and cooling indicated by heat budgets (averaged over 2 days). Concentrations of CH_4 also increased inshore (data not shown). Thus, rainfall events appear to have caused loading of either organic matter or CO_2 in both years, with stronger stratification, anoxia and weaker mixing in 2012 enabling the persistence of CH_4 in the hypolimnion.

Surface CO_2 concentrations were significantly higher in 2011 than in 2012 (mean of 16.3 and 8.6 μM respectively, $P < 0.05$). On average, surface CH_4 concentrations were higher in 2012 than in 2011 (0.89 and 2.1 μM respectively, $P < 0.05$), but this latter difference was mainly due to the large increase of dissolved CH_4 in the water column during fall overturn. The variability in GHG saturation levels was higher in 2012 (CV = 145 and 210% for CO_2 and CH_4 respectively) than in 2011 (CV = 58 and 45%).

Comparison of gas transfer coefficients calculated with wind-based and surface renewal models

The comparison of gas transfer coefficients was done for data with wind speeds above 1 m s^{-1} (anemometer thresholds) and below 6 m s^{-1} (upper wind speed limit for the *CW* model), thus allowing similar conditions for

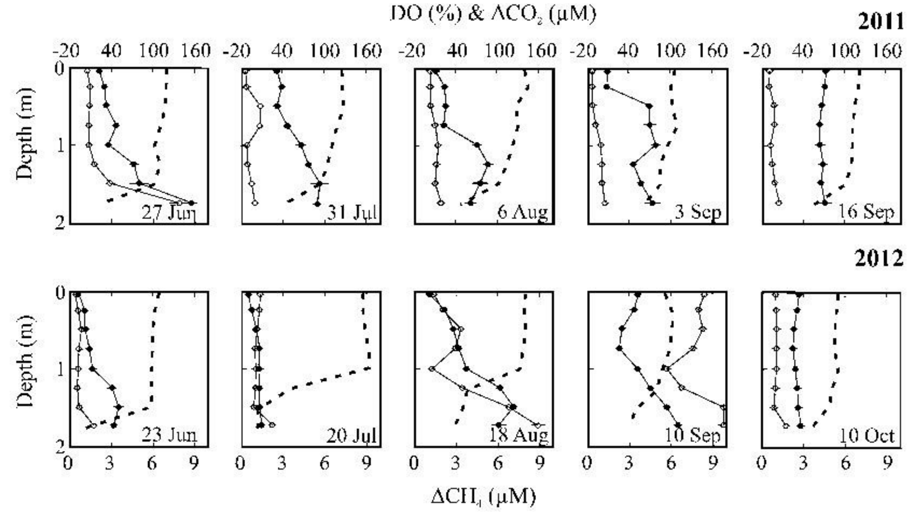

Fig. 4 Profiles of dissolved oxygen saturation (*dotted line*, *upper axis*) as well as the departure from saturation of CO_2 (*closed circles*, *upper axis*) and CH_4 (*open circles*, *lower axis*) in Lake Jacques during 2011 and 2012

Table 2 The gas transfer coefficient k_{600} and its variability (CV) for two wind-based models (*CW* and *CC*) and a surface renewal model (*SC*) at Lake Jacques during the rainy, cooler (2011) and dry, hotter summer (2012)

Model	2011		2012	
	k_{600} (cm h^{-1})	CV (%)	k_{600} (cm h^{-1})	CV (%)
CW	2.94	80	2.22	100
CC	3.47	27	3.15	28
SC	5.22	30	5.28	29

all models used in calculations. Overall, k was higher when estimated with the surface renewal (*SC*) model than when estimated with wind-based models and k was higher with *CC* than *CW* (Table 2). The contribution from buoyancy flux was negligible. The application of different models to estimate k_{600} revealed significant differences between years in average values (Mann–Whitney test, $P < 0.05$), significant differences between models ($0.001 < P < 0.05$) in the same year, and significant differences in the magnitude of k_{600} variability (coefficient of variation, CV) calculated with wind-based (*CW*) and surface renewal models ($P < 0.05$). The higher values using the surface renewal model result because k_{600} increases more rapidly with wind speed than in the Cole & Caraco (1998) formulation, similar to results shown in MacIntyre et al. (2010) under heating. The larger variance with *CW* results because it is a power law formulation with steep curvature relative to *CC*, and the lower average values result because the curve goes through zero at low winds and provides lower estimates of k at low winds than *CC* (Banerjee & MacIntyre, 2004). Because of this bias, we do not include *CW* in the comparisons with the floating chamber to follow.

