



# Modeling the interplay between deepwater oxygen dynamics and sediment diagenesis in a hard-water mesotrophic lake



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## ABSTRACT

Sediment diagenesis can be a significant driver of oxygen depletion in lakes and may dramatically impact the hypolimnetic oxygen concentrations. In this study, our aim is to simulate sediment oxygen demand (SOD) dynamics under varying conditions of organic matter sedimentation and hypolimnetic oxygen levels. Specifically, we use a process-based sediment diagenesis model to identify the critical processes that regulate dissolved oxygen levels in the hypolimnion of the mesotrophic Lake Simcoe, Ontario, Canada. We quantify the spatial distribution of organic matter mineralization and subsequently assess the role of sediment oxygen demand in hypolimnetic oxygen depletion. Our model reinforces the notion that aerobic mineralization is a major diagenetic process that shapes sediment oxygen demand in the system. Our model confirms existing empirical evidence that SOD contribution to the hypolimnetic oxygen deficit is less than 30% in Lake Simcoe. Our analysis also sheds light on the potential drivers of the significant spatial heterogeneity of the sediment oxygen demand among Kempenfelt Bay, Cook's Bay, and the main basin of Lake Simcoe, namely, the differences in primary production rates, the origins of the settling organic matter, the redistribution of sediments, and the oxygen concentration at the sediment–water interface due to differences in morphology and hydrodynamics. We conclude by arguing that the pace of the planned re-oligotrophication and the anticipated hypolimnetic oxygen improvements, induced by nutrient loading reductions, may experience short-term delays from years to several decades due to the potential effects of a number of feedback mechanisms across the sediment–water interface in Lake Simcoe.

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## 1. Introduction

The tight linkage between lake productivity and hypolimnetic dissolved oxygen (DO) concentration is well established in deep stratified lakes (Wetzel, 2001). Elevated lake productivity and organic matter (OM) sedimentation lead to anoxic conditions in deepwaters, which subsequently induce excessive release of nutrients from the sediments as well as accumulation of metals/toxic substances (e.g., methane, ammonium, and sulfide) near the lake bottom (Carignan and Lean, 1991; Gelda et al., 1995). Sediment oxygen demand (SOD) in lakes can be connected to both autochthonous and allochthonous OM loading rates and may significantly impact the rate of hypolimnetic DO depletion along with the oxygen penetration depth (OPD) in the sediments (Katsev et al., 2007; Matzinger et al., 2010; Müller et al., 2012). Besides the toxic impact of substances released from sediments on benthic organisms, the occurrence and extension of anoxic deepwater zones poses constraints on the integrity of fish habitats, especially those for cold-water species (Wu et al., 2003; Evans, 2007).

Because of the dire ramifications of hypoxia, lake management often revolves around the establishment of hypolimnetic DO threshold

concentrations in deepwaters, e.g., <2–4 mg L<sup>-1</sup>. Various strategies have been developed to meet such targeted hypolimnetic DO levels, including the reduction of external nutrient loading (Gächter and Wehrli, 1998), deepwater oxygenation (Liborius et al., 2009), aeration or mixing in winter (Mueller and Stadelmann, 2004), and removal of sediments containing high amount of organic matter (Annadotter et al., 1999). However, the effects of these restoration strategies are often delayed (Mueller and Stadelmann, 2004; Liborius et al., 2009). In other instances, decline in primary production/organic carbon sedimentation and ultimately hypoxia alleviation are only observed after a disproportional reduction of external nutrient loading (Charlton et al., 1993; Matthews and Efler, 2006). Importantly, SOD can drive hypolimnetic DO deficit for decades after lake re-oligotrophication, also known as the “sediment memory effect” (Gelda et al., 2012a, 2012b). The reason for the latter pattern is the slow degradation of accumulated OM from past periods of eutrophication coupled with the diffusion of degradation products from the sediments to the water column. For example, Carignan and Lean (1991) found that the degradation of the refractory portion of deposited OM to reduced substances could last from decades to centuries. Thus, detailed knowledge of the processes occurring within the top few centimeters of the sediment is essential for assessing water quality conditions, understanding the manifestation of hypoxia, and the management of surface waters.

