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Dominant processes in organic carbon cycling in lakes revealed by dynamic mechanistic modeling

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## **ABSTRACT**

Lakes are active processors of organic carbon (OC) and are thought to play important roles in landscape and global carbon cycles. OC is produced and consumed during primary production and respiration 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 various fluxes under a dynamic modeling framework to examine their interactions and relative magnitudes. We used our conceptual understanding of lake OC budgets to develop a flexible, dynamical mass balance model for OC, and applied the model to a heterogeneous set of five lakes. We examined the relative magnitudes of OC fluxes and found that long-term lake OC dynamics were predominantly driven by allochthonous loads in four of the five lakes, underscoring the importance of terrestrially-derived OC in lake ecosystems. Burial, the main OC storage mechanism in lakes, represented a relatively small component of the total budget and was driven largely by autochthony. Respiration, the mechanism by which lakes export carbon to the atmosphere, ranged from 14-90% of total OC loads, whereas burial accounted for 7-37% of total OC loads. Additionally, we found that lake OC budgets generally transitioned seasonally from burial- to respiration-dominated as water temperatures and lake productivity increased. Finally, we highlighted critical research needs, which include surface water DOC observations in paired tributary and lake settings, measurements of OC burial rates, and budgets of particulate OC. These additional data will better constrain parameter estimates in future lake OC models and improve our understanding of landscape carbon cycling.

Key words: carbon cycle, carbon flux, limnology, mass balance, allochthony, autochthony, sedimentation, dissolved organic carbon, particulate organic carbon, LTER

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## **INTRODUCTION**

Lakes are dynamic components of the landscape that actively process and transport terrestrially derived organic carbon (OC; Cole et al. 2007, Tranvik et al. 2009, Tanentzap et al. 2017), as well as emit inorganic carbon to the atmosphere (Arvola et al. 2002, Raymond et al., 2013, Weyhenmeyer et al., 2015), making them important in global carbon (C) cycling. Owing to a dearth of ecosystem-scale studies, however, there remains a considerable knowledge gap in lake OC dynamics. Therefore, the use of models, whether steady-state or dynamical mass balance or statistical, is necessary to estimate the importance of lakes to global C budgets, and quantifying dominant processes in lake OC budgets would represent an important step in incorporating lakes into the global C cycle (Hanson et al. 2015). Dynamical mass balance approaches to modeling the internal processing of OC in lakes, which incorporate all critical OC fluxes, are still relatively uncommon. Furthermore, existing mass balances are generally based on low frequency data, confined to single lakes, and tend to be concentrated in boreal regions (Jonsson et al. 2001, Urban et al. 2005, Andersson and Sobek 2006, Cremona et al. 2014). Here we developed and applied a dynamical mass balance model to examine the relative magnitudes of OC fluxes within a heterogeneous set of five lakes.

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

For lakes, the term “mass balance” has been broadly used to quantify budgets as the combination of inputs, internal processes, and outputs. Inputs to lake ecosystem OC budgets are the sum of allochthonous (externally derived) dissolved (DOC) and particulate OC (POC) inflows from surface or groundwater sources, precipitation, and litterfall. Internal processes consist of autochthonous (in-lake) mechanisms that produce, consume, and store OC (i.e., primary production, respiration, burial in sediments). Outputs include DOC and POC that exit a lake via surface or groundwater exports. The balance of inputs, internal processes, and outputs determine the fate of OC in lake ecosystems and the role of lakes in broader C cycling. (Box 1, Fig. 1).

At the global scale, lakes are thought to be net sources of carbon to the atmosphere based on the super saturation of carbon dioxide (Kortelainen et al. 2006, Tranvik et al. 2009, Raymond et al. 2013) and methane (Bastviken et al. 2011) in lake surface waters. OC export is less frequently considered, but equally important, both in terms of burial and the quality and quantity of OC ultimately reaching the ocean via tributaries (Raymond and Bauer 2001, Santoso et al. 2017). Because lakes store OC in sediments, they act as sinks in the global carbon cycle (Mulholland and Elwood 1982, Dillon and Molot 1997, Einsele et al. 2001, Einola et al. 2011).

