



Modeling CO<sub>2</sub>  
diffusive emissions  
from lake systems

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# A coupled two-dimensional hydrodynamic and terrestrial input model to simulate CO<sub>2</sub> diffusive emissions from lake systems

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## Abstract

Most lakes worldwide are supersaturated with carbon dioxide (CO<sub>2</sub>) and consequently act as atmospheric net sources. Since CO<sub>2</sub> is a major greenhouse gas (GHG), the accurate estimation of CO<sub>2</sub> exchanges at air/water interfaces of aquatic ecosystems is vital in quantifying the carbon budget of aquatic ecosystems overall. To date, lacustrine CO<sub>2</sub> emissions are poorly understood, and lake carbon source proportions remain controversial, largely due to a lack of integration between aquatic and terrestrial ecosystems. In this paper a new process-based model (TRIPLEX-Aquatic) is introduced incorporating both terrestrial inputs and aquatic biogeochemical processes to estimate diffusive emissions of CO<sub>2</sub> from lake systems. The model was built from a two-dimensional hydrological and water quality model coupled with a new lacustrine CO<sub>2</sub> diffusive flux model. For calibration and validation purposes, two years of data collected in the field from two small boreal oligotrophic lakes located in Québec (Canada) were used to parameterize and test the model by comparing simulations with observations for both hydrodynamic and carbon process accuracy. Model simulations were accordant with field measurements in both calibration and verification. Consequently, the TRIPLEX-Aquatic model was used to estimate the annual mean CO<sub>2</sub> diffusive flux and predict terrestrial dissolved organic carbon (DOC) impacts on the CO<sub>2</sub> budget for both lakes. Results show a significant fraction of the CO<sub>2</sub> diffusive flux (~ 30–45 %) from lakes was primarily attributable to the input and mineralization of terrestrial DOC, which indicated terrestrial organic matter was the key player in the diffusive flux of CO<sub>2</sub> from oligotropical lake systems in Québec, Canada.

## 1 Introduction

Lakes account for more than 3% of land surface area (Downing et al., 2006) and are an important component in terrestrial carbon cycling. Substantial evidence indicates that the transfer of terrestrial carbon to lake ecosystems is considerably larger than

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the carbon flux to marine systems and approximately coequal to estimates of the net ecosystem productivity (NEP) of the terrestrial biosphere (Richey et al., 2002; Cole et al., 2007; Battin et al., 2009). In addition, a significant fraction of terrestrial carbon can be mineralized in lake systems (Kling et al., 1991; Cole et al., 1994, 2007; Hope et al., 1996; del Giorgio et al., 1997; Striegl et al., 2001; Algesten et al., 2003; Sobek et al., 2003; Rantakari and Kortelainen, 2005). Lake surveys carried out worldwide have demonstrated that boreal, temperate, and tropical lakes are typically supersaturated with CO<sub>2</sub> and consequently release significant amounts of CO<sub>2</sub> into the atmosphere (Kling et al., 1991; Cole et al., 1994, 2007; Sobek et al., 2003; Roehm et al., 2009; Battin et al., 2009).

The northern latitude biomes have been identified as important for CO<sub>2</sub> exchange between ecosystems and the atmosphere, with a net sink of CO<sub>2</sub> for temperate forests (Chapin III et al., 2000; Dunn et al., 2007). However, there are few quantitative estimates of lake emission in relation to current assessments of the CO<sub>2</sub> balance. To date, the lake CO<sub>2</sub> emissions over space are poorly understood (Duchemin et al., 2002; Sobek et al., 2003; Cardille et al., 2007; Demarty et al., 2011; Roehm et al., 2009; Tedoru et al., 2011), and lake carbon source proportions in different ecosystems remain controversial (del Giorgio et al., 1999; Cole et al., 2000; Jonsson et al., 2001, 2003; Prairie et al., 2002; Algesten et al., 2003; Hanson et al., 2003, 2004; Karlsson et al., 2007; McCallister and del Giorgio, 2008). Therefore, estimates of the fraction of terrestrial organic carbon that is exported to lakes and then routed into atmospheric CO<sub>2</sub> and the evaluation of the role of lakes in regional carbon budget require the integrated studies of the entire lake-watershed system (Algesten et al., 2003; Jenerette and Lal, 2005; Cole et al., 2007; Battin et al., 2009; Buffam et al., 2011).

Identifying CO<sub>2</sub> emissions from lakes is challenging and tends to be fraught with uncertainty since complex links exist between terrestrial and aquatic ecosystems (Hutjes et al., 1998; Wagener et al., 1998; Kalbitz et al., 2000; Smith et al., 2001; McDowell, 2003; Hanson et al., 2004; Jenerette and Lal, 2005; Cole et al., 2007; Buffam et al., 2011). In addition, water bodies exhibit significant multidimensional variations caused

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by interactions among hydrodynamic, biological, and chemical processes (Cole and Wells, 2006). Although lacustrine biogeochemistry is an integrative discipline, previous terrestrial and lake models have developed somewhat independently of each other (Grimm et al., 2003; Jenerette and Lal, 2005; Hanson et al., 2004; Cole et al., 2007; Cardille et al., 2007; Debele et al., 2008; Jones et al., 2009). Therefore, understanding the connectivity between each process and scaling up biogeochemical information must rely on coupled terrestrial and aquatic carbon cycle models essential in reducing uncertainty in carbon fluxes from and into lake systems (Grimm et al., 2003; Jenerette and Lal, 2005; Chapin III, 2006; Cole et al., 2007; Battin et al., 2009; Buffam et al., 2011).

In this paper a new process-based two-dimensional model (TRIPLEX-Aquatic) was developed to investigate lake carbon cycles with a particular emphasis on CO<sub>2</sub> diffusion. This model incorporates both terrestrial inputs and an aquatic carbon cycle model with exceptional spatial and temporal resolution. Thus, the TRIPLEX-Aquatic model constitutes an improved tool to investigate the primary processes involved in aquatic carbon cycling (including CO<sub>2</sub> diffusive exchanges between air and water bodies). Here, we seek to address two questions: (1) Is the TRIPLEX-Aquatic model able to capture the dynamics of CO<sub>2</sub> diffusive flux in boreal lakes? (2) What is the contribution of terrestrial DOC to lake CO<sub>2</sub> emission?

## 2 Model description and methods

To achieve the objectives of this study, TRIPLEX-Aquatic model need to capture the principal hydrological characteristics, the detailed carbon cycle accounting for inputs of DOC from the watershed in lake carbon processing, and the accurate CO<sub>2</sub> diffusive flux simulation to the atmosphere.

Figure 1 provides a schematic of the applied method based upon hydrological, carbon models, and CO<sub>2</sub> diffusive exchanges between air and water in the lake. The first model, hydrological processes start by representing hydrodynamic conditions. It

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is important in modeling carbon cycle since hydrology controls physical mixing processes between spatial components, factors that can directly or indirectly control biotic and abiotic processes. The second model, the lake carbon processes focus primarily on the prediction of organic/inorganic pools via photosynthesis and respiration, and their effects on dissolved oxygen and conventional cycles of nitrogen and phosphorus. This approach represents a substantial progression in lacustrine biogeochemical models since the 1970s (Harris, 1980; Beck, 1985; Ambrose et al., 1993; Kayombo et al., 2000; Chapelle et al., 2000; Omlin et al., 2001; Cole and Wells, 2006). In this paper, lake hydrodynamic and carbon simulations follow the approach of the CE-QUAL-W2 model (Cole and Wells, 2006) since the model has coupled between two-dimensional hydrodynamics and carbon cycle simulations with the same time steps and spatial grid, as well as it having already been successfully applied to rivers, lakes, reservoirs, and estuaries for several decades in the past. The CE-QUAL-W2 model is available at <http://www.ce.pdx.edu/w2>, and its program code is not changed in this study.

The third model, the simulation of CO<sub>2</sub> diffusive fluxes at the air/water interface uses a new boundary layer model developed by Cole and Caraco (1998) for lacustrine CO<sub>2</sub> diffusive flux, because this simulation in CE-QUAL-W2 model was simply designed the gas transfer coefficient for CO<sub>2</sub> is related to that of oxygen transfer using a factor of 0.923 (Cole and Wells, 2006). The program code of CO<sub>2</sub> diffusive flux submodel was developed using the Fortran language as in the CE-QUAL-W2 model.

The inputs of TRIPLEX-Aquatic model and file format are same as the CE-QUAL-W2 model, including climate data (e.g. air average temperature, dew point temperature, wind speed and direction, cloud cover), inflow and constituent concentrations (e.g. DOC, dissolved inorganic carbon – DIC, phosphate – PO<sub>4</sub><sup>3-</sup>, ammonium – NH<sub>4</sub><sup>+</sup>, nitrate – NO<sub>3</sub><sup>-</sup>, and dissolved oxygen – DO), and bathymetric and geometric data of lake. The model outputs represent the characteristics of hydrology (e.g. water velocity, density, temperature) and carbon processes (e.g. DOC, DIC, bicarbonates, carbonates, CO<sub>2</sub> concentration in water) in the lake, especially the CO<sub>2</sub> diffusive fluxes to the atmosphere. A brief overview of the TRIPLEX-Aquatic model is presented below.

## 2.1 The hydrodynamic submodel

The hydrodynamic simulation is able to characterize time variable longitudinal/vertical distributions of thermal energy in water bodies, based upon a finite difference solution applied to laterally averaged equations of fluid motion including momentum balance, continuity, constituent transport, free surface elevation, hydrostatic pressure, and equation of state (Cole and Wells, 2006) (see the Appendix A for a detail description of the model).

The model quantifies the free surface elevation, pressure, and density as well as the horizontal and vertical velocities (Cole and Wells, 2006). Explicit numerical schemes are also used to compute water velocities that affect the spatiotemporal distribution of temperature and biological/chemical constituents. The model simulates the average temperature for each model cell based upon water inflows/outflows, solar radiation, and surface heat exchanges. An equilibrium temperature approach was used to characterize the surface heat exchange. Spatial and temporal variations are permitted for longitudinal diffusion. The model computes the vertical diffusion coefficient from the vertical gradient of longitudinal velocities, water densities, and decay of surface wind shear. A full description of the model is offered by Cole and Wells (2006).

