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Tracking the carbon footprint of lakes: dynamic modeling of organic carbon fates in lake ecosystems

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Abstract:	Lakes are active processors of organic carbon (OC) and play important roles in landscape and global carbon cycling. Allochthonous OC loads from the landscape, along with internal OC autochthonous loads from primary production, are mineralized in lakes, buried in lake sediments, and exported via surface or groundwater outflows. Although these processes provide a basis for a conceptual understanding of lake OC budgets, few studies have integrated these fluxes under a dynamic modeling framework to examine their interactions and relative magnitudes. We developed a simple, dynamic mass balance model for OC, and applied the model to a set of five lakes. We examined the relative magnitudes of OC fluxes and found that long-term (> 10 year) lake OC dynamics were predominantly driven by allochthonous loads in four of the five lakes, underscoring the importance of terrestrially-derived OC in northern lake ecosystems. Respiration, the primary mechanism by which lakes convert OC to inorganic carbon, ranged from 14-90% of total OC loads (allochthony + autochthony). In contrast, burial, the main OC storage mechanism in lakes, represented a relatively small component of total OC loads (3-37%). Our model highlighted seasonal patterns in lake OC budgets, with increasing water temperatures and lake productivity throughout the growing season corresponding to a transition from burial- to respiration-dominated OC fates. Ratios of respiration to burial, however, were also mediated by the source (autochthony vs. allochthony) of total OC loads. Autochthonous OC is more readily respired and may therefore proportionally reduce burial under a warming climate, but allochthonous OC may increase burial due to changes in precipitation. Interactions between ratios of autochthony vs. allochthony and respiration vs. burial demonstrate the importance of dynamic models for examining both the seasonal and inter-annual roles of lakes in landscape and global carbon cycling, particularly in a global change context. Finally, we highlighted critical data needs, which include surface water DOC observations in paired tributary and lake systems, measurements of OC burial rates, and budgets of particulate OC.

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1 Running head: Organic carbon fates in lakes

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4 ecosystems

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39

40 **ABSTRACT**

41   Lakes are active processors of organic carbon (OC) and play important roles in landscape

42 and global carbon cycling. Allochthonous OC loads from the landscape, along with internal OC

43 autochthonous loads from primary production, are mineralized in lakes, buried in lake sediments,

44 and exported via surface or groundwater outflows. Although these processes provide a basis for a

45 conceptual understanding of lake OC budgets, few studies have integrated these fluxes under a

46 dynamic modeling framework to examine their interactions and relative magnitudes. We

47 developed a simple, dynamic mass balance model for OC, and applied the model to a set of five

48 lakes. We examined the relative magnitudes of OC fluxes and found that long-term (> 10 year)

49 lake OC dynamics were predominantly driven by allochthonous loads in four of the five lakes,

50 underscoring the importance of terrestrially-derived OC in northern lake ecosystems.

51 Respiration, the primary mechanism by which lakes convert OC to inorganic carbon, ranged

52 from 14-90% of total OC loads (allochthony + autochthony). In contrast, burial, the main OC

53 storage mechanism in lakes, represented a relatively small component of total OC loads (3-37%).

54 Our model highlighted seasonal patterns in lake OC budgets, with increasing water temperatures

55 and lake productivity throughout the growing season corresponding to a transition from burial- to

56 respiration-dominated OC fates. Ratios of respiration to burial, however, were also mediated by

57 the source (autochthony vs. allochthony) of total OC loads. Autochthonous OC is more readily

58 respired and may therefore proportionally reduce burial under a warming climate, but

59 allochthonous OC may increase burial due to changes in precipitation. Interactions between

60 ratios of autochthony vs. allochthony and respiration vs. burial demonstrate the importance of

61 dynamic models for examining both the seasonal and inter-annual roles of lakes in landscape and

62 global carbon cycling, particularly in a global change context. Finally, we highlighted critical

63   data needs, which include surface water DOC observations in paired tributary and lake systems,

64   measurements of OC burial rates, and budgets of particulate OC.

65

66   Key words: carbon cycle, carbon flux, limnology, mass balance, allochthony, autochthony,

67   sedimentation, dissolved organic carbon, particulate organic carbon, LTER, GLEON

5

68   **INTRODUCTION**

69   Lakes are dynamic components of the landscape that actively process, store, and transport

70   terrestrially derived organic carbon (OC) (Cole et al. 2007, Tranvik et al. 2009, Tanentzap et al.

71   2017), as well as emit inorganic carbon to the atmosphere (Arvola et al. 2002, Raymond et al.,

72   2013, Weyhenmeyer et al. 2015), making them important in global carbon (C) cycling. Owing to

73   a dearth of ecosystem-scale studies that fully account for OC budgets (Hanson et al. 2014, 2015),

74   there remains a considerable knowledge gap in lake OC dynamics, and thus in fully

75   understanding the role of lakes in the global C cycle. Global estimates of CO_2 emissions (i.e.,

76   evasion) from lakes and reservoirs are 0.32 Pg (petagrams) C yr^{-1} (Raymond et al. 2013),

77   whereas 0.02-0.07 Pg C yr^{-1} are stored in sediments (Tranvik et al. 2009). These estimates,

78   however, are highly uncertain, and models that fully account for major OC fluxes and storage

79   terms in lakes and that explore uncertainties around those terms are needed to advance our

80   understanding of lake OC cycling and their contribution to global C budgets (Hanson et al.

81   2015). Existing mass balance models are generally based on low spatio-temporal frequency data,

82   confined to single lakes, and tend to be concentrated in boreal regions (Jonsson et al. 2001,

83   Urban et al. 2005, Andersson and Sobek 2006, Cremona et al. 2014). In a first step in

84   overcoming some of these limitations, here we developed and applied a dynamic mass balance

85   model to examine the relative magnitudes of OC fluxes across a set of five lakes with whole

86   ecosystem OC budget data.

87

88   *Overview of concepts of key OC fluxes in lake ecosystems*

89   For lakes, the term "mass balance" has been broadly used to quantify carbon or nutrient

90   budgets as the combination of inputs, outputs, and changes to standing stocks in the water

91   column and sediments (Pace and Lovett 2013). Inputs to lake ecosystem OC budgets are the sum

92   of allochthonous (externally derived) dissolved (DOC) and particulate OC (POC) inflows from

93   surface and groundwater sources, atmospheric deposition via precipitation, dry deposits, and

94   litterfall, and autochthonous (internally derived) DOC and POC from microbial exudates

95   (Kawasaki and Benner 2006) and primary production. Outputs from the OC pool include

96   mechanisms that mineralize (i.e., photo-oxidation and respiration) and export OC via surface and

97   groundwater outflows. Here, for simplicity, all mineralization processes that convert OC to CO_2

98   are collectively modeled as respiration. The mass change in OC in the water column and lake

99   sediments is considered as change in storage. Outputs and storage are the fates of OC loads, and

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Box 1. Mass balance conceptual equations for organic carbon (OC) in lake ecosystems

$OC_{ALLOCHTHONOUS}$: surface and groundwater inflows + litterfall + atmospheric deposition

$OC_{AUTOCHTHONOUS}$: gross primary production - autotrophic respiration

Full budget: $OC_{ALLOCHTHONOUS}$ + $OC_{AUTOCHTHONOUS}$ = respiration + burial + export + ΔOC (in water column)

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100   their balances define the role of lakes in broader C cycling (Box 1, Fig. 1).

101

102   At the global scale, lakes are thought to be net sources of C to the atmosphere based on

103   the super-saturation of CO_2 (Kortelainen et al. 2006, Tranvik et al. 2009, Raymond et al. 2013)

104   and methane (Bastviken et al. 2011) in lake surface waters. OC export is less frequently

105   considered, but equally important, both in terms of burial and the quality and quantity of OC

106   ultimately reaching the ocean via tributaries (Raymond and Bauer 2001, Santoso et al. 2017).

107   Because lakes store OC in sediments, they can also act as sinks in the global C cycle

108   (Mulholland and Elwood 1982, Dillon and Molot 1997, Einsele et al. 2001, Einola et al. 2011).

109   We synthesized existing knowledge of lake OC budgets into a model that integrates these

110 important mechanisms, including both in-lake as well as external (i.e., watershed) processes (Fig.

111 1). Below we described these processes in three main categories 1) allochthony, 2) autochthony,

112 and 3) storage and export.

113

114 *Allochthony*

115   Allochthonous inputs include all externally derived OC, including terrestrial DOC and

116 POC from surface and groundwater inflows, litterfall, and direct-fall precipitation (Box 1).

117 Although surface water inflows regularly deliver DOC to lake ecosystems, the uncertainties

118 around their sources and magnitudes are perhaps the most commonly overlooked aspect in OC

119 budgets, largely owing to data limitations (Hanson et al. 2015). Prior studies have included direct

120 measurements of inflow stream concentrations of DOC when available (Schindler et al. 1997,

121 Jonsson et al. 2001, Urban et al. 2005, Klump et al. 2009), but other approaches have included

122 literature-derived input estimates (Striegl et al. 1998), equations based on watershed area (Sobek

123 et al. 2006), watershed-wetland area ratios (O'Connor et al. 2009), precipitation (Hanson et al.

124 2004, Staehr et al. 2010), or GIS-based estimates based on land cover and distance-weighted

125 hydrological flow paths (Canham et al. 2004). In lakes without surface inflows, including closed-

126 basin and seepage lakes, groundwater can be the dominant hydrological input (e.g., Gaiser et al.

127 2009) and can deliver DOC to lakes, especially in organic-rich soils (Schindler and Krabennhoft

128 1998). Empirical measurements of groundwater discharge and DOC concentration, however, are

129 rare and difficult to estimate (Hanson et al. 2014). POC inputs from litterfall, and wet and dry

130 atmospheric deposition are typically small and are generally estimated as a function of lake size

131 and literature- or expert-based loading coefficients (Hanson et al. 2004). While low in

132   magnitude, these OC sources can be important for lakes with high perimeter to surface area

133   ratios or with large surface areas and long hydrologic residence times.

134

135   *Autochthony*

136   Autochthonous DOC and POC originate within lakes through bacterial exudates and

137   photosynthesis by primary producers. Since gross primary production (GPP) is difficult to

138   measure at the ecosystem level, net primary production (NPP), considered the difference

139   between GPP and autotrophic respiration, is typically measured instead (Pace and Lovett 2013;

140   Box 1). Approaches to estimate NPP include bottle incubations (Urban 2005, Yang et al. 2008)

141   and high frequency measurements of dissolved oxygen or CO_2 concentrations (Cole et al. 2002,

142   Staehr et al. 2010). Statistical relationships have also been developed to estimate NPP from lake

143   temperature and total phosphorus (TP; Hanson et al. 2004), chlorophyll-a (chl-*a*; Jonsson et al.

144   2001, Ramlal et al. 2003), or static proportions of the overall OC pool (Åberg et al. 2004).

145

146   *Storage and export*

147   Long-term burial of POC in lake sediments is the mechanism by which lakes remove C

148   from the global C cycle, and is therefore a critical component of our understanding of the fate of

149   both allochthonous and autochthonous POC (Cole et al. 2002, Tranvik et al. 2009). POC burial

150   in lakes is a product of in-lake POC concentrations, POC particle sizes and associated settling

151   rates, sediment particle size and density that control resuspension, lake hydrodynamics that affect

152   settling rates and resuspension, and benthic biogeochemistry (Downing et al. 2008, Xu et al.

153   2013). Methods for estimating sediment accumulation rates are diverse and commonly include

154   functions based on lake area, particularly for studies containing multiple lakes (Canham et al.

9

155   2004, Hanson et al. 2004). Previous studies have estimated burial rates using sediment cores

156   (Yang et al. 2008, Klump et al. 2009, Heathcote & Downing 2012), sediment traps (Jonsson et

157   al. 2001, Ramlal et al. 2003), or bathymetry (Downing et al. 2008). A challenge associated with

158   estimating accumulation rates is the reliance on point measurements to characterize sediment

159   accumulation rates that can vary widely over both space and time. Allochthonous and

160   autochthonous POC that is not buried is mineralized directly or leached in the form of DOC and

161   exported via surface or groundwater (Cole et al. 1984). In some lakes, there is carbon efflux in

162   the form of insect production and emergence (Vander Zanden and Gratton 2011), though that

163   flux is not included in this study. Exports represent allochthonous inputs to downstream aquatic

164   ecosystems and therefore contribute to landscape C cycling (Kling et al. 2000).