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Diagenetic modeling is an indispensable tool to investigate the interplay among the sediment diagenesis processes, to generate hypotheses about the sediment–water column coupling, and to predict potential ecosystem behaviors. The demand for robust sediment diagenesis predictions is often underscored as a focal point when attempting to elucidate future lake hypoxia patterns and to gain insights into sediment functioning under projected changes in lake trophic status (Zhang and Arhonditsis, 2008). Nonetheless, this type of process-based diagenetic modeling as well as the data that necessitate to ground-truth those models (e.g., depth profiles of concentrations of dissolved and solid substances, phosphorus binding forms, organic matter depth profiles) are still missing in the context of water quality management (Smits and van Beek, 2013; Paraska et al., 2014). For example, existing sediment modeling studies have profoundly overlooked critical mechanisms, such as the diffusive boundary layer conditions (Brand et al., 2009) or the macrophyte subsidies to OM pool, while others lacked essential features in their configuration, such as flexible sediment depth profiles, dynamic behavior of forcing conditions in lake sediments, and lateral variability of sediment parameter vectors (see review by Paraska et al., 2014). In the same context, Kim et al. (2013) emphatically argued that field, experimental, and modeling work should be designed to shed light on the mechanisms of nutrient mobilization in the sediments and to identify process controls under a variety of conditions. The knowledge obtained from process-based diagenetic modeling will allow addressing research questions, such as the following: Can nutrient retention in lake sediments be predicted based on the sediment mineralogy, sedimentation substance inputs, catchment type, and other characteristics? How does sediment retention capacity with respect to nutrients respond to changes caused by human activities and/or climate change?

Recognizing the importance of the dynamic nature of diagenetic processes in lake sediments, the present study is founded upon a 1-D non-steady-state reaction-transport model for Lake Simcoe, Ontario, Canada (McCulloch et al., 2013). Lake Simcoe has experienced varying degrees of eutrophication problems since the establishment of the first European settlers in the 17th century (North, 2013). Increasing urbanization, intensive agricultural practices, atmospheric deposition, and internal and multiple external phosphorus loading have impacted the ecological health of the lake system (Gudimov et al., 2012). The depletion of hypolimnetic DO following eutrophication has been identified as a main reason for the recent collapse in cold-water fishery recruitment (Young et al., 2011). In this study, our intent is to quantify the contribution of the sediments to hypolimnetic DO depletion as well as to investigate the impact of OM loading on seasonal SOD dynamics and sediment diagenesis processes in three basins of Lake Simcoe. We also explicitly accommodate the lateral heterogeneity of the processes associated with the oxygen demand, thereby challenging the validity of model parameterizations that postulate spatial homogeneity. Finally, we conduct local sensitivity analysis to identify critical processes at the sediment–water interface or near-bottom conditions that may shape O<sub>2</sub> concentration profiles in sediments.

## 2. Methods

### 2.1. Study site and experimental data

Lake Simcoe is located 44 km north of Toronto with 11.6 km<sup>3</sup> of water volume and a catchment area of 2,840 km<sup>2</sup> (Fig. 1a). It is a dimictic lake that completely freezes over during most winters. In its current mesotrophic state, Lake Simcoe receives wastewater from fourteen municipal wastewater treatment plants, which constitute sources of phosphorus (P) loading (6 ± 1 tonnes year<sup>-1</sup> between 2004 and 2007) and substantial phosphorus loads are also deposited from the atmosphere (18 ± 4 tonnes year<sup>-1</sup>) or emanate from other non-point sources, including runoff from agricultural, urban and natural areas (43 ± 5 tonnes year<sup>-1</sup>), and rural septic systems (4.4 ± 0.1 tonnes

year<sup>-1</sup>) (Table 1; see also Gudimov et al., 2012). Lake Simcoe consists of a large main basin (mean depth 14 m, maximum depth 33 m) and two large bays: the narrow and deep Kempenfelt Bay on the west side of the lake (area 34 km<sup>2</sup>, mean depth 20 m) and the shallow Cook's Bay at the south end of the lake (area 44 km<sup>2</sup>, mean depth 13 m). Cook's Bay is connected to Holland Marsh, an agricultural cluster of artificially reclaimed land, rich with organic matter, through dikes and drainage canals, while the Kempenfelt Bay subwatershed is the location of the city of Barrie, where a population of about 135,000 inhabitants resides.