## 2.2 The carbon cycle submodel

The carbon submodel explicitly depicts organic and inorganic carbon processes in lake system. The organic carbon process includes four interacting systems: phytoplankton kinetics, nitrogen cycles, phosphorus cycles, and the dissolved oxygen balance (Fig. 2) (see the appendix A for a detail description of the model). The model accepts inputs in terms of different pools of organic matter (OM) and various species of algae. OM is partitioned into four pools according to a combination of its physical state (dissolved – DOM versus particulate – POM) and reactivity (labile – L versus refractory – R) characterizing the mineralization/decay rate of organic compounds. Labile OM (LDOM and LPOM) is more readily mineralized (i.e. having faster decay rates) whereas refractory

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OM (RDOM and RPOM) is less readily mineralized (i.e. having slower decay rates). All OM decay and decomposition processes in the model follow first order kinetics with temperature-dependent coefficients. The inorganic carbon processes include carbon dioxide input and output the inorganic carbon pool among carbonate species via two major pathways: atmospheric and biological exchange processes.

### 2.3 The CO<sub>2</sub> diffusive flux submodel

CO<sub>2</sub> diffusion across the air/water interface ( $F_{\text{CO}_2}$ ) is driven by the concentration gradient between the atmosphere and surface water and regulated by the gas exchange velocity  $K$ . Hence:

$$F_{\text{CO}_2} = K_{\text{CO}_2} (\Phi_{\text{CO}_2} - p\text{CO}_{2\text{atm}} K_{\text{H}}) \quad (1)$$

where  $K_{\text{CO}_2}$  is the piston velocity ( $\text{cm h}^{-1}$ );  $\Phi_{\text{CO}_2}$  is the CO<sub>2</sub> concentration in water ( $\text{g m}^{-3}$ ); and  $(p\text{CO}_{2\text{atm}} K_{\text{H}})$  is the CO<sub>2</sub> concentration in equilibrium with the atmosphere.  $p\text{CO}_{2\text{atm}}$  represents the CO<sub>2</sub> partial pressure in the atmosphere, and  $K_{\text{H}}$  is the Henry's constant corrected for water temperature.

$K_{\text{CO}_2}$  is the piston velocity constant for CO<sub>2</sub> calculated as follows:

$$K_{\text{CO}_2} = K_{600} \left( \frac{600}{\text{Sc}_{\text{CO}_2}} \right)^n \quad (2)$$

$n$ , the exponent, was used the value 0.5, which is appropriate for low-wind systems (Jahne et al., 1987).  $K_{600}$ , the piston velocity measured with SF<sub>6</sub> and normalized to a Schmidt number of 600, was determined according to the power function developed for low-wind speed conditions by Cole and Caraco (1998) where  $U_{10}$  is the wind speed ( $\text{m s}^{-1}$ ) at a height of 10 m:

$$K_{600} = 2.07 + 0.215U_{10}^{1.7} \quad (3)$$

$Sc_{CO_2}$ , representing the Schmidt number for carbon dioxide, is calculated according to Eq. (4) (Wanninkhof, 1992):

$$Sc_{CO_2} = 1911.1 - 118.11T_W + 3.4527T_W^2 - 0.04132T_W^3 \quad (4)$$

where  $T_W$  is the water surface temperature ( $^{\circ}C$ ).

### 3 Model input and test data

The computational grid of the two-dimensional lake model was developed based upon the bathymetric and geometric data collected from the unperturbed oligotrophic Lake Mary (46.26° N, 76.22° W) and Lake Jean (46.37° N, 76.35° W) in Québec, Canada, with a surface area of 0.58 and 1.88 km<sup>2</sup>, respectively. The watershed areas are 1.19 km<sup>2</sup> for Lake Mary and 5.43 km<sup>2</sup> for Lake Jean. The region has an average altitude of 230 m, and is characterized by an average temperature of approximately 5 °C, with 1000 mm of annual precipitations. Dominant tree species are red pine and yellow birch in mature. Soils are Brunisolic Luvisols. The lake areas were divided into 24 horizontal segments and 10 vertical layers. Longitudinal segments were 50 m in length for Lake Mary and 160 m in length for Lake Jean. The vertical layers were 2 m thick for both lakes (Fig. 3).

Time-varying boundary conditions at the surface of the lakes were set up with regard to meteorological influences. Hourly meteorological data, such as air average temperature, dew point temperature, wind speed and direction as well as cloud cover were obtained from weather monitoring stations located closet to the sites (Maniwaki Airport, Québec). Daily inflow and constituent concentrations of DOM at branch – estimated by the TRIPLEX-DOC model (Wu et al., 2013) in unit multiplying the watershed forest landscape areas of Lake Mary and Lake Jean, and adapted to TRIPLEX-Aquatic formats – were used as time-series inflow boundary conditions. Other inflow constituents – included POM, DIC, phosphate ( $PO_4^{3-}$ ), ammonium ( $NH_4^+$ ), nitrate ( $NO_3^-$ ), one species of

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blue-green algae, and DO – were compared to data from the nearby tributary in Eastern Canada with sampled data (Wang and Veizer, 2000; Hélie et al., 2002; Hélie and Hillaire-Marcel, 2006; Teodoru et al., 2009) because these have not been sampled in the present study.

Hydraulic parameters governing horizontal dispersion and bottom friction were set to default values using the Chezy friction model (Cole and Wells, 2006). Parameters affecting constituent kinetics are also required by the model. Initially, kinetic coefficients were set to default values (Cole and Wells, 2006) but subsequently tuned during the aquatic carbon process calibration so that the model output agreed with the field data. Kinetic coefficients were adjusted within acceptable ranges based upon data in published literature (Table 1). Although site-specific data are preferable, the paucity of details on hydraulic and kinetic coefficients in the lakes under study made it difficult to rely on site-specific data alone.

To test the model, four times campaigns were conducted in the two lakes from 2006 to 2007 because of the remote region, during periods following ice breakup in May 2006 (16 sampling in 6 days) and 2007 (15 sampling in 2 days), summer stratification in July 2006 (10 sampling in 2 days) and when fall overturn occurred in October 2006 (14 sampling in 3 days) for Lake Mary, and during periods in July 2006 (27 sampling in 2 days), October 2006 (1 sampling in 1 day), May 2007 (14 sampling in 1 day), July 2007 (20 sampling in 2 days) for Lake Jean. During each field trip, surface layer samples and information on water temperature, dissolved CO<sub>2</sub> concentrations (*p*CO<sub>2</sub>) as well as DOC at 15 cm depth was collected in pelagic sites of lake. An about 10 m depth profile of temperature, pH, DO and *p*CO<sub>2</sub> was also carried out at the central point of lake.

To determine *p*CO<sub>2</sub>, three 30 mL water samples were collected in 60 mL polypropylene syringes from each depth and carried out within 6 h of return to the field laboratory. They were equilibrated with an equal volume (30 mL) of ultrapure nitrogen (N<sub>2</sub>) by vigorous shaking for two minutes. Water was then flushed gently and the gaseous phase finally injected into the gas chromatograph (GC) (Star-3400CX; Varian, Palo Alto, CA,

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USA). Equilibrated CO<sub>2</sub> concentrations in the gaseous phase were calculated according to their solubility coefficients as a function of laboratory temperature (Flett et al., 1976). The CO<sub>2</sub> diffusive fluxes were therefore estimated from CO<sub>2</sub> saturation measured in the lakes in conjunction with wind speed. DOC concentration was analyzed in 0.2 μm filtered water samples in an OI-1010 Total Carbon Analyzer (OI Analytical, TX, USA) using wet persulfate oxidation. In addition, water temperature, DO, and pH profiles were taken with a YSI-6600 probe.

#### 4 Model calibration and validation

Calibration of the TRIPLEX-Aquatic model in middle segment (the center of lake) was carried out by tuning appropriate model parameters to match the predicted and measured data from Lake Mary in 2007 to obtain the best possible fit within acceptable ranges specified by Cole and Wells (2006) (Table 1). The model was verified against more data measured at Lake Mary in 2006 during which it was subjected to different ambient weather and flow conditions from those prevailing during model calibration in 2007, in order to test if the model was capable of accurately simulating the hydrodynamic regime and aquatic carbon dynamics under climatic conditions differing from those used for calibration. The model was also validated against measurements taken in Lake Jean from 2006 to 2007. System coefficients used in the model were the same as those determined during model calibration. Measurements serve to validate model results related to water temperature, pH, DO, pCO<sub>2</sub>, DOC, and the CO<sub>2</sub> diffusive flux.

##### 4.1 Temperature, pH, dissolved oxygen, and pCO<sub>2</sub>

Hydrodynamic calibration is typically performed by examining vertical and longitudinal concentration gradients of conservative constituents. Cole and Wells (2006) recommend the use of temperature gradients as a first step for hydrodynamic calibration. The prediction of surface water temperature for 2007 was in agreement with the mea-

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sured data from Lake Mary (Fig. 4a) despite high variability in the calibration data. The root mean squared error (RMSE) for the calibration period was 0.9 °C. The verification of surface layer water temperature during 2006 for Lake Mary and from 2006 to 2007 for Lake Jean (Fig. 4) shows sufficient agreement between the model simulations and field measurements. The water temperature RMSE was 1.6 °C during all simulation periods in both lakes.

With regard to the validation of the vertical simulation of lake hydrodynamics and carbon cycle, the water temperature, pH, DO, and  $p\text{CO}_2$  between model reconstructions and measurements were examined. Figure 5 shows that model simulation results with respect to depth were also accordant with the recorded observations: the RMSE were 0.37 °C for temperature, 0.24 for pH, 9.23 for DO (%), and 4.73 for  $p\text{CO}_2$  during fall turnover (Fig. 5a, c, e, g), and 1.36 °C for temperature, 0.39 for pH, 11.34 for DO, and 5.55 for  $p\text{CO}_2$  during spring stratification in Lake Mary (Fig. 5b, d, f, h). However, predicted values showed lower gradients than measured values during the spring period, especially for DO (Fig. 5f). The model also tended to underestimate water DO (%) by approximately 9 % for complete profile during fall turnover (Fig. 5e).