165

166   *Objective and research question*

167   Our broad objectives were (1) to quantify long-term (i.e., > 10 year) dynamics and

168   magnitudes of key DOC and POC fluxes through the development of a simple dynamic model,

169   (2) to use this model to both reveal key drivers of OC fates (burial, respiration, and export), and

170   (3) to quantify uncertainties associated with these processes in lakes. We applied the model to

171   five lakes that encompass strong contrasts in morphology, hydrology, and trophic state to

172   understand the relative influence of these lake characteristics on OC cycling (Hanson et al.

173   2011), and to address our overarching research question: What are the magnitudes and

174   uncertainties in processes governing lake OC cycling and how do these change through time?

175   The balance between burial and respiration is particularly important, given that this represents

176   the pathway by which lake OC enters long-term storage on geological time scales or effluxes to

177   the atmosphere, primarily as CO_2.

178

179   **METHODS**

180   *Study lakes and data sources*

181   We modeled temporal dynamics of OC budgets for five lakes that span a range of

182   limnological characteristics (e.g., hydrologic residence time, depth, trophic state; Table 1). Lakes

183   were selected based on contrasting characteristics and availability of observational data.

184   Required observational data included precipitation, hydrological inflow (discharge), inflow DOC

185   concentration, and various in-lake measurements (surface water temperature, chl-*a*

186   concentration, and Secchi depth). All lakes had a minimum of 10 years of limnological data used

187   for model training and at least four years of in-lake DOC and DO measurements for model

188   validation. See the appendix for detailed data descriptions and sources (Appendix S1). Our

189   dataset included four oligotrophic lakes and one eutrophic lake. Lake areas ranged from 71.38 ha

190   to 565000 ha and mean depths ranged from 7 m to 27 m. Hydrologic residence times ranged

191   from 0.8 years to 6.3 years. In-lake mean annual DOC concentrations ranged from 3 g m^{-3} to 6 g

192   m^{-3}. Watersheds are primarily forested for Harp Lake, Trout Lake, and Lake Vanern; whereas

193   Toolik Lake is in a tundra-dominated watershed and Monona is in an agricultural and heavily

194   developed watershed. Although the lakes span a set of relatively wide environmental gradients,

195   we did not include a formal analysis of OC fates along these gradients due to the small number

196   of lakes.

197

198   *General model approach*

199   We developed a relatively simple, dynamic mass balance model (Fig. 1, Tables 2-3), that

200   included four state variables representing OC (Table 3, Eqs. 1-4) and one representing dissolved

11

201   oxygen (DO, Eq. 5). Static and calibrated parameters for the equations are in Table 2.

202   Allochthonous DOC and POC (Eqs. 1-2) for the lakes were modeled separately from

203   autochthonous DOC and POC (Eqs. 3-4). The model complexity was commensurate with the

204   modest number of observational variables available. We operated the model on a daily time step

205   for 10-13 years, based on data availability. Lake volume was assumed static and inflow volume

206   was assumed equal to outflow volume. Observational data of inflow DOC, in-lake chl-a, Secchi

207   depth, and temperature were typically weekly or bi-weekly and were linearly interpolated to a

208   daily time step. Precipitation was assumed zero for missing dates; however, precipitation data

209   gaps were rare. Daily evaporative losses were assumed to be approximately equal to

210   precipitation. To account for the absence of winter data at Toolik, we set inflow DOC to 0 when

211   the main inflow (Toolik Inlet) was frozen (Appendix S1). The model was written and executed in

212   R version 3.3.2 (R Core Team 2016).

213

214   *Allochthonous DOC and POC*

215   Changes in allochthonous DOC were modeled as a function of DOC load, deposition,

216   leaching, mineralization, and export (DOC_{Alloch}, Eq. 1). Allochthonous DOC load was calculated

217   as the sum of inflows (I_{DOC}, Eq. 1.1) from both surface ($I_{DOC.SW}$, Eq. 1.11) and groundwater

218   ($I_{DOC.GW}$, Eq. 1.12), and deposition (DDOC, Eq. 1.2) from precipitation (D_{Precip}, Eq. 1.21) and

219   adjacent wetlands ($D_{Wetland}$, Eq. 1.22). Mass loads were calculated as the product of DOC

220   concentrations and flows, except for $D_{Wetland}$, which was the product of the proportion of lake

221   perimeter that is wetland ($P_{Wetland}$), a parameter representing a transfer coefficient ($CDOC_{Wetland}$)

222   of DOC from the wetland to the lake, and lake perimeter (LakePerimeter). The third input

223   (L_{Alloch}, Eq. 1.3) represented in-lake leaching of POC_{Alloch} to DOC_{Alloch} as the product of a first-

224   order decay rate ($C_{LAlloch}$) and the POC_{Alloch} concentration. There were two fates of DOC_{Alloch}

225   (Eq. 1). The first was mineralization ($MDOC_{Alloch}$, Eq. 1.4), which was the product of a first-

226   order decay rate ($RDOC_{Alloch}$), the DOC_{Alloch} concentration, and a Q_{10} temperature adjustment

227   using a standard Arrhenius equation. The second was export downstream ($EDOC_{Alloch}$, Eq. 1.5),

228   which was the product of DOC_{Alloch} and outflow ($Q_{Outflow}$).

229   Changes in allochthonous POC were modeled as a function of POC load, deposition,

230   leaching, burial, and export (POC_{Alloch}, Eq. 2). Allochthonous POC input (I_{POC}, Eq. 2.1) was

231   modeled as a proportion of $I_{DOC,}$ ($CPOC_{Factor}$). Deposition (DPOC, Eq. 2.2) was the sum of

232   canopy ($DPOC_{Canopy}$) and wetland ($DPOC_{Wetland}$) inputs, where $DPOC_{Canopy}$ (Eq. 2.21) was the

233   product of the proportion of lake perimeter that is canopy (P_{Canopy}), a parameter representing a

234   transfer coefficient ($CPOC_{Aerial}$) of POC from the canopy to the lake, and LakePerimeter.

235    $DPOC_{Wetland}$ (Eq. 2.22) was assumed to scale with $DDOC_{Wetland}$ by the proportion $CPOC_{Factor}$.

236   POC_{Alloch} had a burial fate (B_{Alloch}, Eq. 2.3), calculated as the product of a burial coefficient

237   ($BPOC_{Alloch}$) and POC_{Alloch}. As with DOC_{Alloch}, downstream export ($EPOC_{Alloch}$, Eq. 2.4) was

238   included as the product of POC_{Alloch} and outflow.

239   Daily surface water inflow (m^3 s^{-1}) was generally available for the main tributaries of all

240   study lakes. Inflow DOC concentration (mg L^{-1}) was less frequently measured, and was linearly

241   interpolated to a daily time step from sub-weekly to monthly data (Appendix S1). When inflow

242   DOC concentrations were not available for all tributaries, DOC contributions for each tributary

243   were estimated based on the proportion of total inflow volume and the assumption that inflow

244   DOC concentration was equal across all tributaries. Daily precipitation (Q_{Precip}, mm) was based

245   on measurements from the weather station nearest to each lake. The concentration of DOC in

246   precipitation was set to 2 g m^{-3} (Hanson et al. 2014). Time series of lake-specific groundwater

247   inflow volume (m^3) and DOC concentration ($g\ m^{-3}$) were not available. We estimated the

248   proportion of inflow as groundwater in our study lakes based on literature values when available,

249   but assumed no groundwater in the absence of data (Appendix S1). Resulting estimated

250   groundwater proportions ranged from 0-19%. Groundwater DOC concentration was assumed to

251   be 10 $g\ m^{-3}$ (Table 2: DOC_{GWConc}, Hanson et al. 2014). Shoreline-adjacent wetlands and forests

252   were estimated from GIS and publicly available spatial datasets (Appendix S1). We focused on

253   wetlands adjacent to the shoreline because they contribute most of wetland-derived DOC to lakes

254   not already captured in Eq. 1.1 (Hanson et al. 2014). To account for potential misalignment

255   among spatial wetland and forest data and lake boundaries, we defined adjacency as within 30 m

256   of lake boundaries.

257

258   *Autochthonous DOC and POC*

259   Our approach to modeling autochthony (Table 3, Eqs. 3-4) was generally similar to that

260   of allochthony for leaching, mineralization, export, and burial (Eqs. 3.3-3.5, 4.2-4.3), but differed

261   in the input terms: NPPDOC (Eq. 3.2) and NPPPOC (Eq. 4.1). Total autochthony (NPP_{TOT}, Eq.

262   3.1) was the product of GPP, which was modeled as a function of chl-*a* ($\mu g\ L^{-1}$), mixing depth

263   (Z_{mix}), and surface water temperature (T, °C) per Morin et al. (1999), and the proportion of GPP

264   not respired by autotrophs ($1-R_{Autotroph}$). The GPP function was calculated using observed

265   temperature and chl-*a* data that ranged from 5-25 °C and 1-1000 $mg\ m^{-2}$, respectively, across all

266   lakes. Since models of GPP are not well constrained at low temperatures, we set GPP to zero if

267   surface water temperatures were $< 4\ °C$, as this is the maximum density of water and a point at

268   which lakes would no longer be stratified. Chl-*a* concentrations were converted from volumetric

269   to areal units by multiplying by photic depth, which was estimated from Secchi depth (m; Wetzel

270   2001). The DOC fraction of total NPP (NPPDOC) was calculated using the Pace and Prairie

271   (2005) negative exponential equation (Eq. 3.2). The remainder of NPP_{TOT} was attributed to POC

272   (NPPPOC, Eq. 4.1).

273   Water column dissolved oxygen (DO) was used to constrain net ecosystem production

274   (NEPOC, Eq. 5.1), under the assumption that at short time scales and under pseudo-equilibrium

275   conditions, atmospheric exchange (F_{atm}) approximated NEPOC. F_{atm} (Eq. 5.2) was calculated as

276   a function of piston velocity (k), set to 0.7 m d^{-1}, DO and DO saturation, and the mixing depth,

277   Z_{mix}, which was set equal to half the photic zone depth (m). The saturation of DO (DO_{sat}) is

278   temperature dependent and was determined using the Garcia-Benson method in the

279   LakeMetabolizer R package (Winslow et al. 2016). Heterotrophic respiration was calculated as a

280   function of DOC_{Autoch} and DOC_{Alloch} concentration (g m^{-3}) in the photic zone, epilimnion

281   temperature (assumed to be uniform through the photic zone), and two calibrated parameters:

282    $RDOC_{Autoch}$ and $RDOC_{Alloch}$ (Tables 2-3, Eqs. 1.4, 3.4, 5.12) (see Model calibration and

283   uncertainty analysis). We determined epilimnion temperature by averaging observed

284   temperatures throughout the photic zone when data were available from multiple depths, but

285   otherwise used surface temperature (Appendix S1).

286

287   *Model calibration and uncertainty analysis*

288   The collinearity of the four free parameters in the model (respiration: $RDOC_{Alloch}$,

289    $RDOC_{Autoch}$, burial: $BPOC_{Alloch}$, and $BPOC_{Autoch}$; Table 2) was tested using the *collin* function in

290   the R package FME (Soetaert and Petzoldt 2010). In general, when the collinearity index is less

291   than 20, linear independence is assumed (Brun et al. 2001, Omlin et al. 2001). Finding low

292   collinearity, the four parameters were fit by minimizing the sum of the squared residuals of DOC

15

293   and DO, equally weighted by number of observations. The model was fit using a pseudo-random

294   search algorithm in the FME package. Burial parameters were constrained in the model as a

295   proportion between 0 (no burial of POC) and 1 (all POC is buried). $RDOC_{Alloch}$ was constrained

296   between 0.0003 and 0.03 (d^{-1}) based on the range of OC decomposition rates for inland waters

297   with residence times between 1-10 years presented in Catalan et al. (2016). $RDOC_{Autoch}$ was

298   constrained between 0.003 and 0.3 (d^{-1}) (Hanson et al. 2004). Goodness of fit was evaluated with

299   root mean square error (RMSE) and Nash-Sutcliffe efficiency (NSE) scores calculated for DOC

300   and DO for each lake using the hydroGOF R package (Zambrano-Bigiarini 2017). Goodness of

301   fits were reported for DOC and DO because the model was fit to both simultaneously rather than

302   individually. A sensitivity analysis of each parameter was conducted by allowing the parameter

303   to vary at 100 different values within the set bounds while fixing the other three parameters at

304   their calibrated values.

305   We estimated parameter means and uncertainties using a bootstrapping routine (per

306   Dugan et al. 2017). Using the bootstrapped parameters, we calculated residual errors between

307   observed and modeled DOC and DO. We created 100 pseudo-observational datasets by

308   randomizing these residuals 100 times and adding each randomized residual set to the observed

309   data. We then re-fit the parameters to the pseudo-observational datasets to provide 100 new

310   parameter estimates. Finally, we recorded parameter distribution characteristics and assessed

311   correlations among parameters within each lake.