Comparison of GHG fluxes estimated with floating chambers and models

The relative magnitude of fluxes measured with the floating chambers was similar to modeled values (Table 3). The winds were above the anemometer threshold when chamber measurements were conducted. With respect to CO_2, chamber fluxes were more similar to fluxes estimated using the surface renewal model (*SC*) than with *CC* wind-based model (Fig. 5A, B). The correspondence was still good between measured fluxes and *SC* estimates during autumnal overturn. With respect to CH_4, results of both models differed at most by 30% from these calculated with the floating chamber in 2011 (Fig. 5C). During 2012, *FC* fluxes were again similar to both models early in the season, they were lower than both models in mid-summer, and similar to *CC*-modeled fluxes during autumnal overturn (Fig. 5D).

Temporal variability in GHG diffusive fluxes and storage

On average, higher CO_2 diffusive fluxes occurred in 2011 than in 2012 (June–September; $P < 0.01$, ANOVA with Tukey test; Table 4; Fig. 5). In summer 2011, the lake was continuously a net source of CO_2 to the atmosphere. Similarly, it was a source in early and late summer 2012, but a sink in mid-summer. In contrast, diffusive CH_4 fluxes were higher on average in 2012 than in 2011 ($P < 0.01$). The highest CO_2 fluxes occurred in September when stratification was eroded, with estimates averaging 89 and 97 mmol m^{-2} day^{-1} for *SC* model and 86.5 and 96.5 mmol m^{-2} day^{-1} for *FC*, in 2011 and 2012, respectively. The CH_4 emissions also increased at the autumnal overturn, but only in 2012, with diffusive fluxes reaching 14 mmol m^{-2} day^{-1} for *SC* model and 7.95 mmol m^{-2} day^{-1} for *FC* measurements (Fig. 5).

The assessment of GHG storage revealed that, despite an efflux at the surface, the lake was gaining CO_2 early in 2011 (June–July) in association with heavy rainfalls (Figs. 2, 6). By the end of August, rainfall increased again, and so did the CO_2 concentrations in the lake. This rise continued until mid-September when CO_2 exceeded 16×10^3 mol. The CH_4 tended to follow a pattern opposite to CO_2 as it decreased early in the summer during the rainy period, increased when rainfall was relatively low, and decreased again later in the season. In 2012, the lake lost CO_2 between June and July when rainfall was low and Chl *a* increased from 13 to 37 µg l^{-1}. The lake gained CO_2 beginning in August with large increases in mid-August and in September when rainfall increased and temperatures in the lower water column decreased (Figs. 2, 3). The stable stratification resulted in the development of anoxia in the hypolimnion in the summer, and accumulation of CO_2 and CH_4 reaching 12.6×10^3 and 19.7×10^2 mol, respectively, despite the efflux of both gases at the surface (Fig. 5). While a considerable amount of GHG was

Table 3 Average GHG diffusive fluxes (CO_2/CH_4) in 2011 (June–September) and 2012 (May–October) measured with floating chambers (*FC*) or calculated using a wind-based model (*CC*) and a surface renewal model (*SC*), as well as the correlation coefficients (*R*) between modeled and measured CO_2 fluxes considering *FC* as the reference ($P < 0.01$)

Flux$_d$ (CO_2/CH_4)	*FC* (mmol m^{-2} day^{-1})	*CC* (mmol m^{-2} day^{-1})	*SC* (mmol m^{-2} day^{-1})	$R_{FC:CC}$	$R_{FC:SC}$
2011	29.1/0.6	18.7/0.65	29.4/1.02	0.84	0.96
2012	9.0/1.35	4.5/1.52	9.2/2.80	0.90	0.98