Differences between simulated and measured DO concentration, could partly be explained by lower tributary dissolved oxygen loads, because data was compared from the nearby tributary where may region-specific differences. For thermocline had lower gradients in predicted values than actual, because the stratification is a complex integration of multiple forcing components such as mixing rates, vertical dimensions of layer, layer temperature, basin morphometry, hydrology and, most important, meteorological conditions (Harleman, 1982; Owens and Effler, 1989), thus, it is difficult to accurately simulate the thermocline without intensive meteorological data, while the data used in this study are measured at only one nearby meteorological station. On the other hand, uncertainties resulting from the daily inflow of TRIPLEX-DOC model simulations in the upland watershed likely propagated into the simulations by way of the TRIPLEX-Aquatic model computation for waterbody, since outputs from the TRIPLEX-DOC model were used to run the TRIPLEX-Aquatic model.

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Although it is importance of water temperature and thermal stratification dynamics for temporal variation of surface water CO<sub>2</sub> in boreal lake (Aberg et al., 2010), the RMSE of surface temperature, pCO<sub>2</sub> in model simulation lead to approximately 12 % and 15 % mean errors in CO<sub>2</sub> diffusive flux respectively, they had only minor impacts on lake CO<sub>2</sub> emission. In general, these results suggest that the model reasonably represented surface and vertical variations of water temperature, pH, DO, pCO<sub>2</sub> and hydrodynamics of the lake system.

### 4.2 Dissolved organic carbon

Dissolved organic carbon, a substrate for microbial respiration, is a key constituent in aquatic carbon dynamics and could be the source of significant variations in lake pCO<sub>2</sub> (Hope et al., 1996; Sobek et al., 2003). Figure 6 offers a comparison between simulated and observed daily DOC concentrations from 2006 to 2007 in Lake Mary. Simulated values were reasonably distributed in the middle of the observational period (RMSE = 0.7). This agreement obtained during 2006 demonstrates that the model is capable of modeling DOC carbon-process properties within Lake Mary.

### 4.3 CO<sub>2</sub> diffusive flux

In this study, a zero CO<sub>2</sub> flux was assumed during the ice cover period for the simulations. During the ice-free period, there were considerable seasonal variations in the magnitude of the CO<sub>2</sub> diffusive flux and a distinct seasonal cycle in both Lake Mary and Lake Jean (Fig. 7). Peak fluxes occurred in the month of May following ice breakup and reached a brief, temporary minimum in early July. This minimum was followed by a second peak in late fall associated with autumnal mixing.

In comparing simulated results with observational daily data from 2006 for Lake Mary and from 2006 to 2007 for Lake Jean, the model successfully reproduced the observed distributions of CO<sub>2</sub> flux in both lakes, except for a daily value in autumn 2006 in Lake Jean that may be due to a single measurement. Although more systematic measure-

ments were absent in this study, such reasonable agreement between simulated and observed hydrodynamic plots and aquatic carbon dynamic parameters demonstrates that TRIPLEX-Aquatic was able to model various hydrodynamic and aquatic carbon cycle processes within the lake systems. It can thus be applied to simulate the CO<sub>2</sub> diffusive flux for lakes.

## 5 Terrestrial DOC and lake CO<sub>2</sub> emissions

### 5.1 Seasonal and annual mean lake CO<sub>2</sub> diffusive flux

The most current estimates of the annual CO<sub>2</sub> emission budgets of lakes, based upon measurements, only consider CO<sub>2</sub> produced during ice-free periods. However, CO<sub>2</sub> produced during winter months may accumulate under the ice cover and be subsequently released into the atmosphere once ice break-up occurs in spring (Striegl et al., 2001; Duchemin et al., 2006; Demarty et al., 2011). This early spring CO<sub>2</sub> release accumulated during the winter should be accounted for in order to develop a more realistic annual CO<sub>2</sub> emission budget for boreal lakes.

At the end of the winter season, the TRIPLEX-Aquatic model was well-calibrated to capture the principal characteristics of a high CO<sub>2</sub> flux episode just after ice melt over a period of approximately ten days (Fig. 7a, b). During this period the model estimated that approximately 80 % of the CO<sub>2</sub> contained in the water column of Lake Mary and Lake Jean was emitted into the atmosphere. The values for early spring CO<sub>2</sub> emissions ranged from 5 % to 8 % of the annual CO<sub>2</sub> diffusive emission budget for both lakes during the 2006 and 2007 period, which are thus an important portion in the annual CO<sub>2</sub> budget.

For Lake Mary and Lake Jean, variations in daily CO<sub>2</sub> flux were greatest during spring and fall and smallest during summer stratification (Fig. 7a, b). The average summer (from July to August) values were approximately 24–88 % lower than the average

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calculated values for the entire open water period in both lakes, a typical situation for northern temperate dimictic lakes (Hesslein et al., 1990).

Although there is a reasonable agreement between model simulations and field measurements for daily CO<sub>2</sub> diffusive flux (Fig. 7), when comparisons are based on seasonal CO<sub>2</sub> diffusive flux in Lake Mary (Fig. 8a), it was noted that the observations made during the autumn of 2006 were much higher than those in the simulation. For Lake Jean (Fig. 8b) measurements taken in the summer of 2006 were lower than those in the model simulations.

In respect to the annual mean CO<sub>2</sub> diffusion during the open water period in 2006, the simulated value was 447 mg CO<sub>2</sub> m<sup>-2</sup> day<sup>-1</sup> in Lake Mary, broadly consistent with the measured value of 493 mg CO<sub>2</sub> m<sup>-2</sup> day<sup>-1</sup>. In Lake Jean the simulated annual mean CO<sub>2</sub> emission was 589 mg CO<sub>2</sub> m<sup>-2</sup> day<sup>-1</sup>, significantly higher than the measurement of 360 mg CO<sub>2</sub> m<sup>-2</sup> day<sup>-1</sup> taken during the same period.

## 5.2 Impact of terrestrial DOC on lacustrine CO<sub>2</sub> diffusive emissions

A large body of literature suggests net heterotrophy is the key factor responsible for the often observed supersaturation of CO<sub>2</sub> in lake systems (del Giorgio et al., 1999; Cole et al., 2000; Jonsson et al., 2001, 2003; Prairie et al., 2002; Algesten et al., 2003; Hanson et al., 2003, 2004; Sobek et al., 2003; Karlsson et al., 2007; McCallister and del Giorgio, 2008), but this inference is tempered by uncertainties in the magnitude of the carbon load to lakes, and the relative contributions to lake CO<sub>2</sub> emission (Hanson et al., 2004; Karlsson et al., 2007; McCallister and del Giorgio, 2008).

To evaluate impacts of terrestrial DOC on the lake CO<sub>2</sub> emission regime, a comparison between DOC inputs and CO<sub>2</sub> fluxes was performed where the DOC data was simulated by way of the TRIPLEX-DOC model. Figure 9 shows a positive relationship between terrestrial DOC and CO<sub>2</sub> flux in both Lake Mary (CO<sub>2</sub> flux = 22.86 DOC + 3191, *R* = 0.65, *P* < 0.0001) and Lake Jean (CO<sub>2</sub> flux = 41.86 DOC + 3712,

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$R = 0.63$ ,  $P < 0.0001$ ), underlining the important role of DOC inputs in seasonal  $\text{CO}_2$  diffusive flux variations.

To further estimate the impact of terrestrial DOC on aquatic  $\text{CO}_2$  diffusive flux, a sensitivity analysis was carried out on the modeled results for 2006 to 2007 for both lakes by setting the terrestrial DOC inputs to zero while keeping other variable inputs at normal values, mimicking a situation in which the terrestrial DOC input would be nil. Results showed the annual mean  $\text{CO}_2$  diffusive flux from lakes under no-DOC-input conditions were much lower (approximately 30–45 % lower) than values with DOC inputs (Fig. 10a, b).

## 6 Discussion and conclusion

### 6.1 Comparison model with earlier approaches

There are presently only a handful of model studies (Hanson et al., 2004; Cardille et al., 2007; Buffam et al., 2011) that have tried to link terrestrial watershed carbon inputs to their aquatic components for  $\text{CO}_2$  emission. However, integration is still pending. In this study a comprehensive process-based aquatic carbon model (TRIPLEX-Aquatic) incorporating both terrestrial inputs, an aquatic carbon cycle, and detailed hydrodynamic simulation was developed and applied to investigate aquatic  $\text{CO}_2$  diffusion in lake ecosystems within Québec, Canada.

Although recent lake carbon models (Hanson et al., 2004; Cardille et al., 2007) integrate inputs of terrestrial DOC from watersheds, such models have no or very low hydrodynamic spatial resolution. In addition, these models do not include real-time meteorological conditions, while using constants to represent physical mixing processes between spatial components. The mass balance model (Jones et al., 2009) accounts for real-time meteorological data for lake carbon simulation, but does not include inputs of terrestrial DOC from catchments. Accordingly, the lake hydrodynamic routine is less realistic than the simulation carried out in this study. Moreover, previous photosynthetic

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estimates are based upon empirical models whereas simulations in this study were based on a process-based model.

A previous numerical CO<sub>2</sub> emission model developed by Barrette and Laprise (2002) illustrate the relevant approach to modeling physical processes in the water column based upon an extension of the lake water column model. It was used to study the temporal and spatial distribution of the dissolved CO<sub>2</sub> concentration profile and the CO<sub>2</sub> diffusive flux at the air/water interface. However, this particular model does not include the autotrophic and heterotrophic production of organic matter based upon variables such as water temperature, dissolved oxygen, nutrient salts, and terrestrial organic matter from catchments. All were included in the model used in the present study.