312

313   **RESULTS**

314   *Model performance, parameter estimates, and sensitivity analysis*

315   Modeled DOC and DO generally followed observed temporal patterns across years in

316   each study lake (Fig. 2). There was an overall strong goodness of fit for both DOC and DO based

317   on RMSE for each lake (0.80-1.46 mg L^{-1}; Table 4). The model also accounted for considerably

318   more temporal variability than long-term means of DOC based on NSE values across lakes

319   (0.70-0.96). Although the model captured annual and seasonal DOC and DO dynamics, the

320   model did not consistently characterize the magnitude of short-term spikes in DOC (i.e., days to

321   weeks; Fig. 2). Nonetheless, long-term model performance indicated the ability to account for

322   lake variability in DOC and DO from seasonal to inter-annual time scales.

323   Across all lakes, parameter estimates for the allochthonous components of the budget

324   were generally more consistent and better constrained than those for autochthony (Table 4).

325   Respiration of allochthonous DOC ($RDOC_{Alloch}$) ranged from about 0.0011-0.0025 d^{-1} among

326   lakes and SEM values were about two orders of magnitude lower, indicating tightly constrained

327   mean values. In contrast, respiration of autochthonous DOC ($RDOC_{Autoch}$) was higher and more

328   variable than $RDOC_{Alloch}$ among lakes, ranging from about 0.0034-0.4500 d^{-1}. Burial rates for

329   allochthony ($BPOC_{Alloch}$) were high, with values at or near the upper limit (1.0000 d^{-1}) for all

330   lakes except Lake Monona. Burial of autochthony ($BPOC_{Autoch}$) was comparatively lower and

331   more variable, ranging from approximately 0.0310-0.8700 d^{-1}. SEM values for burial tended to

332   be about one order of magnitude smaller than corresponding parameter means across all lakes.

333   Modeled DOC (mg L^{-1}) was generally most sensitive to $RDOC_{Alloch}$, except for Lake

334   Monona, for which modeled DOC was most sensitive to $BPOC_{Autoch}$ (Fig. 3). The other four

335   lakes were minimally affected by changes in $BPOC_{Autoch}$ (< 1 mg L^{-1} difference across the range

336   of parameter values). Changes in $BPOC_{Alloch}$ had consistently small effects (< 1 mg L^{-1}) on

337   modeled DOC across all lakes. Trout Lake was the most sensitive to $RDOC_{Autoch}$, with modeled

338   DOC ranging about 2-2.5 mg L^{-1} across the range of parameter values, whereas Toolik Lake and

339   Lake Monona were moderately sensitive (1-1.5 mg L^{-1} differences) and Lake Vanern and Harp

340   lake were minimally sensitive (< 1 mg L^{-1} differences). Overall, parameter sensitivity was

341   greatest for Lake Vanern, Harp Lake, and Lake Monona, for which modeled DOC varied as

342   much as 5-6 mg L^{-1} across the range of parameter values (Fig. 3). Conversely, modeled DOC

343   varied no more than 2 and 3 mg L^{-1} for Toolik Lake and Trout Lake, respectively.

344

345   *Summary of fluxes and fates*

346   With the exception of Trout Lake, OC loads were primarily driven by allochthony,

347   underscoring the importance of terrestrially derived OC in overall lake budgets (Table 5, Fig. 4).

348   Additionally, respiration exceeded burial in all lakes but Lake Monona. Trout Lake also had the

349   smallest total annual OC load of the five lakes (41.52 g m^{-2} yr^{-1}), the lowest proportion of total

350   load exported (0.10), and the largest proportional difference between respiration (0.90) and

351   burial (0.03) among all lakes. Lake Monona had the largest total OC load (118.97 g m^{-2} yr^{-1})

352   among lakes, lowest proportion respired (0.14) and greatest proportion buried (0.37). Lake

353   Monona was the only lake dominated by burial long-term; on average, burial rates were greater

354   than twice respiration rates across modeled years. Harp Lake had the second lowest proportion

355   buried (0.07) among all lakes and exhibited proportions of allochthony (0.55) and autochthony

356   (0.45) that were approximately similar in Lake Monona and Lake Vanern. Similar to Trout Lake,

357   Harp Lake demonstrated a relatively large proportional difference between respiration (0.66) and

358   burial (0.07). Toolik Lake had the second largest (mean = 87.45 g m^{-2} yr^{-1}) but most variable

359   (standard deviation; SD = 62.91 g m^{-2} yr^{-1}) total OC load and was the most driven by allochthony

360   (0.87) among all lakes. This inter-annual variability in total OC load for Toolik Lake was driven

361   by highly variable allochthony (SD = 64.78 g m^{-2} yr^{-1}), and also resulted in highly variable

362   export (53.53 g m^{-2} yr^{-1}). Compared to other lakes, Toolik Lake on average demonstrated

363   relatively low proportion buried (0.08) and relatively high proportion exported (0.64).

364

365   *Seasonal fates*

366   Seasonal patterns in OC fluxes were consistent across entire respective time series for

367   each lake, with autochthony and respiration increasing to a summer maximum (Fig. 4). As water

368   temperatures increased during the growing season (e.g., May – Aug.), the balance between

369   allochthony and autochthony generally shifted toward autochthony due to increases in NPP,

370   whereas the ratio between respiration and burial generally shifted towards respiration (Figs. 4-5).

371   There was high seasonal variability in the dominant fluxes in each lake (Fig. 5). Trout Lake

372   remained dominated by respiration year-round, but respiration increased relative to burial as

373   water temperatures warmed. Harp Lake, Toolik Lake, and Lake Vanern were dominated by

374   burial early in the growing season, but were dominated by respiration on an annual basis due to

375   increased respiration rates as temperatures warmed. Whereas respiration in Lake Monona

376   exceeded burial late in the growing season, the lake remained dominated by burial when

377   calculated on an annual basis (Table 5, Fig. 5). Lake Monona and Lake Vanern also showed less

378   of a proportional increase in autochthony (i.e., lacked a negative slope in Fig. 5) as the growing

379   season progressed compared to other lakes, suggesting the importance of continued

380   allochthonous inputs during summer months in these lakes. In the other three lakes, the

381   proportional shift toward autochthony was associated with a proportional shift from burial to

382 respiration as water temperatures increased, but this association was relatively weaker in Lake

383 Monona and Lake Vanern (Fig. 5). Overall, these seasonal dynamics suggest that water

384 temperatures are associated with changes in the balance between key OC fates (burial and

385 respiration), but that such shifts are mediated by the balance between allochthony and

386 autochthony that vary across lake systems.

387

388 **DISCUSSION**

389 *Representing lake processes*

390   Our results indicated that a relatively simple, dynamic model can recreate long-term

391 trends in DOC and represent the set of key biogeochemical, trophic, and landscape processes that

392 combine to determine the fate of OC in lake ecosystems. While postulated previously (Hanson et

393 al. 2011), to our knowledge, this is the first demonstration of a dynamic OC model applied to

394 contrasting lakes. Further, the fluxes we modeled were within the range of other published

395 studies for these lakes based on steady-state models. Using much of the same LTER data,

396 Hanson et al. (2014) provided similar estimates for Trout of allochthony (Hanson: 15.92 g m^{-2} yr^{-}

397 ^{1}, our model: 13.35 g m^{-2} yr^{-1}), burial (Hanson: 3.66 g m^{-2} yr^{-1}, our model: 1.19 g m^{-2} yr^{-1}), and

398 export (Hanson: 4.95 g m^{-2} yr^{-1}, our model: 3.95 g m^{-2} yr^{-1}), but not for respiration (Hanson: 7.31

399 g m^{-2} yr^{-1}, our model: 37.45 g m^{-2} yr^{-1}), because Hanson et al. (2014) did not account for

400 autochthony. Similar to our study, Whalen and Cornwall (1985) demonstrated that Toolik Lake

401 was driven by high proportional allochthony (Whalen and Cornwall 0.91, our study: 0.87)

402 relative to autochthony and low burial (Whalen and Cornwall: 0.02, our study: 0.08). Our

403 proportion exported (0.64) contrasted somewhat with Whalen and Cornwall (0.82), but much of

404 this excess export was respired (0.27) in our model. Dillon and Molot's (1997) proportional

405   estimates for burial in Harp Lake were low and similar to ours (Dillon and Molot: 0.01, our

406   model: 0.07) and the magnitudes of allochthony were somewhat similar (Dillon and Molot: 28.9

407   g m^{-2} yr^{-1}, our study: 39.88 g m^{-2} yr^{-1}), but Dillon and Molot (1997) did not consider

408   autochthony. Therefore, we are unable to compare total loads and differences in the proportion

409   exported in Dillon and Molot (0.58) vs. our study (0.24) may be explained by our inclusion of

410   autochthony and respiration. Differences in budget estimates may also be due to differences in

411   study years (Dillon and Molot: 1981-1989, our study: 1991-2001). Although our results

412   generally agreed with prior studies based on steady-state models, our estimated respiration rates

413   were generally higher than those noted in the literature (Hanson et al. 2014, Dillon and Molot

414   1997). We offer that dynamic models better represent these processes by accounting for seasonal

415   changes in temperature and chl-*a* concentrations. Therefore, although steady-state models may

416   be sufficient for recreating some key ecological processes, dynamic models are needed for

417   determining the relative magnitudes of OC fates in lake ecosystems, given the importance of

418   autochthony and respiration.

419

420   *Key uncertainties in OC fates*

421   Well-constrained estimates of OC burial in lakes remain a challenge to model. Although

422   our estimates for burial parameters have relatively low uncertainties (Table 4), burial may be

423   underestimated for these lakes. A key consideration is that our model buried close to 100% of

424   POC_{Alloch} in four of five lakes (Table 4: $BPOC_{Alloch}$); therefore, any increase in POC_{Alloch} would

425   be directly proportional to increases in burial. Owing to lack of observational data, we assumed

426   POC_{Alloch} was 10% of DOC_{Alloch} ($CPOC_{Factor}$), but this may be an underestimate, and does not

427   account for potential seasonal variation in the DOC:POC ratio. Intense precipitation can increase

21

428   POC concentration disproportionately to DOC concentration in streams (Jeong et al. 2012,

429   Dhillon and Inamdar 2014, which could temporarily increase POC_{Alloch} and thus burial. In

430   addition, our model demonstrated DOC responses to precipitation events when we had

431   corresponding weather and DOC_{Alloch} data, but the assumed linear changes in inflow volume and

432   DOC_{Alloch} between observation points may also have underrepresented DOC_{Alloch}, leading

433   indirectly to underrepresentation of POC_{Alloch}. Wet years increase DOC_{Alloch} inputs to lakes at

434   regional scales by increasing connectivity among waterbodies (Rose et al. 2016) and therefore

435   probably also increase POC_{Alloch}. Although our burial estimates were somewhat uncertain due to

436   underrepresentation of POC_{Alloch} during precipitation events, burial would have to increase

437   substantially over the course of the entire modeling period for burial to dominate over

438   respiration, including three-fold or greater for Harp Lake, Toolik Lake and Trout Lake (Table 5).

439   Therefore, missing POC_{Alloch} likely leads to underestimates of burial, but is unlikely to account

440   for enough OC to exceed the magnitudes of respiration in many lake ecosystems.

441

442   *On-going research and data needs*

443   Our work is an important advance in quantifying the fates of OC across aquatic

444   ecosystems; however, we encountered constraints associated with current data availability. If

445   requisite data were collected for a larger number of lakes spanning wider environmental

446   gradients (e.g., climate, watershed conditions, trophic state), contributions of lakes to landscape

447   carbon cycles could be better estimated at broad spatial scales. During model development, we

448   encountered a notable paucity of high-frequency measurements of inflow DOC concentration, of

449   which broader collection would facilitate dynamic OC modeling in more lakes. Although

450   collection of these data may be expensive and logistically challenging, the increasing availability

451   of automated, high-frequency sensor equipment may alleviate long-term costs associated with

452   sensor deployment and manual data retrieval. In addition, relatively little is currently known

453   about POC budgets despite their key interactions with DOC (Einsele et al. 2001); we need more

454   POC observational data for incorporation into dynamic models of OC, particularly in inflows for

455   estimating POC_{Alloch}. Such studies would help constrain POC parameters and improve estimates

456   of the fates of POC within overall OC budgets. Thus, a key implication of our study is the need

457   for more observational data, particularly pertaining to surface water DOC concentrations, POC

458   cycling, and burial rates. Despite these uncertainties, lake resource managers should be aware of

459   the importance of the balance between allochthonous and autochthonous inputs to lake OC

460   budgets.