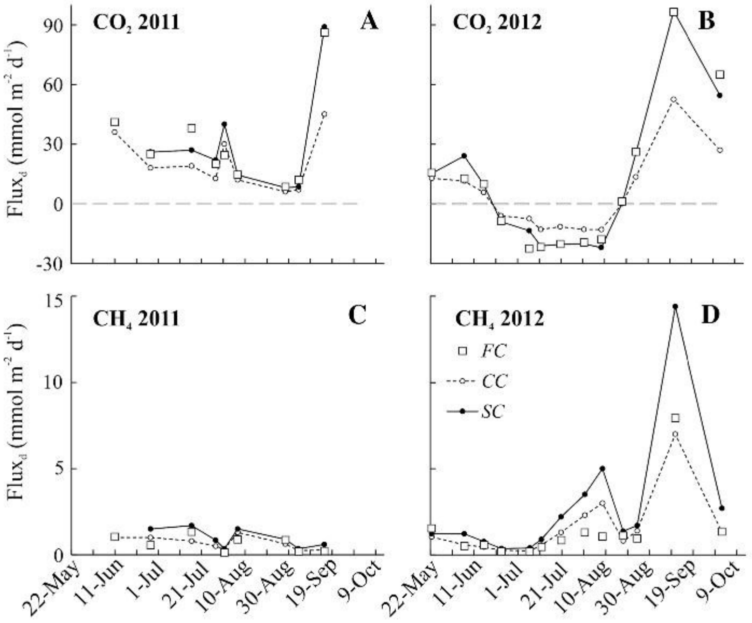

Fig. 5 The CO_2 and CH_4 diffusive fluxes (*Flux*$_d$) in 2011 and 2012 estimated with wind-based (*CC*, *dashed line*) and surface renewal model (*SC*, *solid line*) or measured directly (CO_2) and estimated with floating chambers (*FC*, *open squares*)

Table 4 Monthly average diffusive fluxes (*Flux*$_d$) using surface renewal model (*SC*), ebullition fluxes (*Flux*$_e$), partial pressure of CH_4 and CO_2 in the collected bubbles (P_eCH_4 and P_eCO_2 in %), and the global warming potential (GWP) of GHG emitted by Lake Jacques in summer 2011 and 2012 (given as CO_2-equivalent)

	CO_2 *Flux*$_d$ (mmol m^{-2} day^{-1})	CH_4 *Flux*$_d$ (mmol m^{-2} day^{-1})	CH_4 *Flux*$_e$ (mmol m^{-2} day^{-1})	P_eCH_4 (%)	P_eCO_2 (%)	GWP (mmol m^{-2} day^{-1})
2011						
June	26.5	1.4	3.4	21.4	4.1	192
July	29.7	1.0	5.3	30.5	0.8	242
August	11.3	1.2	5.1	31.2	1.8	225
September	48.8	0.5	2.4	42.3	0.6	146
2012						
June	8.3	0.7	1.1	14.4	5.0	72
July	−18.7	1.7	0.5	39.1	2.3	57
August	1.2	2.6	0.3	42.5	0.7	102
September	75.5	8.5	0.5	51.3	0.02	376

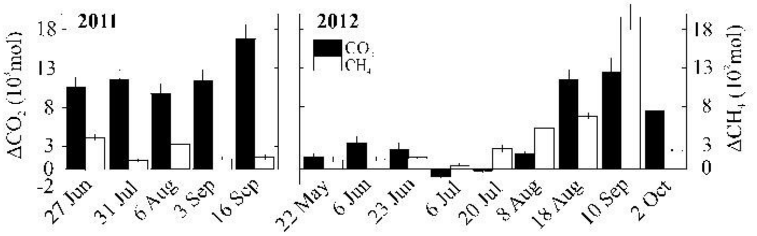

Fig. 6 The storage (mass relative to saturation ± SD) of CO$_2$ (*closed bars, scale on right axis*) and CH$_4$ (*open bars, scale on left axis*) in Lake Jacques in 2011 and 2012

lost during the overturn period in 2012, the mass of each gas above saturation was still significant in October.

GHG ebullition

Methane ebullition was higher during the rainier summer 2011 (4.1 mmol m^{-2} day^{-1}; $n = 14$) than 2012 (0.6 mmol m^{-2} day^{-1}, $n = 20$; $P < 0.01$, Table 4). It was also higher during rainier months of 2012. Overall in 2011, ebullition contributed 66% of the total lake CH$_4$ flux in June, and more than 80% in July, August and September (comparing with *SC* diffusive flux). In 2012, the contribution of ebullition decreased from 58% in June, to 24% in July, 16% in August, and 5% in September. The collected gas bubbles contained, on average, more CH$_4$ in 2012 than in 2011 ($P < 0.01$; $n = 34$, partial pressures are provided in Table 4). The bubbles contained small amounts of CO$_2$, thus ebullition contributed only from 0.02 to 4% of the total CO$_2$ flux, respectively, for both years. Interestingly in 2012, when diffusive CO$_2$ flux was negative, CO$_2$ ebullition flux ranged from 0.28 mmol m^{-2} day^{-1} in July to 0.015 mmol m^{-2} day^{-1} in August. The total GWP of the lake was 32% higher in 2011 (201 mmol CO$_2$ equivalent m^{-2} day^{-1}) than in 2012 (152 mmol CO$_2$ equivalent m^{-2}day^{-1}; Table 4).