A limitation of many existing mass balance studies is the omission of key fluxes, such as burial, which contribute to the full budget, which underscores the importance of a unified model framework (Hanson et al. 2015). We synthesized existing knowledge of lake OC budgets into an applied model that integrates these important mechanisms, including both in-lake as well as external (i.e., watershed) processes (Fig. 1). Below we described these processes in three main categories 1) allochthony, 2) autochthony, and 3) storage and export.

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

*OCALLOCHTHONOUS:* surface and groundwater inflows + litterfall + atmospheric deposition

*OCAUTOCHTHONOUS*: gross primary production - autotrophic respiration - heterotrophic respiration

*Full budget*: *OCALLOCHTHONOUS* + *OCAUTOCHTHONOUS* = respiration + burial + export+ ΔOC storage (in water column)

### *Allochthony*

Allochthonous inputs include all terrestrially derived OC, including DOC and POC from surface and groundwater inflows, litterfall, and direct-fall precipitation (Box 1). Although surface water inflows regularly deliver DOC to lake ecosystems, the role of terrestrially derived OC is perhaps the most commonly overlooked set of processes in OC budgets, largely owing to data limitations (Hanson et al. 2015). Prior studies have included direct measurements of inflow stream concentrations of DOC when available (Schindler et al. 1997, Jonsson et al. 2001, Urban et al. 2005, Klump et al. 2009), but other approaches have included use of literature-derived input estimates (Striegl et al. 1998), equations based on watershed area (Sobek et al. 2006), watershed-wetland area ratios (O’Connor et al. 2009), precipitation (Hanson et al. 2004, Staehr et al. 2010), or GIS-based estimates based on land cover and distance-weighted hydrological flow paths (Canham et al. 2004). In lakes without surface inflows, including closed-basin and seepage lakes, groundwater can be the dominant hydrological input (e.g., Gaiser et al. 2009) and can deliver DOC to lakes, especially in organic-rich soils (Schindler and Krabennhoft 1998). Empirical measurements of groundwater discharge and DOC concentration, however, are rare and difficult to estimate (Hanson et al. 2014). POC inputs from litterfall, and wet and dry atmospheric deposition are typically small and are generally estimated as a function of lake size and literature- or expert-based loading coefficients (Hanson et al. 2004).

### *Autochthony*

Autochthonous DOC and POC originate within lakes through photosynthesis by primary producers. Since gross primary production (GPP) is difficult to measure at the ecosystem level, net primary production (NPP), considered the difference between GPP and autotrophic respiration, is typically measured instead (Pace and Lovett 2013) (Box 1). Approaches previously employed to estimate NPP include using bottle incubations (Urban 2005, Yang et al. 2008), and more recently high frequency measurements of dissolved oxygen or carbon dioxide concentrations (Cole et al. 2002, Staehr et al. 2010). Statistical relationships have also been built to estimate NPP from lake temperature and total phosphorus (TP) (Hanson et al. 2004) or chlorophyll-a (chl-*a*) (Jonsson et al. 2001, Ramlal et al. 2003), or static proportions of the overall OC pool (Aberg et al. 2004).