461

462   *Lessons from a dynamic model: implications in a dynamic world*

463   Prior to our study, it was known that lakes actively process, emit and store globally

464   significant amounts of C (Cole et al. 2007, Tranvik et al. 2009, Raymond et al. 2013). Our results

465   demonstrate that a dynamic model can considerably advance knowledge on the role of lakes in

466   landscape and ultimately global C cycling by highlighting dominant inputs and fates of OC in

467   individual systems. Lakes more readily respire autochthonous than allochthonous OC (Wetzel

468   2001). In our study, our one lake dominated by autochthony (Trout Lake) exhibited the greatest

469   respiration relative to burial (Table 5). Therefore, lakes as global C sources or sinks may depend

470   both on the balances between 1) respiration and burial and 2) allochthony and autochthony. The

471   balance between respiration and burial can vary according to regional climate and respiration is

472   typically greater than burial in boreal lakes compared to those in higher latitudes (Anthony et al.

473   2014). This represents a potential negative feedback for the global C cycle under a warming

474 climate with poleward boreal advance and thawing of frozen, high-latitude lakes (Anthony et al.

475 2014).

476   Our model identified another important global change implication associated with

477 warming water temperatures. Across all lakes in our study, warm surface temperatures were

478 generally associated with a shift toward autochthony relative to allochthony, as well as an

479 increase in respiration relative to burial (Fig. 5). This likely is due to elevated NPP during

480 summer growing seasons accompanied by relatively high respiration rates of autochthony

481 relative to allochthony (Table 4). Although the balance between respiration and burial appears to

482 shift toward respiration with increases in temperature, it is also possible for burial to increase

483 with temperature if temperature increases coincide with greater OC loads (e.g., warm-season

484 precipitation events increasing POC_{Alloch} and consequently burial as a function of $BPOC_{Alloch}$).

485 As such, our results suggest that processes favoring allochthony will generally have a greater

486 effect on OC burial than processes that drive autochthony. More broadly, however, lakes have

487 generally become more productive under recent climate warming (Kraemer et al. 2016), which

488 our study suggests favors autochthony over allochthony and respiration over burial. Therefore,

489 changes in both precipitation (including magnitude, timing, duration, and form) and temperature

490 have key implications for the fate of OC in lake ecosystems under a changing climate; however,

491 effects of warming will vary according to the balance between allochthony and autochthony,

492 which is mediated by precipitation due to its effect on the origin of the total OC load. These

493 findings illustrate the usefulness of a dynamic mass balance model for highlighting key global

494 change processes and interactions that ultimately influence the role of lakes in global C cycling.

495 Improved estimates of the contribution of lakes to global C budgets should account for the

496 source and degradability of total OC loads and consequent effects on respiration and burial.

497

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513

LITERATURE CITED

515 Åberg, J., Bergström, A. K., Algesten, G., Söderback, K., & Jansson, M. (2004). A comparison

516   of the carbon balances of a natural lake (L. Örträsket) and a hydroelectric reservoir (L.

517   Skinnmuddselet) in northern Sweden. *Water Research*, *38*(3), 531-538.

25

518   Andersson, E., & Sobek, S. (2006). Comparison of a mass balance and an ecosystem model

519   approach when evaluating the carbon cycling in a lake ecosystem. *AMBIO: A Journal of*

520   *the Human Environment*, *35*(8), 476-483.

521   Anthony, K. W., Zimov, S. A., Grosse, G., Jones, M. C., Anthony, P. M., Chapin III, F. S., ... &

522   Frolking, S. (2014). A shift of thermokarst lakes from carbon sources to sinks during the

523   Holocene EPOCh. *Nature*, *511*(7510), 452-456.

524   Arvola, L., Kortelainen, P. I. R. K. K. O., Bergström, I., Kankaala, P., Ojala, A., Pajunen, H. A.

525   N. N. U., ... & Rantakari, M. I. I. T. T. A. (2002). Carbon pathways through boreal lakes:

526   A multi-scale approach (CARBO). *Understanding the Global System, The Finnish*

527   *Perspective, edited by Käyhkö J and Talve L*, 97-106.

528   Bastviken, D., Tranvik, L. J., Downing, J. A., Crill, P. M., & Enrich-Prast, A. (2011). Freshwater

529   methane emissions offset the continental carbon sink. *Science*, *331*(6013), 50-50.

530   Brun, R., Reichert, P. and Kunsch, H. R., (2001). Practical Identifiability Analysis of Large

531   Environmental Simulation Models. Water Resources Research. 37(4): 1015-1030.

532   Canham, C. D., Pace, M. L., Papaik, M. J., Primack, A. G., Roy, K. M., Maranger, R. J., ... &

533   Spada, D. M. (2004). A spatially explicit watershed-scale analysis of dissolved organic

534   carbon in Adirondack lakes. *Ecological Applications*, *14*(3), 839-854.

535   Catalán, N., Marcé, R., Kothawala, D. N., & Tranvik, L. J. (2016). Organic carbon

536   decomposition rates controlled by water retention time across inland waters. *Nature*

537   *Geoscience, 9*(7), 501-504.

538   Cole, J. J., McDowell, W. H., & Likens, G. E. (1984). Sources and molecular weight of"

539   dissolved" organic carbon in an oligotrophic lake. *Oikos 42*(1), 1-9.

540   Cole, J. J., Carpenter, S. R., Kitchell, J. F., & Pace, M. L. (2002). Pathways of organic carbon

541   utilization in small lakes: Results from a whole☐lake 13C addition and coupled model.

542   *Limnology and Oceanography*, *47*(6), 1664-1675.

543   Cole, J. J., Prairie, Y. T., Caraco, N. F., McDowell, W. H., Tranvik, L. J., Striegl, R. G., ... &

544   Melack, J. (2007). Plumbing the global carbon cycle: integrating inland waters into the

545   terrestrial carbon budget. *Ecosystems*, *10*(1), 172-185.

546   Cremona, F., Kõiv, T., Nõges, P., Pall, P., Rõõm, E. I., Feldmann, T., ... & Nõges, T. (2014).

547   Dynamic carbon budget of a large shallow lake assessed by a mass balance approach.

548   *Hydrobiologia*, *731*(1), 109-123.

549   Dhillon, G. S., & Inamdar, S. (2013). Storm event patterns of particulate organic carbon (POC)

550   for large storms and differences with dissolved organic carbon (DOC). *Biogeochemistry*,

551   *118*(1-3), 61-81.

552   Dillon, P. J., & Molot, L. A. (1997). Dissolved organic and inorganic carbon mass balances in

553   central Ontario lakes. *Biogeochemistry*, *36*(1), 29-42.

554   Downing, J. A., Cole, J. J., Middelburg, J. J., Striegl, R. G., Duarte, C. M., Kortelainen, P., ... &

555   Laube, K. A. (2008). Sediment organic carbon burial in agriculturally eutrophic

556   impoundments over the last century. *Global Biogeochemical Cycles*, *22*(1), GB1018,

557   doi:10.1029/2006GB002854.

558   Dugan, H. A., Bartlett, S. L., Burke, S. M., Doubek, J. P., Krivak-Tetley, F. E., Skaff, N. K., ... &

559   Weathers, K. C. (2017). Salting our freshwater lakes. *Proceedings of the National

560   Academy of Sciences*, *114*(17), 4453-4458.

561   Einola, E., Rantakari, M., Kankaala, P., Kortelainen, P., Ojala, A., Pajunen, H., ... & Arvola, L.

562   (2011). Carbon pools and fluxes in a chain of five boreal lakes: a dry and wet year

27

563   comparison. *Journal of Geophysical Research: Biogeosciences*, *116*(G03009,

564   doi:10.1029/2010JG001636, 2011.

565   Einsele, G., Yan, J., & Hinderer, M. (2001). Atmospheric carbon burial in modern lake basins

566   and its significance for the global carbon budget. *Global and Planetary Change*, *30*(3),

567   167-195.

568   Gaiser, E. E., Deyrup, N. D., Bachmann, R. W., Battoe, L. D., & Swain, H. M. (2009).

569   Multidecadal climate oscillations detected in a transparency record from a subtropical

570   Florida lake. *Limnology and Oceanography*, 54(6), 2228-2232.

571   Hanson, P. C., Buffam, I., Rusak, J. A., Stanley, E. H., & Watras, C. (2014). Quantifying lake

572   allochthonous organic carbon budgets using a simple equilibrium model. *Limnology and

573   Oceanography*, *59*(1), 167-181.

574   Hanson, P. C., Hamilton, D. P., Stanley, E. H., Preston, N., Langman, O. C., & Kara, E. L.

575   (2011). Fate of allochthonous dissolved organic carbon in lakes: a quantitative approach.

576   *PLoS One*, *6*(7), e21884.

577   Hanson, P. C., Pace, M. L., Carpenter, S. R., Cole, J. J., & Stanley, E. H. (2015). Integrating

578   landscape carbon cycling: research needs for resolving organic carbon budgets of lakes.

579   *Ecosystems*, *18*(3), 363-375.

580   Hanson, P. C., Pollard, A. I., Bade, D. L., Predick, K., Carpenter, S. R., & Foley, J. A. (2004). A

581   model of carbon evasion and sedimentation in temperate lakes. *Global Change Biology*,

582   *10*(8), 1285-1298.

583   Heathcote, A. J., & Downing, J. A. (2012). Impacts of eutrophication on carbon burial in

584   freshwater lakes in an intensively agricultural landscape. *Ecosystems*, *15*(1), 60-70.

585   Jeong, J. J., Bartsch, S., Fleckenstein, J. H., Matzner, E., Tenhunen, J. D., Lee, S. D., ... & Park,

586   J. H. (2012). Differential storm responses of dissolved and particulate organic carbon in a

587   mountainous headwater stream, investigated by high☐frequency, in situ optical

588   measurements. *Journal of Geophysical Research: Biogeosciences*, *117* (G03013,

589   doi:10.1029/2012JG001999).

590   Jonsson, A., Meili, M., Bergström, A. K., & Jansson, M. (2001). Whole☐lake mineralization of

591   allochthonous and autochthonous organic carbon in a large humic lake (Örträsket, N.

592   Sweden). *Limnology and Oceanography*, *46*(7), 1691-1700.

593   Kawasaki, N., & Benner, R. (2006). Bacterial release of dissolved organic matter during cell

594   growth and decline: molecular origin and composition. *Limnology and Oceanography*,

595   *51*(5), 2170-2180.

596   Kling, G. W., Kipphut, G. W., Miller, M. M., & O'Brien, W. J. (2000). Integration of lakes and

597   streams in a landscape perspective: the importance of material processing on spatial

598   patterns and temporal coherence. *Freshwater Biology*, *43*(3), 477-497.

599   Klump, J. V., Fitzgerald, S. A., & Waplesa, J. T. (2009). Benthic biogeochemical cycling,

600   nutrient stoichiometry, and carbon and nitrogen mass balances in a eutrophic freshwater

601   bay. *Limnology and Oceanography*, *54*(3), 692-712.

602   Kortelainen, P., Rantakari, M., Huttunen, J. T., Mattsson, T., Alm, J., Juutinen, S., ... &

603   Martikainen, P. J. (2006). Sediment respiration and lake trophic state are important

604   predictors of large CO2 evasion from small boreal lakes. *Global Change Biology*, *12*(8),

605   1554-1567.

606 Kraemer, B. M., Chandra, S., Dell, A. I., Dix, M., Kuusisto, E., Livingstone, D. M., ... &

607   McIntyre, P. B. (2016). Global patterns in lake ecosystem responses to warming based on

608   the temperature dependence of metabolism. *Global Change Biology 23*(5), 1881-1890.

609 Kvarnäs, H. (2001). Morphometry and hydrology of the four large lakes of Sweden. *AMBIO: A*

610   *Journal of the Human Environment*, *30*(8), 467-474.

611 Morin, A., Lamoureux, W., & Busnarda, J. (1999). Empirical models predicting primary

612   productivity from chlorophyll a and water temperature for stream periphyton and lake

613   and ocean phytoplankton. *Journal of the North American Benthological Society*, *18*(3),

614   299-307.

615 Mulholland, P. J., & Elwood, J. W. (1982). The role of lake and reservoir sediments as sinks in

616   the perturbed global carbon cycle. *Tellus*, *34*(5), 490-499.

617 O'Connor, E. M., Dillon, P. J., Molot, L. A., & Creed, I. F. (2009). Modeling dissolved organic

618   carbon mass balances for lakes of the Muskoka River Watershed. *Hydrology Research*,

619   *40*(2-3), 273-290.

620 Omlin, M., Brun, R. and Reichert, P., (2001). Biogeochemical Model of Lake Zurich:

621   Sensitivity, Identifiability and Uncertainty Analysis. Ecological Modelling. 141*(1)*: 105-

622   123.