Discussion

Despite its small size, Lake Jacques had a persistent layered structure over the summer, similar to deeper lakes, and stratification was strong. For example, buoyancy frequencies in the metalimnion were higher and more persistently higher than those reported during diel stratification in tropical lakes with high insolation (MacIntyre et al., 2002), higher than those observed in the pycnocline in a much larger meromictic lake (MacIntyre et al., 1999), and similar to values computed using data from a turbid subarctic thaw pond (Laurion et al., 2010). The stratification was weakened during the passage of cold fronts, with such events occurring more frequently and causing a greater weakening of the stratification in 2011 than in 2012. The variability in stratification moderated exchanges between the upper and lower water column, the extent of oxygenation of the lower water column, and the speciation of GHG.

The between-year differences in weather led to variability in storage and emissions of GHG. Frequent cold fronts associated with rain in 2011 resulted in accumulation of CO$_2$, but not CH$_4$, in the lower water column of the lake and higher effluxes of CO$_2$ at the surface than in the warmer dryer year. The strong thermal stratification in 2012 resulted in persistent anoxia starting early in the summer. However, increases of both CO$_2$ and CH$_4$ in the lake occurred later in summer 2012 in association with rainfall. As a result of the stratification, emissions were delayed until a large rainfall event weakened the stratification in mid-September. The approach taken here, which included time series data and gas storage assessment, similar to that in Aberg et al. (2010), enabled us to illustrate that increased CO$_2$ within the lake was associated with rainfall and how the between-year differences in meteorology moderated stratification, which in turn moderated concentrations and proportions of CO$_2$ and CH$_4$, and their evasion to the atmosphere.

Our comparison of CO$_2$ fluxes measured with chambers versus surface renewal and wind-based models was good, with comparisons within 30% between the chamber and surface renewal approach (Fig. 5). The comparisons with CH$_4$ were also good, with the largest discrepancy reaching a factor of two.

These comparisons differ from those in Schubert et al. (2012) who report greater than four-fold differences between floating chamber and modeled fluxes of CH_4. In our experiments, the flux measurements and water samples for the model calculations were obtained at the same location. In contrast, Schubert et al. (2012) studied a larger lake and deployed multiple chambers to obtain appropriate coverage. As indicated in the analysis in Heiskanen et al. (2014), the combination of internal wave motions and convective cooling can induce spatial variability in greenhouse gas concentrations. This variability should be taken into account for robust comparisons of measured and modeled results and for model development. The good agreement we demonstrate by working at one location indicates floating chambers and model approaches can obtain similar results and points to the utility of the approach for developing accurate models of the gas transfer coefficient.

Accuracy in predicting GHG emissions also requires understanding controls on temporal variability. Temporal variability in GHG fluxes depends on the size of water bodies (small > large, Roulet et al., 1997) and on sampling frequency (Weyhenmeyer, 1999). Besides our results showing that emissions varies between years in association with difference in ambient meteorology, they also show that they depend on the method used for estimating *k*. Models, such as the power law formulation of Crucius & Wanninkoff (2003), which force *k* to low values at low wind speeds, generate more variable GHG fluxes (max. CV = 100%). The surface renewal model approach gives less variable estimates. As it also includes the various processes mediating turbulence (Zappa et al., 2007; MacIntyre et al., 2010), it offers an approach for inclusion of a range of hydrodynamic controls on the gas transfer coefficient. In support of that model, we obtained better congruence with CO_2 fluxes from floating chamber and the surface renewal model than with the two wind-based models used in our comparison. Since the differences in gas transfer coefficient parameterizations can cause biases in predicting the contribution of lakes to the global carbon cycle (Rypdal & Winiwarter, 2001), model results should be evaluated against direct measurements particularly in changeable weather conditions.