For the CO<sub>2</sub> diffusive flux submodel in TRIPLEX-Aquatic model, although a few studies have indicated that CO<sub>2</sub> diffusive fluxes obtained with the boundary layer technique might have been underestimated (Anderson et al., 1999; Jonsson et al., 2008) in comparison with the eddy covariance technique that is a direct measurement of the CO<sub>2</sub> flux, while the studies of Eugster et al. (2003) and Vesala et al. (2006) showed a good agreement. The boundary layer model of Cole and Caraco (1998) has also been validated and provides accurate or no bias estimations of CO<sub>2</sub> evasion over most of the sampling intervals based on the dry ice sowing experiment in a small boreal oligotrophic lake in Quebec, Canada (Soumis et al., 2008), and whole-lake sulfur hexafluoride (SF<sub>6</sub>) additions in temperate lakes near Land O'Lake, WI, USA (Cole et al., 2010) under a low-wind environment which is similar to the lakes in this study. Even though the Cole and Caraco (1998) model in this study is relatively simple, it is reasonable for estimating the CO<sub>2</sub> diffusive flux, partly because there has been little evidence that incorporation of comprehensive surface forcing provides a better flux field than simple wind speed algorithms (Wanninkhof et al., 2009).

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## 6.2 Impact of terrestrial DOC on CO<sub>2</sub> emission

Based on model validation, the agreement between observations and simulated results indicates the model is able to capture the principal hydrological characteristics and carbon dynamic processes in lake systems, it thus provides a realistic CO<sub>2</sub> diffusive flux simulation.

For the early spring CO<sub>2</sub> emissions, our model can successfully simulate the high CO<sub>2</sub> flux episode following ice breakup events. Such emission peaks were also identified by measurements in boreal lakes (Riera et al., 1999; Duchemin et al., 2006; Huotari et al., 2009; Demarty et al., 2011). Duchemin et al. (2006) estimated during the week following ice breakup 95 % of the dissolved CO<sub>2</sub> contained in the water column was released into the atmosphere. CO<sub>2</sub> emitted during this short period would account for 7–52 % of total annual emissions (Duchemin et al., 2006; Huotari et al., 2009; Demarty et al., 2011). Our results are within the lower end of their estimates, and reveal a significant CO<sub>2</sub> contribution during the ice break-up periods to the annual CO<sub>2</sub> budget of aquatic ecosystems in boreal lakes.

Concerning the seasonal and annual CO<sub>2</sub> emission, the differences between simulated and measured CO<sub>2</sub> diffusive flux values may result, in part, from the absence of systematic (or continuous) measurements of highly variable daily emissions: there are only a few daily observations for each season and these cannot accurately represent the natural CO<sub>2</sub> emission, thus resulting in a substantial overestimation or underestimation of seasonal, or annual flux values. On one hand, for the analysis of seasonal or annual variability, we should, in the future, use the eddy covariance measurements, which provide more frequent sampling and more accurate estimates of the CO<sub>2</sub> emission (Vesala et al., 2006; Jonsson et al., 2008; Huotari et al., 2011). On the other hand, it is our hope that the model simulation could contribute to the development of more effective sampling strategies, based on the characteristics of the simulated temporal CO<sub>2</sub> emission pattern associated with each lake.

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For the impact of terrestrial DOC on lake CO<sub>2</sub> emission, results from this study reveal that approximately 30–45 % of the annual CO<sub>2</sub> diffusive flux is due to terrestrial DOC input. Our study agree with the work ranged from 3 % to 70 % CO<sub>2</sub> flux from terrestrial organic carbon based on the  $\delta^{13}\text{C}$  measurement in lakes of southern Quebec, Canada (McCallister and del Giorgio, 2008), and consistent with the estimates between 30 % and 80 % of organic carbon exported from the watershed was released to the atmosphere CO<sub>2</sub> in lakes in the boreal Scandinavia (Algesten et al., 2003). The fate of terrestrial DOC in this study is also in the same range as the modeled estimates approximately 40–52 % respiration was supported by allochthonous DOC in Wisconsin lakes (Cole et al., 2002; Hanson et al., 2004). Our results thus support the hypothesis that a significant fraction of aquatic CO<sub>2</sub> diffusive flux is attributable to allochthonous organic carbon inputs from lake catchments (del Giorgio et al., 1999; Cole et al., 2000, 2007; Jonsson et al., 2001, 2003; Prairie et al., 2002; Algesten et al., 2003; Hanson et al., 2003, 2004; Karlsson et al., 2007; McCallister and del Giorgio, 2008; Battin et al., 2009; Buffam et al., 2011).

There is generally a net uptake of CO<sub>2</sub> from the atmosphere in boreal forests (Chapin III et al., 2000; Dunn et al., 2007), whereas, lake ecosystems seems to process a large amount of terrestrial derived primary production and alter the balance between carbon sequestration and CO<sub>2</sub> release. It demonstrates that lake ecosystems contribute significantly to regional carbon balances.

### 6.3 Future improvements to the TRIPLEX-Aquatic model

A major challenge for developing a new process-based model is the validation phase. Results presented in this study demonstrate that the TRIPLEX-Aquatic model is able to provide reasonable simulations of hydrodynamic and carbon processes in two selected boreal oligotrophic lakes (Lake Mary and Lake Jean). However, additional system verification and model testing should be conducted when applying the model to lakes with different characteristics in different climatic zones.

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In addition, the aquatic carbon approach is relatively simple in the current version of TRIPLEX-Aquatic. Decomposition processes of organic carbon follow first order kinetics of temperature-dependent coefficients for bacterial degradation. In fact, mineralization of allochthonous organic carbon occurs primarily, if not exclusively, by way of bacterial degradation (Jonsson et al., 2001). Photochemical degradation (Granéli et al., 1996) and its interaction with bacterial mineralization (Bertilsson and Tranvik, 1998) may add substantially to overall lake mineralization. Moreover, groundwater inflow (Kling et al., 1991; Striegl and Michmerhuizen, 1998) and surface water (Dillon and Molot, 1997; Cardille et al., 2007) enriched in inorganic carbon derived from weathering and soil respiration could be an important factor in some lakes.

There is also increasing evidences that gas transfer near the air/water interface cannot be adequately quantified using wind speed alone (Wanninkhof et al., 2009; MacIntyre et al., 2010). Studies have shown that other factors, such as friction velocity, bubbles, buoyancy, energy dissipation, fetch, surface slicks, rain, and chemical enhancement (Asher and Pankow, 1986; Wallace and Wirick, 1992; Erickson III, 1993; Ho et al., 2000; Zappa et al., 2001; McNeil and d'Asaro, 2007; Wanninkhof et al., 2009; MacIntyre et al., 2010), can also affect the gas transfer velocities. Disregarding these factors will undoubtedly add to the analytical uncertainty in relation to the aquatic carbon cycle. These shortcomings will be addressed and minimized in the future.

It is important to point out that the TRIPLEX-Aquatic model, incorporating robust process-based hydrodynamic components, could be feasibly adapted to reservoirs in the future in spite of the fact that their hydrodynamic and biogeochemical characteristics differ from those observed in lake systems. The model can also be coupled with land surface and ecosystem models at various horizontal resolutions or forced with GCM outputs to investigate the potential impact of climate and land use changes on lake carbon cycles. It is hoped that reassessment and future investigation will generate an improved and integrative understanding of carbon flux in lakes and reservoirs as well as a better integration between aquatic and terrestrial components.

### A1 The hydrodynamic submodel

The hydrodynamic simulation follows the approaches of CE-QUAL-W2 (Cole and Wells, 2006). The equations of fluid motion include: (1) momentum balance (Eq. A1); (2) continuity (Eq. A2); (3) constituent transport (Eq. A3); (4) free surface elevation (Eq. A4); (5) hydrostatic pressure (Eq. A5); and, (6) equation of state (Eq. A6)

$$\frac{\partial UB}{\partial t} + \frac{\partial UUB}{\partial x} + \frac{\partial WUB}{\partial z} = -\frac{1}{\rho} \frac{\partial BP}{\partial x} + \frac{\partial(BA_x \frac{\partial U}{\partial x})}{\partial x} + \frac{\partial B\tau_x}{\partial z} \quad (\text{A1})$$

$$\frac{\partial UB}{\partial x} + \frac{\partial WB}{\partial z} = qB \quad (\text{A2})$$

$$\frac{\partial B\Phi}{\partial t} + \frac{\partial UB\Phi}{\partial x} + \frac{\partial WB\Phi}{\partial z} - \frac{\partial(BD_x \frac{\partial \Phi}{\partial x})}{\partial x} - \frac{\partial(BD_z \frac{\partial \Phi}{\partial z})}{\partial z} = q_\Phi B + S_\Phi B \quad (\text{A3})$$

$$\frac{\partial B_\eta \eta}{\partial t} = \frac{\partial}{\partial z} \int_\eta^h UB dz - \int_\eta^h qB dz \quad (\text{A4})$$

$$\frac{\partial P}{\partial z} = \rho g \quad (\text{A5})$$

$$\rho = f(T_W, \Phi_{\text{TDS}}, \Phi_{\text{SS}}) \quad (\text{A6})$$

where  $U$  and  $W$  are the laterally averaged velocity components ( $\text{m s}^{-1}$ ) in each  $x$  and  $z$  direction;  $B$  is the water body width (m);  $t$  is the time (s);  $\rho$  is the water density ( $\text{kg m}^{-3}$ );  $P$  is the pressure ( $\text{N m}^{-2}$ );  $A_x$  is the momentum dispersion coefficient ( $\text{m}^2 \text{s}^{-1}$ );  $q$  is the lateral boundary inflow and outflow ( $\text{m}^3 \text{s}^{-1}$ );  $\Phi$  is the laterally averaged constituent

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concentration ( $\text{gm}^{-3}$ );  $D_x$  and  $D_z$  are the temperature and constituent dispersion coefficients in each  $x$  and  $z$  direction;  $q_\phi$  is the lateral boundary inflow and outflow mass flow rate of the constituent ( $\text{gm}^{-3}\text{s}^{-1}$ );  $S_\phi$  is the kinetics source/sink term for the constituent concentrations ( $\text{gm}^{-3}\text{s}^{-1}$ );  $B_\eta$  is the time and spatially varying surface width (m);  $\eta$  is the free water surface location (m);  $h$  is the total depth (m);  $g$  is the gravitational acceleration ( $\text{ms}^{-2}$ ); and  $f(T_W, \Phi_{\text{TDS}}, \Phi_{\text{SS}})$  is the density function dependent on water temperature ( $^\circ\text{C}$ ), total solids or salinity, and suspended solids.