*Storage and export*

Long-term burial of POC in lake sediments is the only mechanism by which lakes permanently remove carbon from the global carbon cycle, and is therefore a critical flux in our understanding of sink dynamics of both allochthonous and autochthonous POC (Cole et al. 2002, Tranvik et al. 2009). Permanent POC burial in lakes is a product of in-lake POC concentrations, POC particle sizes and associated settling rates, sediment particle size and density that affect resuspension, lake hydrodynamics that affect settling rates and resuspension, and benthic biogeochemistry (Downing et al. 2008, Xu et al. 2013). Methods for estimating sediment accumulation rates are diverse and have commonly included functions based on lake area, particularly for studies containing multiple lakes (Canham et al. 2004, Hanson et al. 2004). Previous studies have estimated burial rates using sediment cores (Yang et al. 2008, Klump et al. 2009, Heathcote & Downing 2012), sediment traps (Jonsson et al. 2001, Ramlal et al. 2003), or estimates based on changing bathymetry (Downing 2008). A challenge associated with estimating accumulation rates is the reliance on point measurements to characterize sediment accumulation rates that can vary dramatically over both space and time. Allochthonous and autochthonous POC that is not buried is leached in the form of DOC and exported via surface or groundwater (Cole et al. 1984). In some lakes, although not included in this study, there is carbon efflux from lakes in the form of insect production and emergence (Vander Zanden and Gratton, 2011). Exports represent allochthonous inputs to downstream aquatic ecosystems and therefore contribute to landscape C cycling (Kling et al. 2000).

### *Objective and research questions*

Our broad objective was to develop a flexible OC model for lakes that represented long-term dynamics and magnitudes of key DOC and POC fluxes, and to use that model both to reveal key drivers of OC fates (burial, respiration and export) in lakes as well as uncertainties associated with these processes. The balance between burial and respiration is particularly important, given that these represent the pool of internally processed OC that may be stored in lakes or mineralized. Here, we applied a dynamical mass balance model that incorporates the OC fluxes described above, parameterized for five lakes that span a gradient of morphological characteristics. Quantification of these OC pools would considerably enhance our understanding of the role of lakes in landscape C cycling. We applied the model to address the following questions:

1. What are the relative magnitudes of varying fluxes that drive lake OC budgets?
2. Which mechanisms represent the greatest source of uncertainty in our understanding and modeling of C budgets?

## **METHODS**

### *Study lakes and data sources*

We dynamically modeled OC budgets for five lakes that span a range of environmental conditions and limnological characteristics (e.g., hydrologic residence time, depth; Table 1). Lakes were selected based on the availability of observational data. Required observational data included precipitation, hydrological inflow (discharge), inflow DOC concentration and various in-lake measurements (surface temperature, chl-*a*, and Secchi depth). All lakes had a minimum of 10 years of limnological data used for model training (Table 1) and at least four years of in-lake DOC and DO measurements for model validation. See supplemental material for detailed data descriptions and sources (S2).

Our five study lakes were heterogeneous in terms of morphological and water quality characteristics (Tables 1-2). Lakes ranged from 71.38 ha (Harp Lake, Canada) to 565000 ha (Lake Vanern, Sweden) and mean depths ranged from 7 m (Toolik Lake, USA) to 27 m (Vanern). Hydrologic residence time ranged 0.8 (Lake Monona, USA and Toolik) to 6.3 (Vanern) years. Our dataset included four oligotrophic lakes and one eutrophic lake (Monona). In-lake mean annual DOC concentrations ranged 3 g m-3 (Trout Lake, USA) to 6 g m-3 (Monona). Watersheds are primarily forested for Harp, Trout, Vanern, whereas Toolik occurs in a tundra-dominated watershed and Monona occurs in an agricultural and heavily developed watershed. Although our lakes span a set of relatively wide environmental gradients, we did not include a formal analysis of OC fates along these gradients due to the small sample size of lakes.

*General model approach*

Limited observational data hindered our ability to build an overly complex process model for all of our study lakes. We therefore aimed to build a parsimonious model to maintain generalizability across lake ecosystems, using our conceptual framework as the foundation for the mass balance model (Fig. 1). We operated the model on a daily time step. Lake volume was assumed static and inflow volume was assumed equal to outflow volume. Because daily observational data of inflow DOC, in-lake chl-*a*, Secchi depth, and temperature were not always available (i.e., weekly to bi-weekly for some lakes), we gap-filled to a daily time step using linear interpolations, except for precipitation, which was assumed zero for missing data; however, precipitation data gaps were rare. Daily evaporative losses were assumed to be negligible and were not accounted for in the model. To account for the absence of winter data at Toolik, we set inflow DOC to 0 when the main inflow (Toolik Inlet) was frozen (S2). We summarized static and calibrated model parameters in Table 2. The model was written in R version 3.3.2.