The sediment data set for model calibration was collected in spring and autumn of 2011 from three basins of the lake, i.e., sites K42, K45, and C9 (Fig. 1a; see detailed description in Dittrich et al., 2013). In short, sediment samples of 60 cm length were collected using a core sampler. Microsensor measurements for O<sub>2</sub> and pH were carried out immediately upon arrival to the laboratory. Two cores were used for pore water analysis, and two to three cores were used for the fractionation of phosphorus, porosity analysis, dry weight, and total organic matter. The historical DO profiles in the water column (Fig. 1b) were provided by the Ontario Ministry of the Environment (personal communication Dr. Hamdi Jarjanazi). Historical sedimentation fluxes follow the Hiriart-Baer et al. (2011) dating results. The lake hypolimnetic area during the summer-stratified period refers to layers of the water column below 18 m depth (Young et al., 2011). A P sequential extraction analysis quantified the P pools in the sediments, including mobile pool, which suggested a distinct heterogeneous pattern of the P-binding forms in Lake Simcoe (Dittrich et al., 2013). In Cook's Bay, the predominant fraction of total phosphorus (TP) is carbonate-bound P (apatite-P) mainly due to the accelerated erosion in the catchment. TP content in the sediments of Cook's Bay is the lowest among the three studied basins in Lake Simcoe, providing evidence that the high sedimentation rates and natural watershed sources may lead to a “dilution” of P in the sediment dry matter. In contrast, the hypolimnetic sediments in Kempenfelt Bay are responsible for high diffusive P fluxes into the water column, presumably reflecting the highest proportion of the redox-sensitive P sediment pool compared to other lake segments as well as the occasional hypoxic conditions in the Kempenfelt Bay hypolimnion (Eimers et al., 2005). The sediments in the main basin are mostly driven by fast diagenetic processes of settling organic matter from lake epilimnion, which may lead to internal P loading greater than 9 tonnes P year<sup>-1</sup> (Gudimov et al., 2015). Further details regarding the sampling practices and analytical protocols can be found in Dittrich et al. (2013).

### 2.2. Model description and implementation

The diagenetic model for Lake Simcoe was developed using AQUASIM software in 1-D reactive-transport sediment compartment for solid and dissolved substances (Reichert, 1998; Dittrich et al., 2009). The model accounted for deposition fluxes of particulate matter, sediment compaction, bioturbation/bioirrigation, solute molecular diffusion, primary and secondary redox reactions, mineral precipitation/dissolution and acid dissociation reactions (McCulloch et al., 2013; see also Table S1). The sediment model is based on mass-conservation diagenetic equations for solid and dissolved substances (Bernier, 1980):

$$\frac{\partial(\theta S_i)}{\partial t} = \frac{\partial}{\partial z} \left( D_b \frac{\partial(\theta S_i)}{\partial z} + \theta D_{S_i} \frac{\partial S_i}{\partial z} \right) + r_{S_i} - \alpha_{\text{bioirrig}} \theta (S_i - S_i^{\text{SWI}})$$

$$\frac{\partial(X_i)}{\partial t} = \frac{\partial(v_{\text{sed}} X_i)}{\partial z} + \frac{\partial}{\partial z} \left( D_b \frac{\partial(X_i)}{\partial z} \right) + r_{X_i}$$

where  $X_i$  and  $S_i$  represent solid and dissolved phase species,  $\theta$  is the porosity,  $z$  is the vertical dimension of the sediment core,  $t$  is time,  $r_{S_i}$  and  $r_{X_i}$  are the biogeochemical transformation rates and  $v_{\text{sed}}$  is the velocity of sediment vertical movement,  $D_{S_i}$  is the solute molecular diffusion,  $D_b$  is the bioturbation coefficient,  $\alpha_{\text{bioirrig}}$  is the bioirrigation coefficient,  $S_i^{\text{SWI}}$  is

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