## A2 The carbon cycle submodel

The organic carbon cycle includes phytoplankton kinetics, nitrogen cycles, phosphorus cycles, and the dissolved oxygen balance (Cole and Wells, 2006).

### A2.1 Phytoplankton

Photosynthesis is the primary ecosystem process and the basis of carbon dynamics in aquatic ecosystems. The phytoplankton growth rate ( $K_g$ , in  $\text{day}^{-1}$ ) is estimated as a function of temperature ( $T$ , in  $^\circ\text{C}$ ), light intensity ( $I$ , in  $\text{Wm}^2$ ), and concentrations of limiting nutrients ( $\text{gm}^{-3}$ ) such as nitrogen (N) and phosphorus (P).

$$K_g = K_g^* f(T) f(I) f(N, P) \quad (\text{A7})$$

where  $K_g^*$  is the maximum growth rate ( $\text{day}^{-1}$ ). An asymmetric function is used to simulate the influence of temperature on phytoplankton biological processes (Thornton and Lessem, 1978). This function depicts an optimum range of temperature (from  $T_2$  to  $T_3$ ) for biological process rates that decrease asymmetrically at lower ( $T_1$ ) and higher ( $T_4$ )

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temperatures.

$$f(T) = 0 \quad \text{if } T \leq T_1 \quad (\text{A8})$$

$$f(T) = \frac{K_1 e^{\gamma_r(T-T_1)}}{1 + K_1 e^{\gamma_r(T-T_1)} - K_1} \frac{K_4 e^{\gamma_f(T_4-T)}}{1 + K_4 e^{\gamma_f(T_4-T)} - K_4} \quad \text{if } T_1 < T < T_2 \quad (\text{A9})$$

$$f(T) = 0 \quad \text{if } T \geq T_4 \quad (\text{A10})$$

where

$$\gamma_r = \frac{1}{T_2 - T_1} \ln \frac{K_2(1 - K_1)}{K_1(1 - K_2)} \quad (\text{A11})$$

$$\gamma_f = \frac{1}{T_4 - T_3} \ln \frac{K_3(1 - K_4)}{K_4(1 - K_3)} \quad (\text{A12})$$

10 Compared to most other temperature functions with a single optimum value reported in existing literature (Zeng et al., 2006), this function combines two logistic equations to describe the rising ( $\gamma_r$ ) and falling ( $\gamma_f$ ) limb temperature multipliers.

The light function is described by Steels's (1965) according to the equation below:

$$f(I) = \frac{I}{I_s} e^{(1 - \frac{I}{I_s})} \quad (\text{A13})$$

15 where  $I_s$  is the saturating light intensity ( $\text{Wm}^{-2}$ ), and  $I$  is the available light intensity at depth  $z$  (m):

$$I = (1 - \beta)I_0 e^{-\alpha z} \quad (\text{A14})$$

20  $I_0$  is the light intensity at the water surface ( $\text{Wm}^{-2}$ );  $\beta$  is the fraction of solar radiation absorbed at the water surface; and  $\alpha$  is the light extinction coefficient, a combination of the extinction effects of water, inorganic and organic suspended solids, and algae.

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Nitrogen and phosphorus are the most important modeled limiting nutrients (Ambrose et al., 1993). Their concentrations are calculated based upon the Michaelis–Menten or Monod relationships that assume the nutrient composition of algal cells remains constant, and the external concentrations  $\Phi$  ( $\text{g m}^{-3}$ ) of available nutrients ( $i$ ) affect algal growth rates by a factor of:

$$f(P, N) = \frac{\Phi_i}{P_i + \Phi_i} \quad (\text{A15})$$

where  $P_i$  is the half-saturation constant for nutrient  $i$  ( $\text{g m}^{-3}$ ).

The rate equation for each algal group are:

$$\frac{\partial \Phi}{\partial t} = K_g \Phi - K_r \Phi - K_e \Phi - K_m \Phi - \omega \frac{\partial \Phi}{\partial z} \quad (\text{A16})$$

$$K_r = K_r^* f(T) \quad (\text{A17})$$

$$K_e = (1 - f(I)) f(T) K_e^* \quad (\text{A18})$$

$$K_m = f(T) K_m^* \quad (\text{A19})$$

where  $K_g$ ,  $K_r$ ,  $K_e$ ,  $K_m$ , and  $\omega$  are algal growth, dark respiration, excretion, mortality ( $\text{s}^{-1}$ ), and the settling rate ( $\text{m s}^{-1}$ ), respectively.  $\Phi$  is the algal concentration ( $\text{g m}^{-3}$ );  $z$  is the cell height (m);  $K_r^*$ ,  $K_e^*$ , and  $K_m^*$  are the maximum dark respiration, excretion, and mortality rate ( $\text{s}^{-1}$ ), respectively.

## A2.2 Nitrogen cycle

Three variables are used to characterize the nitrogen cycle: organic nitrogen, ammonium, and nitrate. As phytoplankton proliferates, dissolved inorganic nitrogen is taken up and incorporated into its biomass. As phytoplankton respire and dies, living organic matter is recycled into non-living organic and inorganic matter.

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The rate equations of nitrogen in labile and refractory dissolved organic matter are:

$$\frac{\partial \Phi_{\text{LDOM-N}}}{\partial t} = \sum K_e \delta_N \Phi_a + \sum (1 - P_m) K_m \delta_N \Phi_a - K_{\text{LDOM}} \gamma_{\text{OM}} \Phi_{\text{LDOM-N}} - K_{\text{L} \rightarrow \text{R}} \gamma_{\text{OM}} \Phi_{\text{LDOM-N}} \quad (\text{A20})$$

$$\frac{\partial \Phi_{\text{RDOM-N}}}{\partial t} = K_{\text{L} \rightarrow \text{R}} \Phi_{\text{LDOM-N}} - \gamma_{\text{OM}} K_{\text{RDOM}} \Phi_{\text{RDOM-N}} \quad (\text{A21})$$

- 5 where  $K_e$  and  $K_m$  are the algal excretion and mortality rates ( $\text{s}^{-1}$ ), and  $\delta_N$  is the algal stoichiometric coefficient for nitrogen;  $\Phi_a$ ,  $\Phi_{\text{LDOM-N}}$ , and  $\Phi_{\text{RDOM-N}}$  are algal, labile, and refractory DOM nitrogen concentrations ( $\text{gm}^{-3}$ ), respectively;  $P_m$  is the pattern coefficient for algal mortality, and  $K_{\text{LDOM}}$  and  $K_{\text{RDOM}}$  are the labile and refractory DOM decay rates ( $\text{s}^{-1}$ );  $\gamma_{\text{OM}}$  is the temperature rate multiplier for organic matter decay, and
- 10  $K_{\text{L} \rightarrow \text{R}}$  is the labile to refractory DOM transfer rate ( $\text{s}^{-1}$ ).

For nitrogen in labile and refractory particulate organic matter:

$$\frac{\partial \Phi_{\text{LPOM-N}}}{\partial t} = \sum P_m K_m \delta_N \Phi_a - K_{\text{LPOM}} \gamma_{\text{OM}} \Phi_{\text{LPOM-N}} - K_{\text{L} \rightarrow \text{R}} \Phi_{\text{LPOM-N}} - \omega_{\text{POM}} \frac{\partial \Phi_{\text{LPOM-N}}}{\partial z} \quad (\text{A22})$$

$$\frac{\partial \Phi_{\text{RPOM-N}}}{\partial t} = K_{\text{L} \rightarrow \text{R}} \Phi_{\text{LPOM-N}} - \gamma_{\text{OM}} K_{\text{RPOM}} \Phi_{\text{RPOM-N}} - \omega_{\text{POM}} \frac{\partial \Phi_{\text{RPOM-N}}}{\partial z} \quad (\text{A23})$$

- 15 where  $K_{\text{LPOM}}$  and  $K_{\text{RPOM}}$  are the labile and refractory POM decay rates ( $\text{s}^{-1}$ ), respectively;  $K_{\text{L} \rightarrow \text{R}}$  is the labile to refractory POM transfer rate ( $\text{s}^{-1}$ );  $\omega_{\text{POM}}$  is the POM settling rate ( $\text{ms}^{-1}$ ); and  $\Phi_{\text{LPOM-N}}$  and  $\Phi_{\text{RPOM-N}}$  are the labile and refractory POM nitrogen concentrations ( $\text{gm}^{-3}$ ), respectively.