623 Pace ML & Lovett G. (2013). Primary production: the foundation of ecosystems. In: Weathers

624   K, Strayer D, Likens G, editors. Fundamentals of ecosystem science. Academic Press. p.

625   312.

626 Pace, M. L., & Prairie, Y. T. (2005). Respiration in lakes. *Respiration in aquatic ecosystems*, *1*,

627   103-122.

628 R Core Team (2016). R: A language and environment for statistical computing. R Foundation for

629   Statistical Computing, Vienna, Austria. URL: https://www.R-project.org/.

630   Ramlal, P. S., Hecky, R. E., Bootsma, H. A., Schiff, S. L., & Kingdon, M. J. (2003). Sources and

631   fluxes of organic carbon in Lake Malawi/Nyasa. *Journal of Great Lakes Research*, *29*,

632   107-120.

633   Raymond, P. A., & Bauer, J. E. (2001). Riverine export of aged terrestrial organic matter to the

634   North Atlantic Ocean. *Nature*, *409*(6819), 497-500.

635   Raymond, P. A., Hartmann, J., Lauerwald, R., Sobek, S., McDonald, C., Hoover, M., ... &

636   Kortelainen, P. (2013). Global carbon dioxide emissions from inland waters. *Nature*,

637   *503*(7476), 355-359.

638   Rose, K. C., Greb, S. R., Diebel, M., & Turner, M. G. (2017). Annual precipitation regulates

639   spatial and temporal drivers of lake water clarity. *Ecological Applications*, *27*(2), 632-

640   643.

641   Santoso, A. B., Hamilton, D. P., Hendy, C. H., & Schipper, L. A. (2017). Carbon dioxide

642   emissions and sediment organic carbon burials across a gradient of trophic state in eleven

643   New Zealand lakes. *Hydrobiologia, 795*(1): 341-354.

644   Schindler, D. W., Curtis, P. J., Bayley, S. E., Parker, B. R., Beaty, K. G., & Stainton, M. P.

645   (1997). Climate-induced changes in the dissolved organic carbon budgets of boreal lakes.

646   *Biogeochemistry*, *36*(1), 9-28.

647   Schindler, J. E., & Krabbenhoft, D. P. (1998). The hyporheic zone as a source of dissolved

648   organic carbon and carbon gases to a temperate forested stream. *Biogeochemistry*, *43*(2),

649   157-174.

650   Sobek, S., Söderbäck, B., Karlsson, S., Andersson, E., & Brunberg, A. K. (2006). A carbon

651   budget of a small humic lake: an example of the importance of lakes for organic matter

652   cycling in boreal catchments. *AMBIO: A Journal of the Human Environment*, *35*(8), 469-

653   475.

654   Soetaert, K. and Petzoldt, T., (2010). Inverse Modelling, Sensitivity and Monte Carlo Analysis in

655   R Using Package FME. *Journal of Statistical Software 33*(3) 1-

656   28. http://www.jstatsoft.org/v33/i03

657   Staehr, P. A., Sand-Jensen, K., Raun, A. L., Nilsson, B., & Kidmose, J. (2010). Drivers of

658   metabolism and net heterotrophy in contrasting lakes. *Limnology and Oceanography*,

659   *55*(2), 817-830.

660   Striegl, R. G., & Michmerhuizen, C. M. (1998). Hydrologic influence on methane and carbon

661   dioxide dynamics at two north-central Minnesota lakes. *Limnology and Oceanography*,

662   *43*(7), 1519-1529.

663   Tanentzap, A. J., Szkokan-Emilson, E. J., Desjardins, C. M., Orland, C., Yakimovich, K.,

664   Dirszowsky, R., ... & Gunn, J. (2017). Bridging between litterbags and whole-ecosystem

665   experiments: a new approach for studying lake sediments. *Journal of Limnology 76(2),*

666   *431-437*.

667   Tranvik, L. J., Downing, J. A., Cotner, J. B., Loiselle, S. A., Striegl, R. G., Ballatore, T. J., ... &

668   Kortelainen, P. L. (2009). Lakes and reservoirs as regulators of carbon cycling and

669   climate. *Limnology and Oceanography*, *54*(6part2), 2298-2314.

670   Urban, N. R., Auer, M. T., Green, S. A., Lu, X., Apul, D. S., Powell, K. D., & Bub, L. (2005).

671   Carbon cycling in Lake Superior. *Journal of Geophysical Research: Oceans (1978–*

672   *2012)*, *110* (C06S90), doi:10.1029/2003JC002230.

673   Vander Zanden, M. J., & Gratton, C. (2011). Blowin'in the wind: reciprocal airborne carbon

674   fluxes between lakes and land. *Canadian Journal of Fisheries and Aquatic*

675   *Sciences*, *68*(1), 170-182.

676   Webster, K. E., Kratz, T. K., Bowser, C. J., Magnuson, J. J., & Rose, W. J. (1996). The influence

677   of landscape position on lake chemical responses to drought in northern Wisconsin.

678   *Limnology and Oceanography*, *41*(5), 977-984.

679   Wetzel, R. G. (2001). *Limnology: lake and river ecosystems*. Gulf Professional Publishing.

680   Weyhenmeyer, G. A., Kosten, S., Wallin, M. B., Tranvik, L. J., Jeppesen, E., & Roland, F.

681   (2015). Significant fraction of CO2 emissions from boreal lakes derived from hydrologic

682   inorganic carbon inputs. *Nature Geoscience 8*(12), 933-936.

683   Whalen, S. C., & Cornwell, J. C. (1985). Nitrogen, phosphorus, and organic carbon cycling in an

684   arctic lake. *Canadian Journal of Fisheries and Aquatic Sciences*, *42*(4), 797-808.

685   Winslow, L. A., Zwart, J. A., Batt, R. D., Dugan, H., Woolway, R. I., Corman, J., ... & Read, J.

686   S. (2016). LakeMetabolizer: an R package for estimating lake metabolism from free-

687   water oxygen using diverse statistical models. *Inland Waters*, *6*(4), 622-636.

688   Xu, H., Lan, J., Liu, B., Sheng, E., & Yeager, K. M. (2013). Modern carbon burial in Lake

689   Qinghai, China. *Applied Geochemistry*, *39*, 150-155.

690   Yang, H., Xing, Y., Xie, P., Ni, L., & Rong, K. (2008). Carbon source/sink function of a

691   subtropical, eutrophic lake determined from an overall mass balance and a gas exchange

692   and carbon burial balance. *Environmental Pollution*, *151*(3), 559-568.

693   Yao, H., McConnell, C., Somers, K. M., Yan, N. D., Watmough, S., & Scheider, W. (2011).

694   Nearshore human interventions reverse patterns of decline in lake calcium budgets in

695   central Ontario as demonstrated by mass□balance analyses. *Water Resources Research*,

33

696   *47*(W06521), doi:10.1029/2010WR010159.

697   Zambrano-Bigiarini, M. (2017). hydroGOF: Goodness-of-fit functions for comparison of

698   simulated and observed hydrological time series. R package version 0.3-10. URL

699   http://hzambran.github.io/hydroGOF/. DOI:10.5281/zenodo.84008.

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701

702   **Tables**

703   **Table 1. Lake characteristics**

Lake	Harp	Monona	Toolik	Trout	Vanern
Location	Ontario, Canada	Wisconsin, USA	Alaska, USA	Wisconsin, USA	Sweden
Lat, Long	45.38, -79.14	43.06, -89.36	68.63, -149.61	46.04, -89.69	58.87, 13.41
Data years	1991-2001	2003-2014	2001-2010	2004-2013	2001-2013
Lake area (ha)	71	1326	149	1610	565000
Perimeter (m)	4000	35200	8104	25900	2007000
z_{mean} (m)	12	8.3	7	14.6	27
RT (yr)	2.5	0.8	0.8	5.9	6.3
Trophic state	oligotrophic	eutrophic	oligotrophic	oligotrophic	oligotrophic
Secchi (m)	4.3	2.7	4.7	5.3	4.5
Chl-a ($\mu g\ L^{-1}$)	2.4	9.2	1.1	2.2	2.1
SW DOC ($g\ m^{-3}$)	9.9	5.2	6.8	5.1	9.4
Lake DOC ($g\ m^{-3}$)	4	6	5	3	4
P_{Canopy}	1.000	0.167	0.000	0.780	0.615
$P_{Wetlands}$	0.000	0.026	0.133	0.011	0.037
Burial rate ($g\ m^{-2}\ yr^{-1}$)	78	249	153	27	186
References	Yao et al. 2011	NTL LTER	Kling et al. 2000	Webster et al. 1996, NTL LTER	Kvarnäs 2001

704   z_{mean} = mean depth, RT = hydrologic residence time, Secchi = Secchi depth, Chl-a = chlorophyll-a, SW DOC = inflow dissolved
705   organic carbon, Lake DOC = in-lake DOC (mean water column). All values are means from all available model calibration data or
706   were derived from cited references. NTL LTER = https://lter.limnology.wisc.edu. See Appendix S1 for sources of burial rates.

35

707 **Table 2. Lake model parameters (calibrated parameters italicized, n = 4)**
708

Parameter	Description	Value	Units
DOC_{GWconc}	DOC concentration of groundwater	10	g m^{-3}
$DOC_{PrecipConc}$	DOC concentration of precipitation	2	g m^{-3}
$CDOC_{Wetland}$	Loading rate of POC from wetlands	1	g m-shoreline^{-1} d^{-1}
$C_{LAlloch}$	Proportion of allochthonous POC that is leached to DOC	1- $BPOC_{Alloch}$	Unitless
$RDOC_{Alloch}$	*Decomposition rate of allochthonous DOC in heterotrophic respiration*	*Calibrated*	*d^{-1}*
θ	Temperature multiplier	1.08	Unitless
$CPOC_{Factor}$	Concentration of inflow POC relative to DOC	0.10	Unitless
$CPOC_{Aerial}$	Loading rate of aerial POC (i.e., leaflitter)	1	g m-shoreline^{-1} d^{-1}
$BPOC_{Alloch}$	*Proportion of allochthonous POC buried in sediments*	*Calibrated*	Unitless
$C_{LAutoch}$	Proportion of autochthonous POC that is leached to DOC	1- $BPOC_{Auto}$	Unitless
$RDOC_{Autoch}$	*Decomposition rate of autochthonous DOC in heterotrophic respiration*	*Calibrated*	*d^{-1}*
$BPOC_{Autoch}$	*Proportion of autochthonous POC buried in sediments*	*Calibrated*	Unitless
k	Gas flux coefficient	0.7	m d^{-1}
$R_{Autotroph}$	Proportion of GPP respired by autotrophs	0.8	Unitless

Table 3. Model Equations

No.	Equation
1	$DDOC_{Alloch}/dt = I_{DOC} + DDOC + L_{Alloch} - MDOC_{Alloch} - EDOC_{Alloch}$
2	$DPOC_{Alloch}/dt = I_{POC} + DPOC - L_{Alloch} - B_{Alloch} - EPOC_{Alloch}$
3	$DDOC_{Autoch}/dt = NPPDOC + L_{Autoch} - MDOC_{Autoch} - EDOC_{Autoch}$
4	$DPOC_{Autoch}/dt = NPPPOC - L_{Autoch} - B_{Autoch} - EPOC_{Autoch}$
5	$dDO/dt = NEPOC + F_{atm}$
	Allochthony, DOC
1.1	$I_{DOC} = I_{DOC.SW} + I_{DOC.GW}$
1.11	$I_{DOC.SW} = DOC_{SWconc} * Q_{SW}$
1.12	$I_{DOC.GW} = DOC_{GWconc} * Q_{GW}$
1.2	$DDOC = DDOC_{Precip} + DDOC_{Wetland}$
1.21	$DDOC_{Precip} = DOC_{PrecipConc} * Q_{Precip}$
1.22	$DDOC_{Wetland} = P_{Wetland} * CDOC_{Wetland} * LakePerimeter$
1.3	$L_{Alloch} = C_{L.Alloch} * POC_{Alloch}$
1.4	$MDOC_{Alloch} = RDOC_{Alloch} * DOC_{Alloch} * \theta^{(T-T_{Base})}$
1.5	$EDOC_{Alloch} = DOC_{Alloch} * Q_{Outflow}$
	Allochthony, POC
2.1	$I_{POC} = I_{DOC} * CPOC_{Factor}$
2.2	$DPOC = DPOC_{Canopy} + DPOC_{Wetland}$
2.21	$DPOC_{Canopy} = P_{Canopy} * CPOC_{Aerial} * LakePerimeter$
2.22	$DPOC_{Wetland} = DDOC_{Wetland} * CPOC_{Factor}$
2.3	$B_{Alloch} = BPOC_{Alloch} * POC_{Alloch}$
2.4	$EPOC_{Alloch} = POC_{Alloch} * Q_{Outflow}$
	Autochthony, DOC
3.1	$NPP_{Tot} = 10^{(1.18 + (0.92 * log10(chl\text{-}a * Zmix) + (0.014 * T)))} * (1-R_{Autotroph})$
3.2	$NPPDOC = 0.2 * NPP_{Tot} * (chl\text{-}a * z_{mix})^{-0.22} * (0.714)$
3.3	$L_{Autoch} = C_{LAutoch} * POC_{Autoch}$
3.4	$MDOC_{Autoch} = RDOC_{Autoch} * DOC_{Autoch} * \theta^{(T-T_{Base})}$
3.5	$EDOC_{Autoch} = DOC_{Autoch} * Q_{Outflow}$
	Autochthony, POC
4.1	$NPPPOC = NPP_{Tot} - NPPDOC$
4.2	$B_{Autoch} = BPOC_{Autoch} * POC_{Autoch}$
4.3	$EPOC_{Autoch} = POC_{Autoch} * Q_{Outflow}$
	DO
5.1	$NEPOC = F_{atm} = NPP_{Tot} - R_{Tot}$

37

5.11	$NPP_{Tot} = NPPDOC + NPPPOC$
5.12	$R_{Tot} = MDOC_{Alloch} + MDOC_{Autoch}$
5.2	$F_{atm} = k * (DO - DO_{Sat}) * Z_{mix}^{-1}$
	Abbreviations: I = Inflow, E = Export, D = Deposition, L = Leaching, M = mineralization, R = Respiration, B = Burial

38

Table 4. Model goodness of fit and estimates of the parameter means (parentheses include standard error of the mean estimate, SEM). All parameters had a collinearity < 20 and were assumed independent. RMSE = root mean square error, NSE = Nash-Sutcliffe Efficiency score.