### *Allochthonous DOC and POC*

Allochthonous DOC load (DOCalloch) was calculated as a combined function of 1) stream load, 2) precipitation load, 3) wetland load, and 4) groundwater load. Observational data for POCalloch (5) was not available and was estimated as the sum of an aerial load based on canopy cover and 10% of DOCalloch (Hanson et al. 2014).

1) Daily surface water inflow discharge (m3 s-1) was generally available for the main tributaries of all lakes. Inflow DOC concentration was less frequently measured, and was interpolated to a daily timestep from sub-weekly to monthly data (S2). When inflow DOC concentrations were not available for all tributaries, DOC contributions for each tributary were estimated based on the proportion of total inflow volume and the assumption that inflow DOC concentration was equal across all tributaries. Inflow DOC load (DOC SW; g d) was calculated as a product of discharge volume and measured concentration (Table 3: Eq. 1a).

2) DOC loads from precipitation were calculated as the product of lake surface area (m2) and daily precipitation (mm) measured at the weather station nearest to each lake (Table 3: Eq. 1b-c). The concentration of DOC in precipitation was set to 2 g m-3 (Hanson et al. 2014) (Table 2: DOC\_precip\_conc).

3) Contributions of DOC from shoreline-adjacent wetlands were estimated using GIS and publicly available spatial datasets (S2). We focused on wetlands adjacent to the shoreline because they contribute most of wetland-derived DOC to lakes; distant wetlands contribute negligible DOC (Hanson et al. 2014). DOC contribution of wetlands was calculated by multiplying the proportion of lake shoreline covered by wetlands (Table 2: PropWetland) by lake perimeter (m), and then multiplying this value by a static parameter representing wetland DOC in g m-1 shoreline d-1 (Table 3: Eq. 1d). To account for potentially misaligned topology among spatial wetland data and lake boundaries, we defined adjacency as within 30 m of lake boundaries.

4) Lake-specific groundwater inflow volume (m3) and DOC concentration (g m-3) were not available for any lake. The proportion of inflow as groundwater in our study lakes ranged from 0-19%; we used literature values when available, but assumed no groundwater inflow in the absence of data (S2). Groundwater DOC concentration was assumed to be 10 g m-3 (Table 2: DOC\_GW, Hanson et al. 2014). The contribution of groundwater to the allochthonous DOC load was therefore calculated as the product of DOC concentration and a percentage of total surface inflow (groundwater inflow rate, Table 3: Eq. 1e).

5) We calculated the contribution of all adjacent forests, including coniferous, deciduous, and mixed stands (POC canopy), to the OC budget via litterfall by multiplying the proportion of lake shoreline covered by forests (Table 2: PropCanopy) by lake perimeter (m), and multiplying this value by an aerial POC rate of 1 g m-1 shoreline-1 day-1 (Table 3: Eq. 1f, Hanson et al. 2014). Adjacency was defined in the same manner as for PropWetlands.