For ammonium (NH<sub>4</sub>):

$$\frac{\partial \Phi_{\text{NH}_4}}{\partial t} = \sum K_r \delta_N \Phi_S - \sum K_g \delta_N \Phi_a P_{\text{NH}_4} + \sum_{i=1}^4 K_i \delta_{\text{NOM}} \gamma_{\text{OM}} \Phi_i + K_S \delta_{\text{NOM}} \gamma_{\text{OM}} \Phi_S \quad (\text{A24})$$

$$+ \sum K_{\text{CBOD}} R_{\text{CBOD}} \delta_{\text{N-CBOD}} \Theta^{T-20} \Phi_{\text{CBOD}} + K_{\text{NO}_x} \gamma_{\text{NO}_x} \Phi_{\text{NO}_x} - K_{\text{NH}_4} \gamma_{\text{NH}_4} \Phi_{\text{NH}_4}$$

where  $K_i$  ( $i = 1, 2, 3, 4$ ) are the labile and refractory DOM and the labile and refractory POM decay rates ( $\text{s}^{-1}$ ), respectively;  $K_S$ ,  $K_{\text{CBOD}}$ ,  $K_{\text{NO}_x}$ , and  $K_{\text{NH}_4}$  are the sediment, CBOD, nitrate-nitrogen, and ammonium decay rates ( $\text{s}^{-1}$ ), respectively;  $\delta_N$ ,  $\delta_{\text{NOM}}$ , and  $\delta_{\text{N-CBOD}}$  are the algal, organic matter, and CBOD stoichiometric coefficients for nitrogen, respectively;  $\Phi_S$ ,  $\Phi_a$ ,  $\Phi_{\text{CBOD}}$ ,  $\Phi_{\text{NO}_x}$ , and  $\Phi_{\text{NH}_4}$  are the organic sediment, algal, CBOD, nitrate-nitrogen, and ammonium concentrations ( $\text{g m}^{-3}$ ), respectively;  $\Phi_i$  ( $i = 1, 2, 3, 4$ ) are the labile and refractory DOM and the labile and refractory POM concentrations ( $\text{g m}^{-3}$ ), respectively;  $\gamma_{\text{OM}}$ ,  $\gamma_{\text{NO}_x}$ , and  $\gamma_{\text{NH}_4}$  are the temperature rate multipliers for organic matter decay, denitrification, and nitrification, respectively;  $\Theta$  is the temperature rate multiplier for CBOD decay; and  $P_{\text{NH}_4}$  is the ammonium preference factor expressed as:

$$P_{\text{NH}_4} = \Phi_{\text{NH}_4} \frac{\Phi_{\text{NO}_x}}{(K_{\text{NH}_4} + \Phi_{\text{NH}_4})(K_{\text{NH}_4} + \Phi_{\text{NO}_x})} + \Phi_{\text{NH}_4} \frac{K_{\text{NH}_4}}{(\Phi_{\text{NH}_4} + \Phi_{\text{NO}_x})(K_{\text{NH}_4} + \Phi_{\text{NO}_x})} \quad (\text{A25})$$

For nitrite-nitrate:

$$\frac{\partial \Phi_{\text{NO}_x}}{\partial t} = K_{\text{NH}_4} \gamma_{\text{NH}_4} \Phi_{\text{NH}_4} - K_{\text{NO}_x} \gamma_{\text{NO}_x} \Phi_{\text{NO}_x} - \omega_{\text{NO}_x} \frac{\partial \Phi_{\text{NO}_x}}{\partial z} - \sum K_g \delta_N \Phi_a (1 - P_{\text{NH}_4}) \quad (\text{A26})$$

where  $\omega_{\text{NO}_x}$  is the sediment transfer velocity ( $\text{m s}^{-1}$ ).

## A2.3 Phosphorus cycle

Phosphorus is an important element in aquatic ecosystems since it serves as one of the primary nutrients in phytoplankton growth (Carlson, 1977; Carpenter et al., 1998). Phosphorus is assumed to be completely available for phytoplankton as orthophosphate. It, however, becomes unavailable when it is removed through adsorption by way of suspended sediment particles and iron.

$$\frac{\partial \Phi_P}{\partial t} = \sum (K_r - K_g) \delta_P \Phi_a + \sum_{i=1}^4 K_i \delta_{POM} \gamma_{OM} \Phi_i + K_S \delta_{POM} \gamma_{OM} \Phi_S + \sum K_{CBOD} R_{CBOD} \delta_{P-CBOD} \Theta^{T-20} \Phi_{CBOD} - \frac{(\sum \omega_{ISS} \Phi_{ISS} + \omega_{Fe} \Phi_{Fe}) P_P}{\Delta z} \Phi_P \quad (A27)$$

where  $\delta_P$ ,  $\delta_{POM}$ , and  $\delta_{P-CBOD}$  are the algal, organic matter, and phosphorus/CBOD stoichiometric coefficients for phosphorus, respectively;  $\omega_{ISS}$ , and  $\omega_{Fe}$  are the inorganic suspended solids and particulate organic matter settling velocities ( $\text{m s}^{-1}$ );  $\Phi_{ISS}$ ,  $\Phi_{Fe}$ , and  $\Phi_P$  are the inorganic suspended solids, particulate organic matter, and phosphorous concentrations ( $\text{g m}^{-3}$ ), respectively; and  $\Delta z$  is the model cell thickness (m).

## A2.4 Dissolved oxygen balance

Oxygen is one of the most important elements in aquatic ecosystems. Dissolved oxygen (DO) concentrations depend upon exchanges with the atmosphere, consumption through algal respiration, production by algal photosynthesis, biochemical oxygen demand (BOD) decay, nitrification, and sediment organic matter decay.

$$\frac{\partial \Phi_{DO}}{\partial t} = \sum (K_g - K_r) \delta_{OM} \Phi_a + A_{sur} K_L (\Phi'_{DO} - \Phi_{DO}) - \sum_{i=1}^4 K_i \delta_{OM} \gamma_{OM} \Phi_i - K_S \delta_{OM} \gamma_{OM} \Phi_S - \sum K_{CBOD} R_{CBOD} \Theta^{T-20} \Phi_{CBOD} - K_{NH_4} \delta_{NH_4} \gamma_{NH_4} \Phi_{NH_4} \quad (A28)$$

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where  $A_{\text{sur}}$  is the water surface area ( $\text{m}^2$ );  $K_L$  is the interfacial exchange rate for oxygen ( $\text{ms}^{-1}$ ); and  $\Phi_{\text{DO}}$  and  $\Phi'_{\text{DO}}$  are the dissolved oxygen and saturation DO concentrations ( $\text{gm}^{-3}$ ).

## A2.5 pH and carbonates

- 5 Both pH and inorganic carbon species are calculated using water temperature, total dissolved solids (TDS) or salinity, alkalinity, and the total inorganic carbon concentration using basic inorganic carbon relationships. The computed inorganic carbon species are carbon dioxide, bicarbonates, and carbonates.

10 The concentration of total inorganic carbon ( $C_T$ ) is calculated according to the following equation:

$$\frac{\partial C_T}{\partial t} = \sum (K_r - K_g) \delta_C \Phi_a + \sum_{i=1}^4 K_i \delta_{\text{COM}} \gamma_{\text{OM}} \Phi_i + K_S \gamma_{\text{OM}} \delta_{\text{COM}} \Phi_S + A_{\text{sur}} F_{\text{CO}_2} + \sum K_{\text{CBOD}} R_{\text{CBOD}} \delta_{\text{C-CBOD}} \Theta^{T-20} \Phi_{\text{CBOD}} \quad (\text{A29})$$

15 where  $\delta_C$ ,  $\delta_{\text{COM}}$ , and  $\delta_{\text{C-CBOD}}$  are algal, organic matter, and the CBOD stoichiometric coefficient for carbon, respectively;  $A_{\text{sur}}$  is the surface area of the surface computational cell ( $\text{m}^2$ ); and  $F_{\text{CO}_2}$  is the  $\text{CO}_2$  interfacial diffusive flux ( $\text{gm}^{-2}$ ).

Inorganic carbon species are equated by:

$$[\text{H}^+] = -\frac{K_1}{2(1 - \frac{C_T}{A_C})} \left( 1 - \sqrt{1 - 4 \frac{K_2(1 - \frac{C_T}{A_C})}{K_1(1 - \frac{C_T}{A_C})^2}} \right) \quad (\text{A30})$$

$$[\text{H}_2\text{CO}_3] = \frac{C_T [\text{H}^+]^2}{[\text{H}^+]^2 + K_1 [\text{H}^+] + K_1 K_2} \quad (\text{A31})$$

$$[\text{HCO}_3^-] = \frac{C_T K_1 [\text{H}^+]^2}{[\text{H}^+]^2 + K_1 [\text{H}^+] + K_1 K_2} \quad (\text{A32})$$

$$[\text{CO}_3^{2-}] = \frac{C_T K_1 K_2 [\text{H}^+]^2}{[\text{H}^+]^2 + K_1 [\text{H}^+] + K_1 K_2} \quad (\text{A33})$$

where  $A_C$  is the carbonate alkalinity, and  $K_1$  and  $K_2$  are the dissociation constants for carbonic acid and bicarbonate, respectively.

$$[A_C] = [\text{HCO}_3^-] + 2[\text{CO}_3^{2-}] + [\text{OH}^-] - [\text{H}^+] \quad (\text{A34})$$

$$K_1 = \frac{[\text{H}^+][\text{HCO}_3^-]}{[\text{H}_2\text{CO}_3]} \quad (\text{A35})$$

$$K_2 = \frac{[\text{H}^+][\text{CO}_3^{2-}]}{[\text{HCO}_3^-]} \quad (\text{A36})$$

$$K_W = \frac{[\text{H}^+][\text{OH}^-]}{[\text{H}_2\text{O}]} \quad (\text{A37})$$

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**Table 1.** Final calibration values for hydrodynamic and ecological input variables.