Lake	RMSE	NSE	RDOC$_{Alloch}$ (d^{-1})	RDOC$_{Autoch}$ (d^{-1})	BPOC$_{Alloch}$	BPOC$_{Autoch}$
Harp	1.22	0.86	0.0025 (2.1e-5)	0.0034 (9.4e-5)	1.0000 (4.2e-5)	0.8400 (2.2e-2)
Monona	1.46	0.70	0.0009 (9.3e-5)	0.1600 (1.3e-2)	0.4100 (3.2e-2)	0.6800 (3.5e-2)
Toolik	1.19	0.79	0.0025 (3.2e-5)	0.0350 (5.6e-3)	1.0000 (1.4e-5)	0.0310 (1.7e-2)
Trout	0.80	0.95	0.0014 (4.7e-5)	0.0320 (7.6e-3)	0.9300 (2.4e-2)	0.8700 (2.8e-2)
Vanern	0.87	0.96	0.0011 (5.1e-5)	0.4500 (8.0e-2)	0.9500 (1.8e-2)	0.5900 (4.7e-2)

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Table 5. Summary of annual mass balances (g m^{-2} y^{-1}), including allochthonous (Alloch) and autochthonous (Autoch) loads, respiration (Resp), burial, and export. Standard deviations (SD) of the annual means are shown, as well as the proportions of load by source and fate.

Lake	Alloch	Autoch	Total Load	Resp	Burial	Export
Annual means						
Harp	39.88	32.03	71.91	47.46	5.30	17.60
Monona	64.74	54.23	118.97	16.76	43.81	59.95
Toolik	76.19	11.27	87.45	23.68	6.67	55.59
Trout	13.35	28.17	41.52	37.45	1.19	3.95
Vanern	32.25	26.08	58.32	29.20	19.85	12.12
SD of annual means						
Harp	10.71	7.99	11.42	9.42	0.95	4.86
Monona	24.16	18.05	21.81	1.99	12.85	20.58
Toolik	64.78	3.36	62.91	3.52	5.55	53.53
Trout	2.68	4.59	4.41	5.98	0.18	0.88
Vanern	5.81	4.91	5.86	2.35	3.14	1.84
Proportion of total load						
Harp	0.55	0.45	1.00	0.66	0.07	0.24
Monona	0.54	0.46	1.00	0.14	0.37	0.50
Toolik	0.87	0.13	1.00	0.27	0.08	0.64
Trout	0.32	0.68	1.00	0.90	0.03	0.10
Vanern	0.55	0.45	1.00	0.50	0.34	0.21

FIGURES
Figure 1.
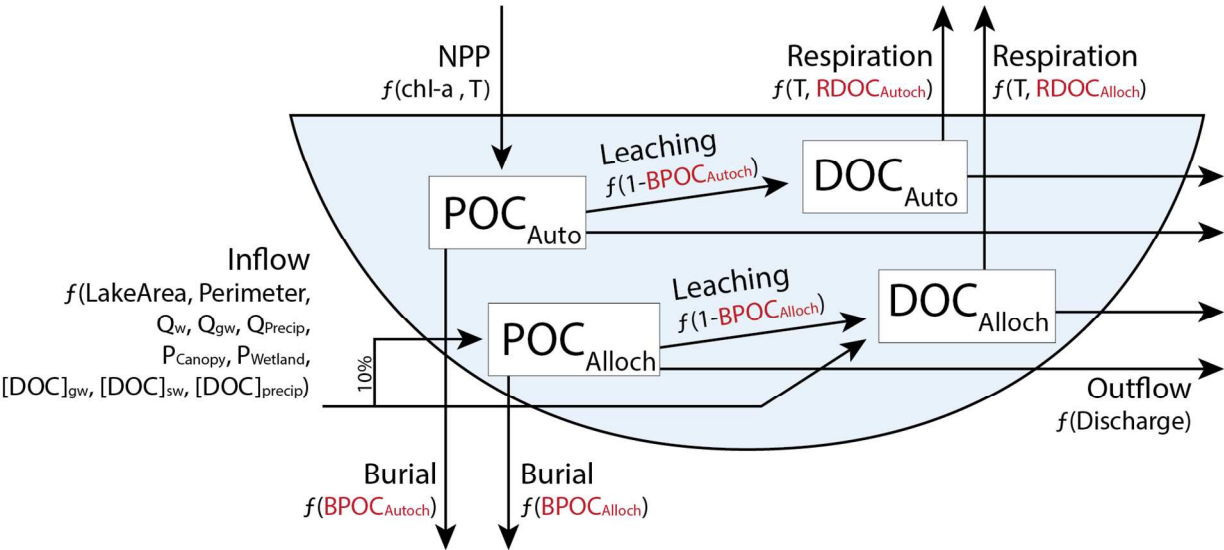
Figure 1. Conceptual diagram of the organic carbon lake model depicting fluxes based on allochthonous (alloch; externally derived) and autochthonous (autoch; internally derived; NPP) inputs of organic carbon, long-term burial, leaching of particulate organic carbon (POC) to dissolved organic carbon (DOC), respiration of DOC to CO_2, and export via outflow. Four parameters ($RDOC_{Alloch}$, $RDOC_{Alloch}$, $BPOC_{Autoch}$, and $BPOC_{Alloch}$) were calibrated for each lake. Parameters and equations are defined in Tables 2 and 3.

Figure 2.
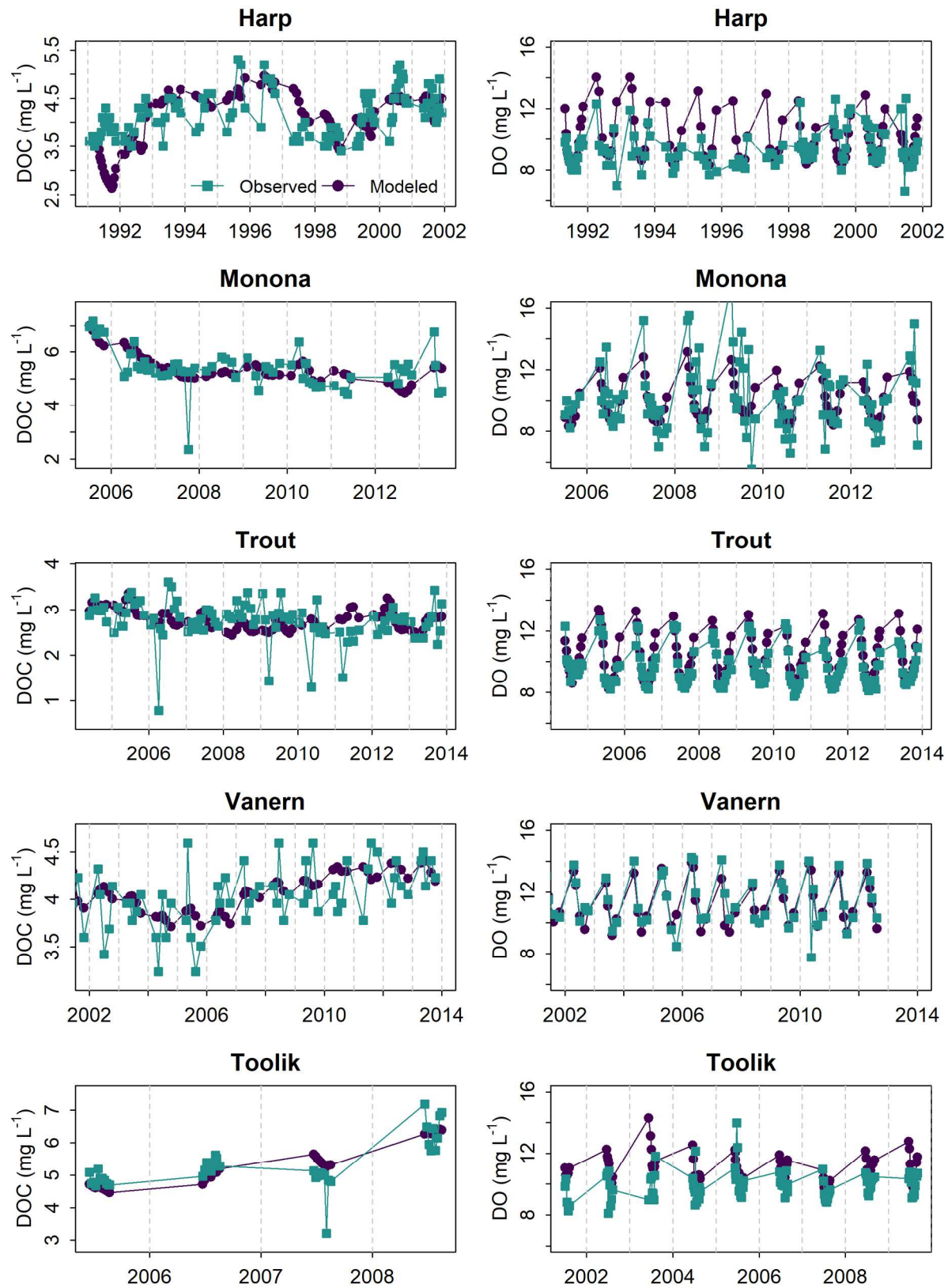
Figure 2. Observed dissolved organic carbon (DOC) and dissolved oxygen (DO) concentrations in each lake (teal squares) compared to modeled concentrations (purple circles) for the same date. For some lakes, years differed between DOC and DO based on availability of observed data (Appendix S1). Toolik data are temporally clustered due to the short ice-free season.