During the rainy summer 2011, Lake Jacques was continuously a source of CO_2 to the atmosphere indicating the prevalence of respiration and net heterotrophy. The increased CO_2 concentrations lower in the water column tended to co-occur with rainfall, thus it may have resulted from direct inputs of CO_2 or labile organic carbon from the drainage basin which fueled bacterial respiration in the lake. The higher winds in that year caused more frequent water column mixing, enabling evasion and possibly more frequent re-suspension of organic matter from the sediments, which also may have stimulated bacterial respiration. In 2012, surface waters were undersaturated in CO_2 from the end of June until early August, concomitant with supersaturation in dissolved oxygen. This depression indicates a drawdown of CO_2 by photosynthesis combined with reduced vertical fluxes due to stable stratification separating the lower and upper water column. In 2012, if CH_4 flux was not taken into account, the lake would have been a net sink for atmospheric carbon. However, when CH_4 was included in the carbon budget, the lake became a source of carbon to the atmosphere except in July (GWP in Table 4).

Weather conditions also affected the ratios of CO_2 to CH_4 diffusing from the lake and the amount of CH_4 emitted via ebullition. High amounts of oxygen introduced to the water column during stormy weather led to lower concentrations of dissolved CH_4, likely resulting from enhanced methanotrophy and suppressed methanogenesis (Huttunen et al., 2006; Juutinen et al., 2009). When oxygen concentration was low, CH_4 accumulated in the lower water column. We did not determine whether CH_4 was derived from microbial processing of the CO_2 associated with rainfall events or directly from the sediments as in other studies (Rudd & Hamilton, 1978; Huttunen et al., 2003). Nevertheless, since the CH_4 only began to accumulate after rainfall, even though the lower water column was anoxic for most of 2012, it suggest that methanogenesis was stimulated by the microbial processing of the organic matter (Huttunen et al., 2003) or CO_2 (Wand et al., 2006) introduced with runoff. Moreover, stormy weather resulted in significantly higher CH_4 release from the lake via ebullition, possibly linked to warmer sediment temperatures (Wik et al., 2014) due to weaker stratification enabling a larger downward flux of heat. The higher frequency of frontal activity would have led to rapid changes in atmospheric pressure that also induces ebullition (Fechner-Levy & Hemond, 1996; Tokida et al., 2007). In Lake Jacques, ebullition was a more important CH_4 emission pathway than diffusion, as demonstrated in several recent studies (e.g., Delsontro et al., 2010; Schubert et al., 2012; Shakhova et al., 2014), but only during stormy weather. Interestingly, during the dry, hot summer, the volume of gas evading from the sediments was considerably lower, but it contained a higher proportion of CH_4. This inter-annual variability may have resulted from the difference in the size of bubbles, with smaller ones rising more often from the sediments during the more frequent mixing events in 2011. Relatively smaller bubbles would have lost relatively more of their initial CH_4 (Ostrovsky et al., 2008).

Weather patterns and oxygen availability may have also mediated the cycling of CH_4 by microbial processes. Significantly, more dissolved CH_4 was found in lake water after summer rainfall events when the lake was more strongly stratified. The stratification could have allowed a more efficient exploitation of this carbon source by methanotrophic bacteria and a greater input of CH_4-derived carbon to higher trophic levels (Bastviken et al., 2003; Sanseverino et al., 2012). In contrast, during stormy weather, most of the CH_4 was released from the sediments as gas bubbles avoiding oxidation in the water column and incorporation to the lake food web. Hence, CH_4 may be a larger carbon source for bacteria during hot and dry weather or in wind-sheltered lakes than during stormy weather or in wind-exposed lakes (Kankaala et al., 2013).

In summary, depending on weather conditions, small and shallow temperate lake can be polymictic or remain stably stratified for periods longer than a month. Our correlative data indicated that rainfall and associated runoff introduced carbon directly into the lake with inflows generally near the bottom. The duration and extent of stratification moderated both the vertical distribution and processing of CO_2 and CH_4, and their flux at the air–water interface. The frequency of frontal events and the magnitude of wind and rain associated with these fronts caused year-to-year variability in GHG emissions and storage. Climate induced modification of the frequency of weather events such as rainstorms and concomitant changes in thermal stratification thus can lead to different pathways of GHG cycling in shallow lakes.

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