Kinetic parameters	Units	Value	Suggested value (Cole and Well, 2006)
Horizontal eddy viscosity	$m^2 s^{-1}$	1.0	1.0
Horizontal eddy diffusivity	$m^2 s^{-1}$	1.0	1.0
Bottom frictional resistance	$m^2 s^{-1}$	70	70
Solar radiation fraction absorbed at the water surface	–	0.45	0.45
Solar radiation extinction – water	$m^{-1}$	0.45	0.25–0.45
Solar radiation extinction – detritus	$m^{-1}$	0.2	0.1
Solar radiation extinction – algae	$m^{-1}$	0.2	0.2
Wind sheltering coefficient	–	1.0	0–1.0
Zero-order sediment oxygen demand	$gm^{-2} day^{-1}$	1.0	0.1–1.0
Algal growth rate	$day^{-1}$	2.0	1.1–2.0
Algal dark respiration rate	$day^{-1}$	0.04	0.02–0.04
Algal excretion rate	$day^{-1}$	0.04	0.01–0.04
Algal mortality rate	$day^{-1}$	0.1	0.01–0.1
Algal settling rate	$day^{-1}$	0.1	0.1–0.14
Phosphorus half-saturation coefficient	$gm^{-3}$	0.003	0.003–0.009
Nitrogen half saturation coefficient	$gm^{-3}$	0.014	0.014, 0.03
Light saturation	$Wm^{-2}$	50	75
Lower temperature for minimum algal rates	$^{\circ}C$	5	5
Lower temperature for maximum algal rates	$^{\circ}C$	12	25
Upper temperature for maximum algal rates	$^{\circ}C$	20	35
Upper temperature for minimum algal rates	$^{\circ}C$	30	40
Lower temperature rate multiplier for minimum algal growth	–	0.1	0.1
Lower temperature rate multiplier for maximum algal growth	–	0.99	0.99
Upper temperature rate multiplier for maximum algal growth	–	0.99	0.99
Upper temperature rate multiplier for minimum algal growth	–	0.1	0.1
Phosphorus-to-biomass ratio	–	0.005	0.005
Nitrogen-to-biomass ratio	–	0.08	0.08
Carbon-to-biomass ratio	–	0.45	0.45
Algae-to-chlorophyll <i>a</i> ratio	–	130	145
Ammonium decay rate	$day^{-1}$	0.3	0.12
Sediment release rate of ammonium	fraction of SOD	0.001	0.001
Lower temperature for ammonium decay	$^{\circ}C$	5.0	5.0
Upper temperature for ammonium decay	$^{\circ}C$	25.0	25.0
Lower temperature rate multiplier for ammonium decay	–	0.1	0.1
Upper temperature rate multiplier for ammonium decay	–	0.99	0.99
Sediment release rate of phosphorus	fraction of SOD	0.001	0.001–0.003
Stoichiometric ratio of phosphorus in organic matter	–	0.005	0.005
Stoichiometric ratio of nitrogen in organic matter	–	0.08	0.08
Nitrate decay rate	$day^{-1}$	0.05	0.05–0.15
Lower temperature for nitrate decay	$^{\circ}C$	5.0	5.0
Upper temperature for nitrate decay	$^{\circ}C$	25.0	25.0
Lower temperature rate multiplier for nitrate decay	–	0.1	0.1
Upper temperature rate multiplier for nitrate decay	–	0.99	0.99

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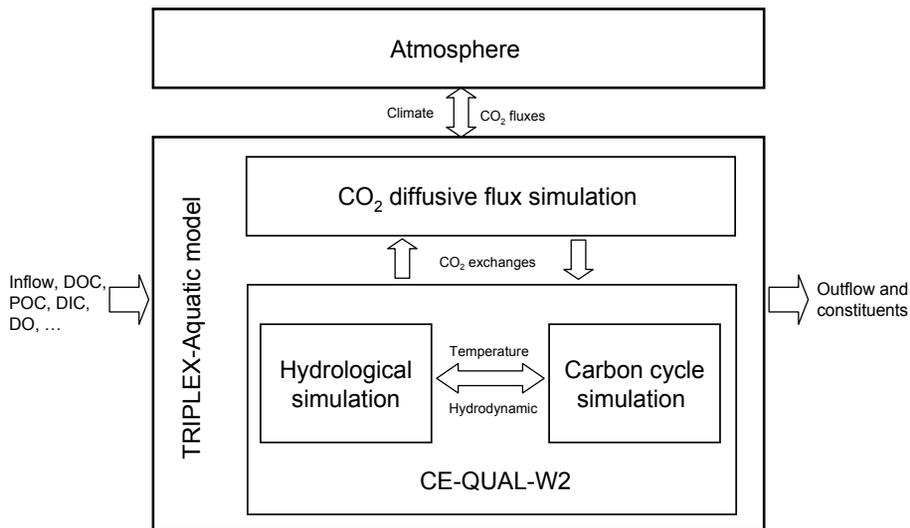
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**Fig. 1.** Modular structure of the TRIPLEX-Aquatic model. DOC: dissolved organic carbon, POC: particulate organic matter, DIC: dissolved inorganic carbon, and DO: dissolved oxygen.

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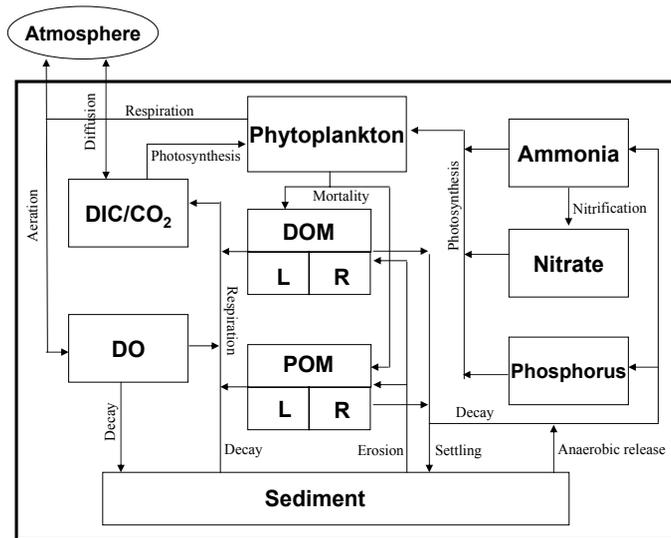
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**Fig. 2.** Flow diagram showing key pools and flux of carbon, nitrogen, and phosphorus simulation in the lake ecosystem. *T*: temperature, *P*: precipitation, DOM: dissolved organic matter, POM: particulate organic matter, L: labile, R: refractory, DO: dissolved oxygen, DIC: dissolved inorganic carbon.

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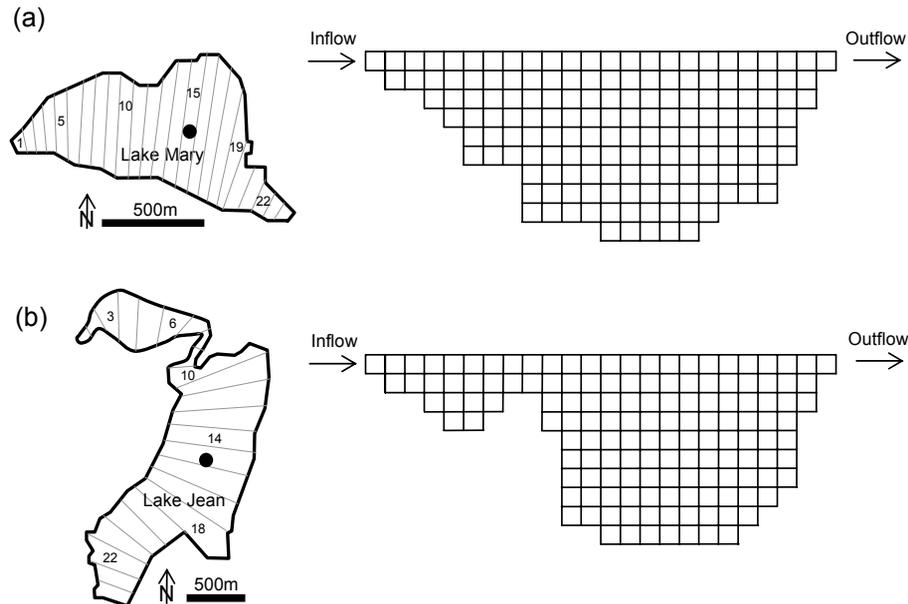
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**Fig. 3.** Map of the study lakes and the grid arrangement with model segment identification numbers for Lake Mary **(a)** and Lake Jean **(b)**. The dark circle is sample site.

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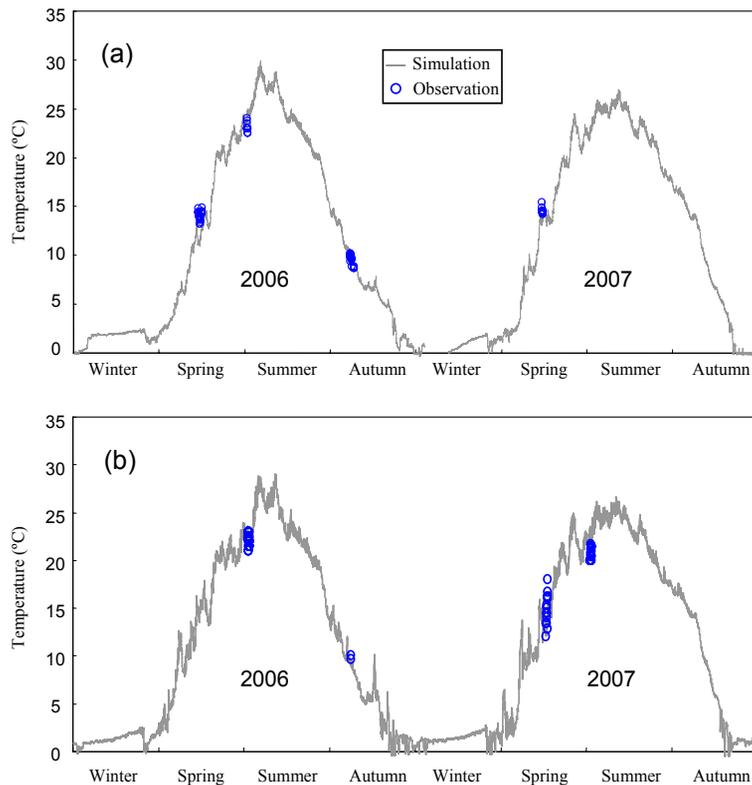
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**Fig. 4.** Observed versus predicted water surface temperatures from 2006 to 2007 for Lake Mary (a) and Lake Jean (b).