43
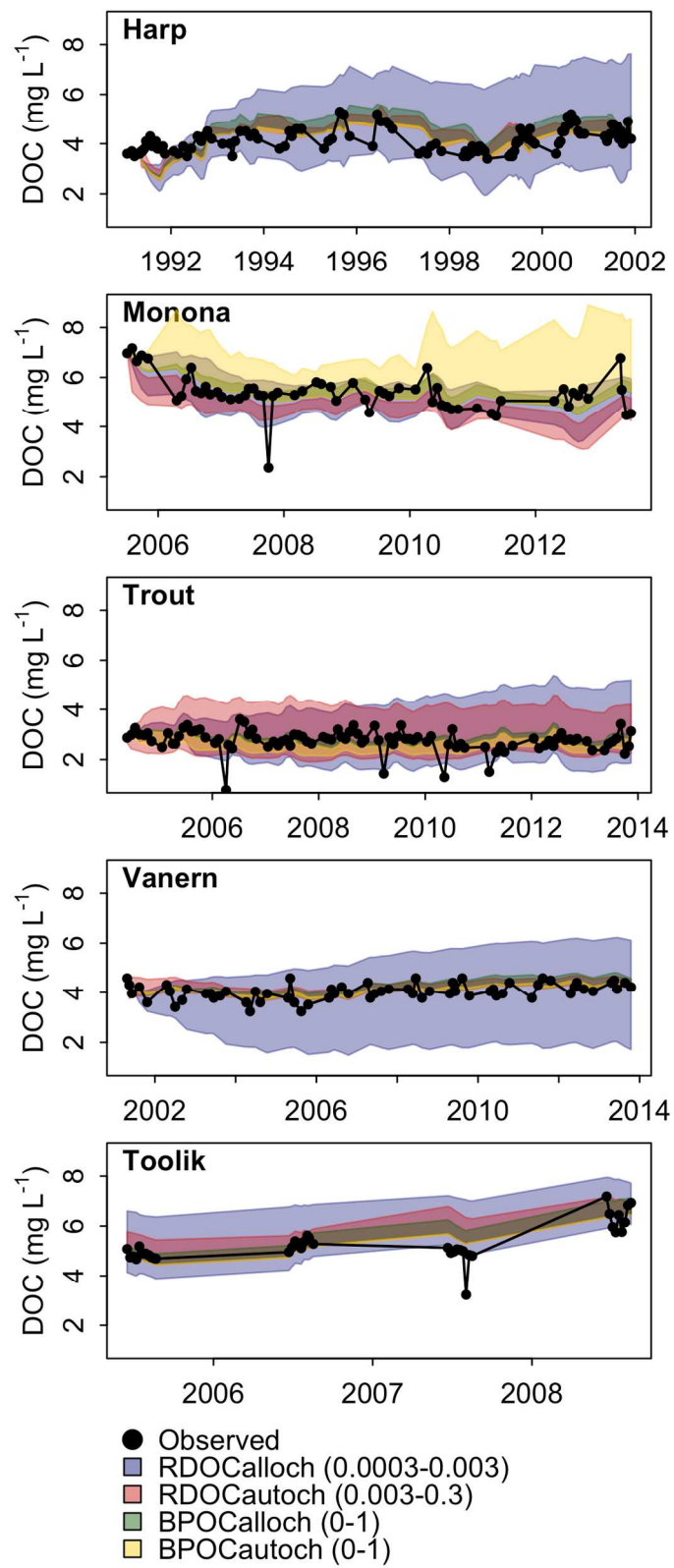
Figure 3.

Figure 3. Sensitivity of modeled dissolved organic carbon (DOC) concentrations to free parameters in the model. Each parameter was varied across a given range (RDOC$_{Alloch}$ 0.0003-0.003 d^{-1}, RDOC$_{Autoch}$ 0.003-0.3 d^{-1}, BPOC$_{Alloch}$, 0-1, BPOC$_{Autoch}$, 0-1) while the other three parameters remained fixed at their calibrated values. Shaded areas represent the range of modeled DOC concentrations as each parameter was varied. Black circles represent observed in-lake DOC concentrations.
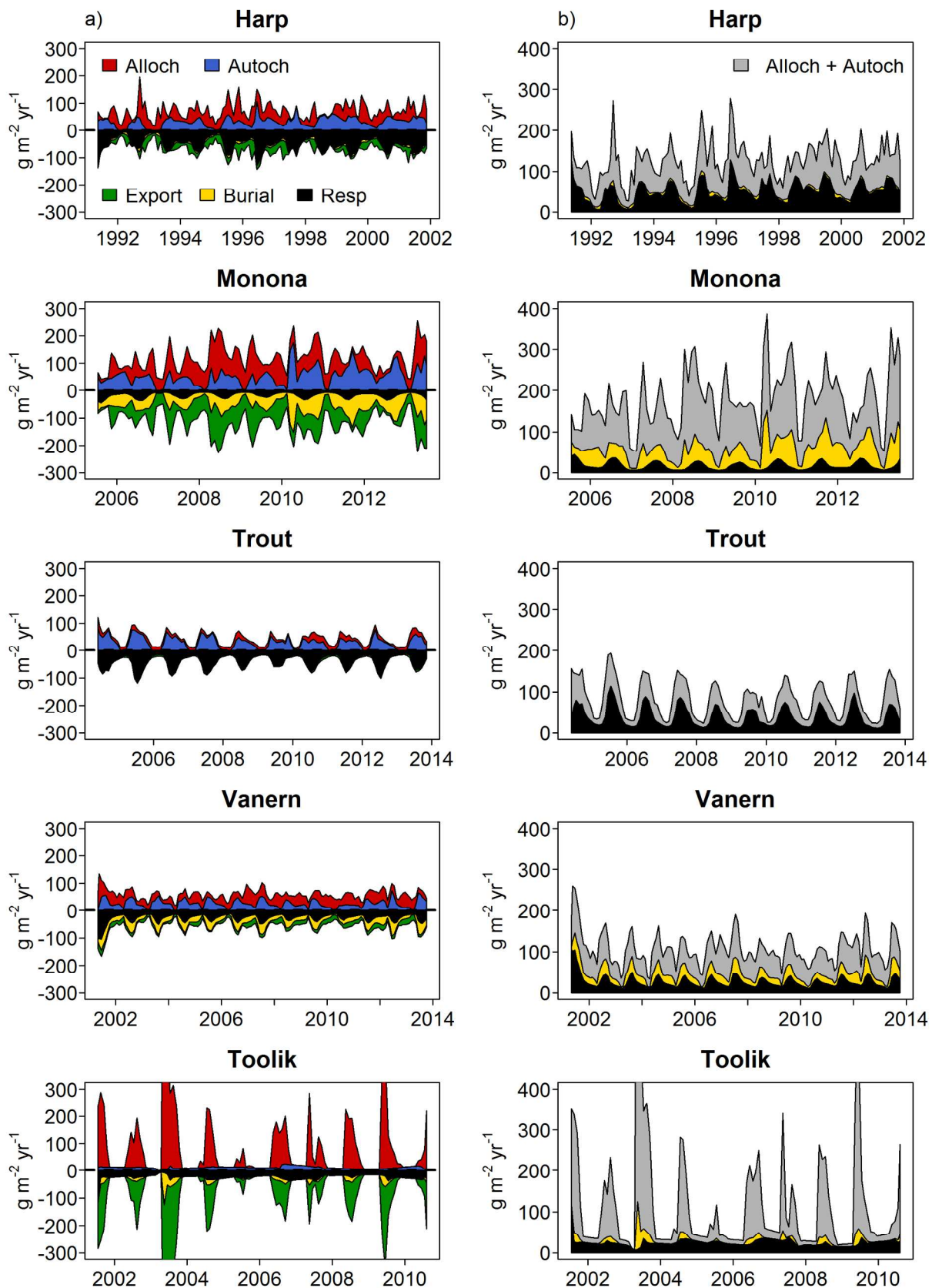
Figure 4.

Figure 4. Time series of organic carbon fluxes and fates. a) Colored areas represent magnitudes of input (allochthonous and autochthonous) and output fluxes (export, burial, and respiration). All lines are stacked to show cumulative magnitudes. b) Absolute values of burial, respiration, and input fluxes. Vertical axes for Toolik Lake plots were truncated to enable visualization of relatively lower fluxes. Maximum autochthony and export for Toolik Lake were 1369 and -848 g $m^{-2} y^{-1}$, respectively (May 2003).

Figure 5.
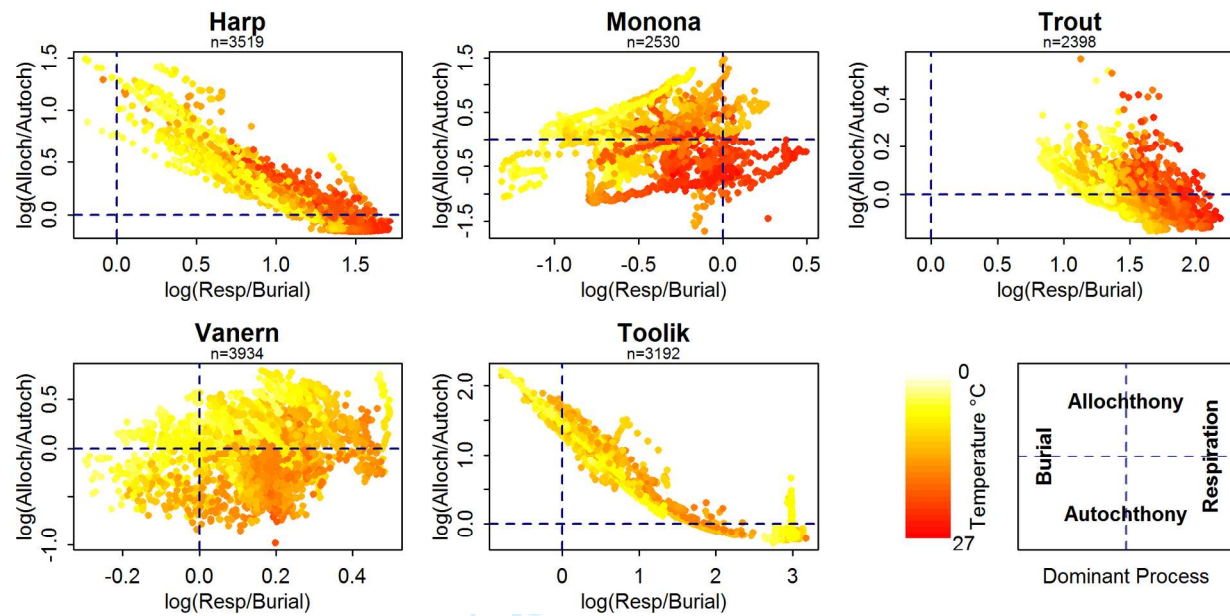
Figure 5. Relationship between $\log_{10}$-transformed allochthony/autochthony and respiration/burial (g m^{-2} yr^{-1}) of organic carbon, colored by epilimnion water temperature. The four quadrants in each figure represent the dominant processes in each lake (Vertical axis: dominant OC fate; horizontal axis: dominant OC input).

AppendixS1

NOTE: moved offline by Ian on 11-12-17
Overview maps of lakes modeled in this study
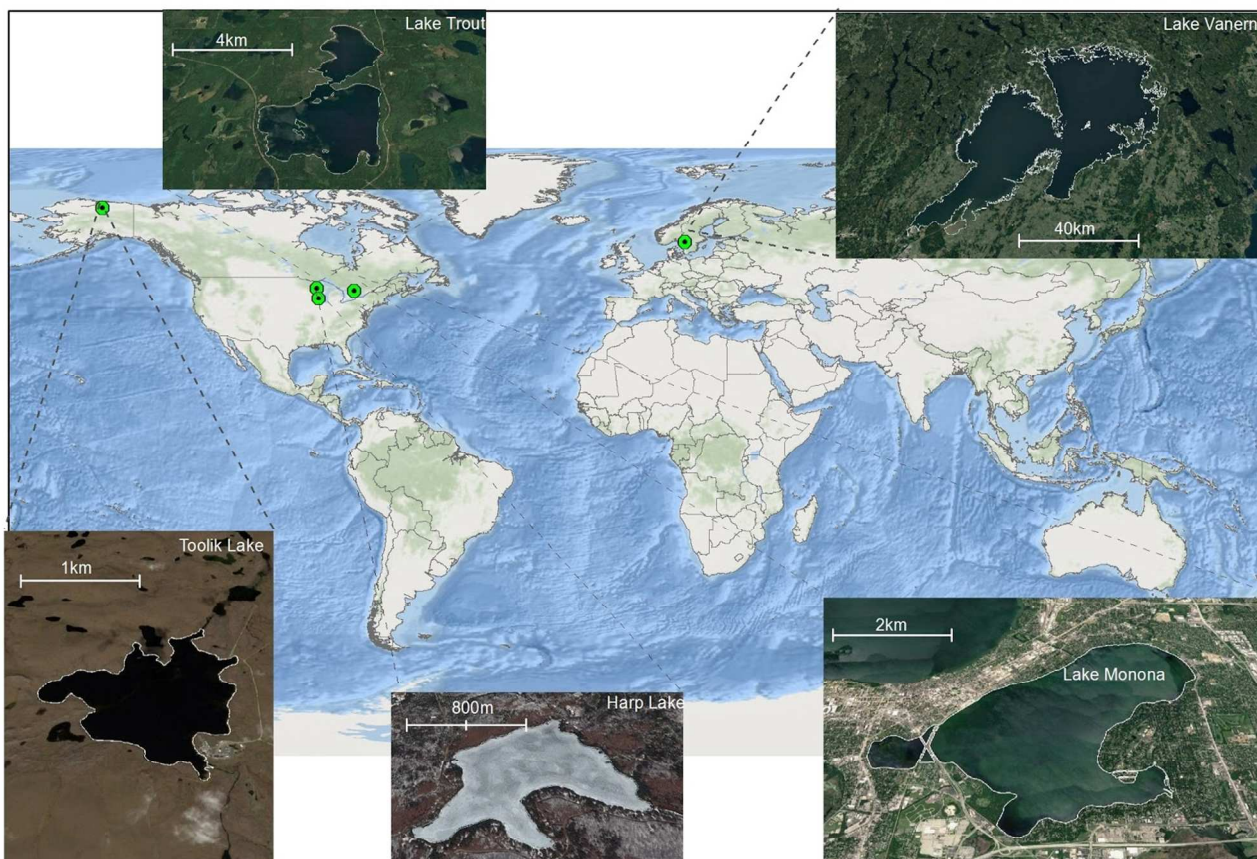
Data sources

Harp Lake

Input model data for Harp Lake were obtained from Ontario Ministry of the Environment and Climate Change Dorset Environmental Science Centre (http://desc.ca/data) Limnological data for the years 1991-2001 were provided. Inflow and lake data were sampled at "HARP INFLOW #3A, #6A, #6, #5, #4, #3 and "HARP LAKE" stations, respectively.
Total inflow DOC was calculated by summing the DOC concentration of each tributary proportionally to the fraction each tributary contributed to total inflow volume. DOC data ("HARP LAKE") from 1991-2001 were used for model validation. Hypolimnion temperature was set to the bottom temperature in the bottom 2 m of the water profile, whereas epilimnion temperature was set to the temperature in the 1-2 m surface water. Precipitation data were obtained from daily weather records for the "HARP LAKE" meteorological station. Sediment mass accumulation rate was inferred from previous studies (Eimers et al. 2006, Mills et al. 2009). A Harp lake shapefile was hand digitized using ESRI aerial photographs (World Imagery)

in ArcGIS (ESRI 2016). PCanopy was estimated using land cover data (15 m resolution) in the Ontario Land Cover Compilation V 2.0 (Ontario Ministry of Natural Resources and Forestry 2014). This dataset showed no adjacent wetlands to Harp Lake. The absence of adjacent wetlands was further verified using aerial photographs (ESRI 2016). As such, PCanopy was set to 1 and PWetland to 0.

Toolik Lake

Input model data for Toolik Lake were obtained from a variety of sources. Limnological data for the years 2001-2010 were freely downloaded from the Arctic LTER website (http://arc-lter.ecosystems.mbl.edu/). Inflow and lake data were sampled at "Toolik Inlet" and "Toolik Main", respectively. Hypolimnion temperature was set to the bottom temperature in the bottom 2 m of the water profile, whereas epilimnion temperature was set to the Temp in the 1-2 m surface water. We made special requests for DOC data (Toolik Main) from 2005-2008 to be used for model validation. This request was generously handled by Dr. George Kling. Precipitation data were obtained from daily weather data records for the Toolik Field Station Meteorological Station. We downloaded a wetlands spatial polygon layer for calculating the percent of the shoreline covered by wetlands (PWetland) from the U.S. Fish and Wildlife Service and National Wetlands Inventory (U.S. Fish and Wildlife Service 2014). Due to the lack of forests at this high latitude, we set PCanopy to 0 and needed no other land cover datasets. We confirmed the lack of tree cover from aerial photography accessed via Arctic LTER. Sediment mass accumulation rate was inferred from previous studies (Cornwell 1985, Whalen and Cornwell 1985). A Toolik lake shapefile was also obtained from Arctic LTER.

A unique challenge associated with Toolik Lake was the relatively short ice-free period each year. Typically, complete input datasets for our model were only available from June - August. Normally, our model would linearly interpolate data gaps, but we sought to avoid interpolating over such a large portion of the year. We used date of ice-off for Toolik Inlet when available (2006-2010) to determine when inflow DOC would be near 0 due to ice. When ice-off dates were not available (2001-2005), we used the 2006-2010 average. Setting inflow DOC to 0 shut down the main input of DOC to the ecosystem and considerably improved model calibration.

Trout Lake

Data for the Trout Lake time series were obtained from a variety of publicly available sources. Water temperature and water chemistry data for the years 2004-2013 was downloaded from the North Temperate Lakes (NTL) LTER data catalog (https://lter.limnology.wisc.edu/researchsite/trout-lake). We defined hypolimnion temperature as the mean temperature below 15 m, whereas epilimnion temperature was the average water temperature above 2 m. Total unfiltered phosphorus (TP) and chlorophyll-a (Chl-*a*) data in the model were each averaged from all sampled depths on each sampling date, which was approximately monthly. Daily water outflow volume data were downloaded from USGS gauge data for the Trout River (USGS Station Number 05357245). While Trout Lake includes multiple surface inflows (Allequash Creek, Mann Creek, Stevenson Creek, North Creek), inflow volume was set equal to surface outflow to maintain constant lake volume. DOC from surface flows was approximated by weighting available surface flow DOC data (https://lter.limnology.wisc.edu/data/filter/32483) for individual streams by flow volume in that stream. Daily precipitation data were downloaded from the Climate and Hydrology Database

Projects website (http://www.fsl.orst.edu/climhy/), using the NTL Minocqua meteorological station.

We obtained lake morphometry parameters from the online NTL Trout Lake profile (https://lter.limnology.wisc.edu/researchsite/trout-lake), and initial concentrations of in-lake DOC were obtained from previously published work on Trout Lake (Hanson et al. 2014, P. Hanson personal communication). Sediment mass accumulation rate was based on the average value of available NTL data for "areal flux of carbon to sediment trap". PWetland for the lake perimeter was estimated using a vector layer obtained from the U.S. Fish and Wildlife Service National Wetlands Inventory (U.S. Fish and Wildlife Service 2014) and the Wisconsin DNR hydrography dataset (Wisconsin Department of Natural Resources 2011). PCanopy was estimated using the WISCLAND land cover raster dataset (30 m resolution) from Wisconsin DNR (Wisconsin Department of Natural Resources 1998). Although we recognize this is a relatively old land cover dataset based on 1993 Landsat imagery, it was only used to estimate forest cover, which is not likely to have changed considerably since 1993.

Model validation in-lake DOC data were obtained from the NTL database (2004-2013) and from unpublished data provided by Mark Gahler (2014; gahler@wisc.edu). Validation data for in lake dissolved oxygen were obtained from the NTL database.

Lake Vänern

Model time series data for Lake Vänern were obtained primarily from publicly available Swedish Meteorological and Hydrological Institute (SMHI; http://www.smhi.se) and Swedish University of Agricultural Sciences (SLU; http://info1.ma.slu.se/db.html) databanks. Water temperature and water chemistry data for the years 2001-2013 were downloaded from the SLU databank (http://info1.ma.slu.se/ma/www_ma.acgi$Project?ID=StationsList&P=V%C4N), and was based on the "Megrundet N" sampling station only. We defined hypolimnion temperature as the mean temperature below 50 m, whereas epilimnion temperature was the average water temperature above 2 m. TP and chl-*a* data in the model were each averaged from all sampled depths on each sampling date, which was approximately monthly during ice-off (Apr-Nov) periods each year. Daily water outflow volume from the single outlet at Göta Älv dam was obtained from SMHI station 1954 (http://vattenweb.smhi.se/station/#). Multiple inflows were set equal to outflow volume to maintain constant lake volume. DOC from surface flows was approximated by as 90% of measured total organic carbon (TOC; P. Hanson personal communication) from all sampled sites on all days data were available (http://info1.ma.slu.se/ma/www_ma.acgi$ProjectP?ID=Intro&P=V%C4NTF). The monitoring site Klarälven Norra Råda was omitted due to distance of sampling site from the lake. Daily precipitation data were downloaded from the SMHI website http://opendata-download-metobs.smhi.se/explore/) using the "Naven A" meteorological station. A data gap from 6/1/2008-7/23/2008 was assumed to be a period of zero precipitation.

We obtained lake morphometry parameters from the SMHI Lake Vänern profile page (http://www.smhi.se/kunskapsbanken/hydrologi/fakta-om-vanern-1.4732), and initial concentrations of in-lake DOC and POC were calaculated from SLU in-lake TOC data on the last date prior to the start of our time series (Oct 1999). Sediment mass accumulation rate was calculated based on previously published data for Lake Vänern sediment linear accumulation rate (Wihlborg and Danielsson 2006) and Lake Mälaren sediment % OC and bulk density data (Håkanson 2004), as those data were not available for Vänern. PWetland and PCanopy for the

lake perimeter were approximated based on whole catchment land use data for "Marsh", "Bog", and "Woodland", respectively, provided by SMHI.

Model validation in-lake DOC data were calculated from SLU TOC data for the Megrundet N station during ice-off season (Apr-Nov 2000-2013). Validation data for in lake dissolved oxygen was obtained from the SLU database for the Megrundet N station during ice-off season (Apr-Nov 2000-2013).

Lake Monona

Inflow discharge data to Lake Monona were downloaded from USGS gauge 05428500. All other time series data for Lake Monona were obtained from from the North Temperate Lakes (NTL) LTER data catalog (https://lter.limnology.wisc.edu/data). Inflow DOC was assumed to equivalent to surface DOC concentrations from upstream Lake Mendota, for which biweekly data were available. A time-series of thermocline depths in Lake Monona was calculated from in-lake temperature profiles via rLakeAnalyzer (Winslow et al. 2016). Epilimnion and hypolimnion temperatures were calculated as the mean temperatures above and below the thermocline depths, respectively. Chl-*a* was calculated as the mean concentration throughout the water column. Other parameter data were acquired from https://lter.limnology.wisc.edu/researchsite/lake-monona and Hanson et al. (2014). The parameters PCanopy and PWetland were estimated using the same spatial data as for Trout Lake.

Other lakes

We additionally acknowledge the efforts of numerous people who provided data for other lakes not ultimately used in this study. In these cases, either we could not obtain all necessary data inputs or a sufficient number of years. Specifically, Hilary Swain of Archbold Biological Station and Evelyn Gaiser provided a long term, curated dataset for Lake Annie, FL. Additionally, several years of discrete nutrient, chlorophyll, temperature, and dissolved oxygen data were provided for West Lake Okoboji, Silver, and Center Lakes in Dickinson County, Iowa, by the Cooperative Lakes Area Monitoring Project (CLAMP).

References

Cornwell, J. C. (1985). Sediment accumulation rates in an Alaskan arctic lake using a modified 210Pb technique. Canadian Journal of Fisheries and Aquatic Sciences, 42(4), 809-814.

Eimers, M. C., Paterson A. M., Dillon P. J., Schiff S. L. ,Cumming B. F. and Hall, R. I. (2006) Lake sediment core records of sulphur accumulation and sulphur isotopic composition in central Ontario, Canada lakes. Journal of Paleolimnology, 35, 99-109.

ESRI, DigitalGlobe, GeoEye, Earthstar Geographics, CNES/Airbus DS, USDA, USGS, AEX, Getmapping, Aerogrid, IGN, IGP, swisstopo, and the GIS User Community. (2016).

Hakanson, L. (2004). Internal loading: a new solution to an old problem in aquatic sciences. Lakes & Reservoirs: Research & Management, 9(1), 3-23.

Hanson, P. C., Buffam, I., Rusak, J. A., Stanley, E. H., and Watras, C. D. (2014) Quantifying lake allochthonous organic carbon budgets using a simple equilibrium model. Limnology and Oceanography 59, 167–181.

Mills, R. B., Paterson, A. M., Jules, M. B., Davis, R. S. L., John, P. S. and Greg, M. (2009). Factors influencing the achievement of steady state in mercury contamination among lakes and catchments of south-central Ontario. Canadian Journal of Fisheries and Aquatic Sciences, 66, 187-200.

Ontario Ministry of Natural Resources and Forestry. (2014). Ontario Land Cover Compilation V.2.0. https://www.ontario.ca/data/ontario-land-cover-compilation-v20.

U.S. Fish and Wildlife Service. (2014). National Wetlands Inventory. U.S. Department of the Interior, Fish and Wildlife Service, Washington, D.C. http://www.fws.gov/wetlands/.

Whalen, S. C., & Cornwell, J. C. (1985). Nitrogen, phosphorus, and organic carbon cycling in an arctic lake. Canadian Journal of Fisheries and Aquatic Sciences, 42(4), 797-808.

Wihlborg, P., & Danielsson, A. (2006). Half a century of mercury contamination in Lake Vanern (Sweden). Water, Air, and Soil Pollution, 170(1), 285-300.

Winslow, L. A., Read, J. S., Woolway, R., Brentrup, J., Leach, T. and Zwart, J. A. (2014). rLakeAnalyzer: package for the analysis of lake physics. *R package version*, *14*.

Wisconsin Department of Natural Resources. (1998). WISCLAND Land Cover (WLCGW930). http://dnr.wi.gov/maps/gis/datalandcover.html.

Wisconsin Department of Natural Resources. (2011). Surface water (hydrography) data. ftp://dnrftp01.wi.gov/geodata/hydro_24k/.