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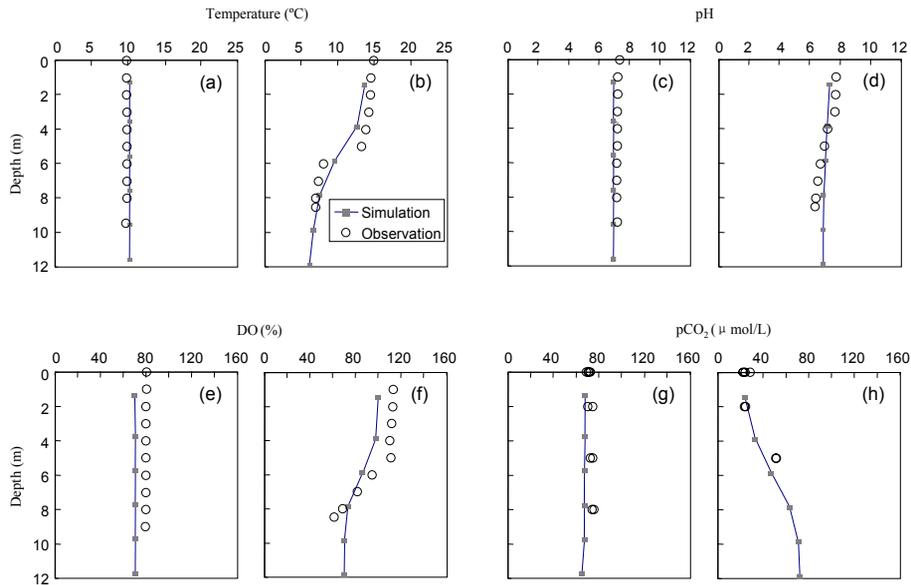
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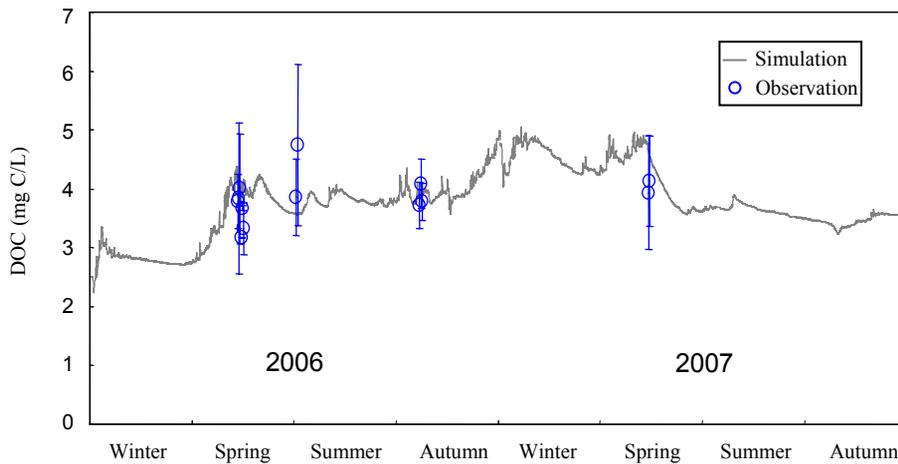


**Fig. 5.** Measured versus simulated vertical temperature, pH, dissolved oxygen (DO), dissolved CO<sub>2</sub> ( $p\text{CO}_2$ ) profiles in Lake Mary during autumn (21 October 2006) (a, c, e, g) and spring (14 May 2007) (b, d, f, h) periods.

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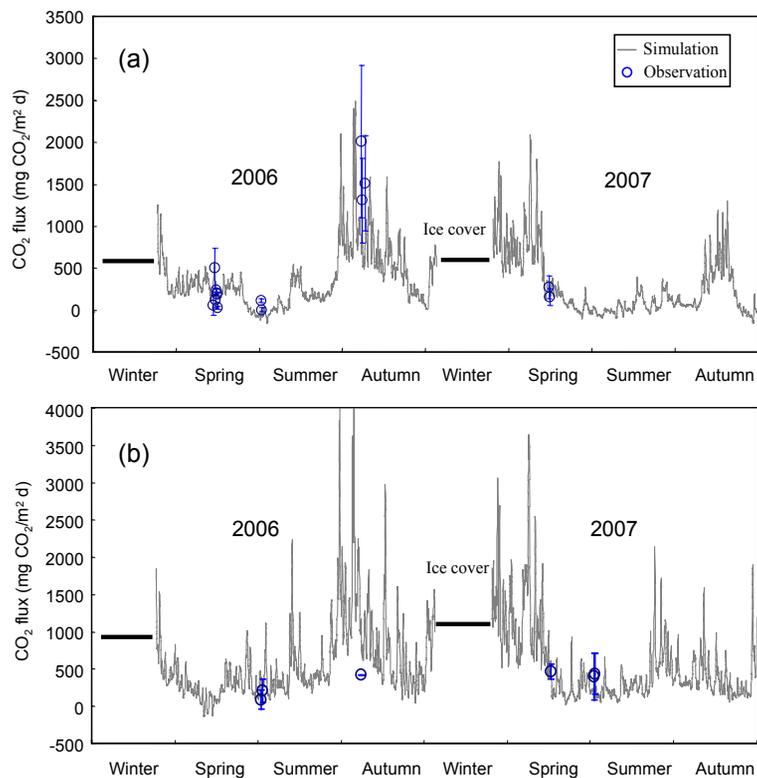


**Fig. 6.** Time series plots of measured versus simulated daily DOC concentrations in Lake Mary. Error bars represent standard deviations.

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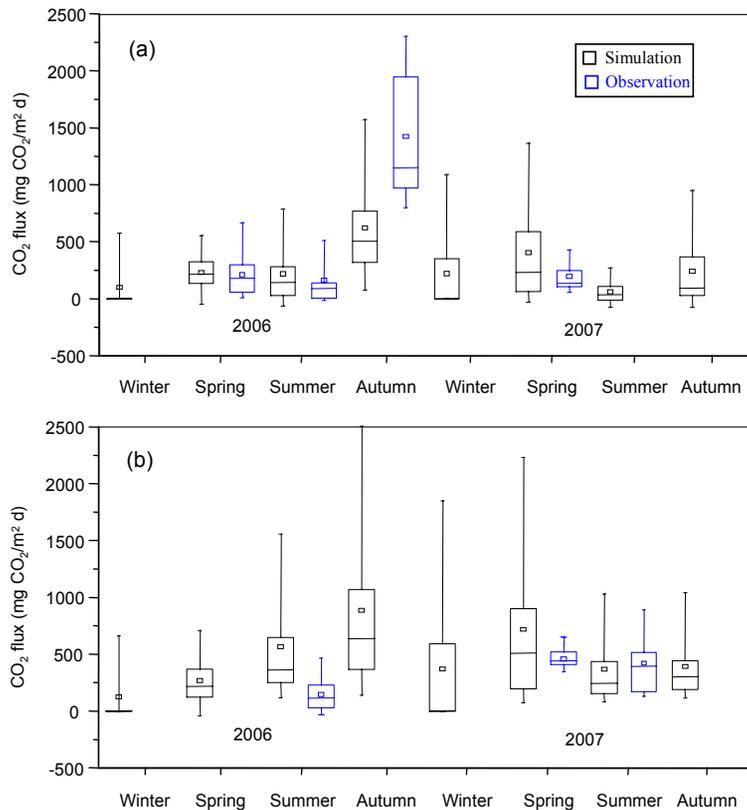
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**Fig. 7.** Time series plots of measured versus simulated daily CO<sub>2</sub> diffusive fluxes from Lake Mary **(a)** and Lake Jean **(b)**. Error bars represent standard deviations.

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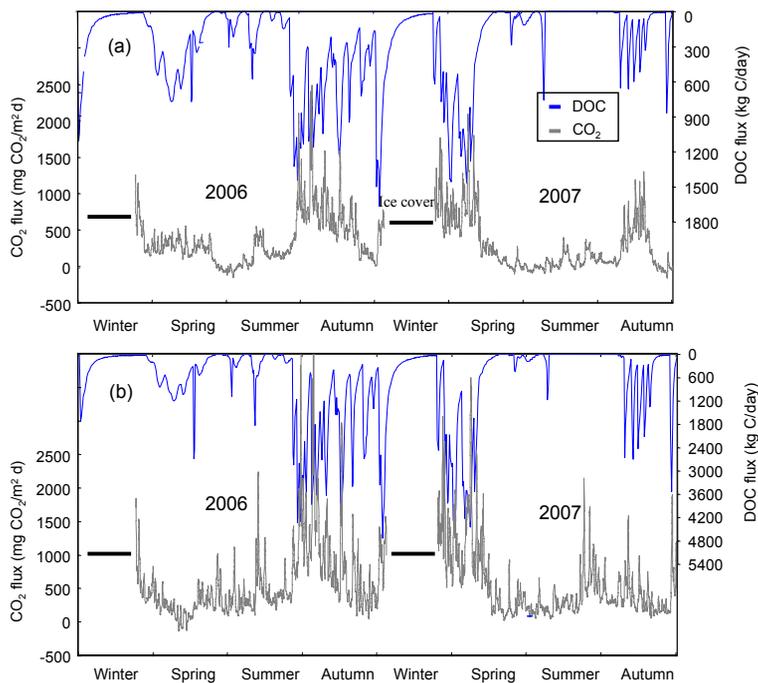


**Fig. 8.** Comparison between measurements and simulations of seasonal CO<sub>2</sub> diffusive fluxes for Lake Mary (a) and Lake Jean (b). Boxes indicate interquartile intervals (25th and 75th percentiles) while bars represent 90 % intervals (5th and 95th percentiles).

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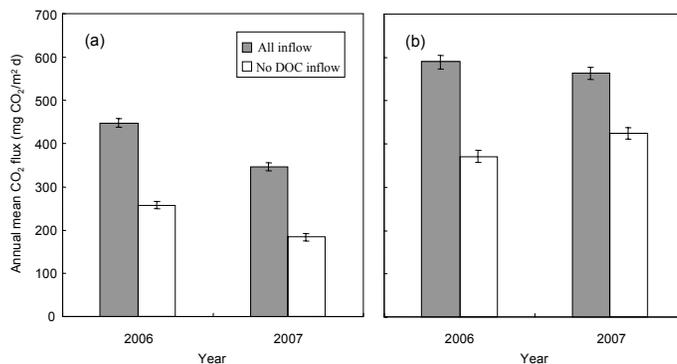


**Fig. 9.** Relationship between terrestrial DOC input and the CO<sub>2</sub> diffusive flux from Lake Mary (a) and Lake Jean (b).

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**Fig. 10.** Sensitivity analysis for the effect of terrestrial DOC inputs on the annual mean CO<sub>2</sub> diffusive flux from Lake Mary (a) and Lake Jean (b). Bars represent standard errors.

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