### *Autochthonous DOC and POC: primary production*

Autochthonous OC production as GPP was modeled as a function of chl-*a* (µg L-1) and surface water temperature (°C) per Morin et al. (1999, Table 3: Eq. 2a). This empirical statistical model was based on observational temperature and chl-*a* data that ranged from 5-25 °C and 1-1000 mg m-2, respectively. Since models of GPP are not well constrained at low temperatures, we set GPP to zero if surface water temperatures were < 4 °C, as this is the maximum density of water and a point at which lakes would no longer be stratified. Chl-*a* concentrations were converted from volume to areal units by multiplying by photic depth, which was estimated from Secchi depth (m; Wetzel 2001). Autotrophic respiration (Table 2: R\_autotroph) was assumed to equal 80% of total GPP; therefore, net primary production (NPP) was set equal to 20% of GPP (Quay et al. 1986, Cole et al. 2002, Hanson et al. 2004). NPP was partitioned into DOCauto and POCauto fractions using the Pace and Prairie (2005) negative exponential equation to estimate the percentage of NPP that is respired and converted to DOC as a function of chl-*a* (Table 3: Eqs. 2b-2d). Oligotrophic lakes will generally respire a greater percentage of NPP than eutrophic lakes (Pace and Prairie 2005). The resulting rates of DOCauto and POCauto (mg m-2 d-1) production were then applied to the whole-lake carbon balance by scaling with lake surface area (Table 3: Eqs. 2e-f).

Heterotrophic respiration was calculated as a function of DOCauto and DOCalloch concentration (g m-3) in the photic zone, epilimnion temperature (assumed to be uniform through the photic zone), and two calibrated parameters: Respiration\_autoch and Respiration\_alloch (Table 2, Table 3: Eqs. 2g-h). We determined epilimnion temperature by averaging observed temperatures throughout the photic zone when data were available, but otherwise used surface temperature (S2). Respiration\_alloch was constrained between 0.0003 and 0.03 (d-1) based on the range of OC decomposition rates for inland waters with residence times between 1-10 years presented in Catalan et al. (2016). Respiration\_autoch was constrained between 0.03 and 0.3 (d-1).

### *Burial of POC*

At each time step, a proportion of POCauto and POCalloch was was removed from the lake via export, and the rest was either buried based on the Burial\_autoch and Burial\_alloch parameters (Table 2, Table 3: Eqs. 3c-h) or leached to DOCauto. Burial parameters were calibrated in the model as a proportion between 0 (no burial of POC) and 1 (all POC is buried). This simple approach allowed burial to function as the expected feedback mechanism to high POC loads from inflows and/or primary production.

### *Model output and calibration*

All fluxes and loads of DOC and POC were tracked at a daily time step. Net ecosystem production (NEP) was calculated as the difference between NPP and heterotrophic respiration (Table 3: Eq. 4a). Atmospheric flux rate (Fatm) was calculated as k × (DOconc - DOsat)/Zmix. The mixed depth (Zmix) was set equal to half the photic depth (m) and the piston velocity (k) was set at 0.7 m d-1 (Table 3: Eq. 4b) The saturation of DO (DOsat) is temperature dependent and was determined using the Garcia-Benson method in the LakeMetabolizer R package (Winslow et al. 2016). NEP, Fatm, and DO were used to calculate the change in DO in the surface water at each time step owing to respiration (Table 3: Eq. 4c).

The four calibrated parameters in the model (Respiration\_autoch, Respiration\_alloch, Burial\_autoch and Burial\_alloch; Table 2) were fit by minimizing the sum of the squared residuals of DOC and DO modeled minus DOC and DO observed (equally weighted by number of observations). The model was fit using a pseudo-random search algorithm in the R package FME (Soetaert and Petzoldt 2010). Modeled DOC and DO were compared to corresponding observed concentrations using root mean square error (RMSE) and Nash-Sutcliffe efficiency scores for each lake to determine model goodness of fit. A sensitivity analysis of each parameter was conducted by allowing the parameter to vary at 100 different values within the set bounds while fixing the other three parameters at their calibrated values.

*Bootstrapping*

We assessed parameter uncertainty using a bootstrapping routine. Residual errors between observed and modeled DOC as well as observed and modeled DO were calculated. We created 100 pseudo-observational data sets by adding randomly selecting residuals to the original observational data. The model was then re-fit to the pseudo-observational data sets to provide 100 new parameter estimates.

## **RESULTS**

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

Modeled DOC and DO generally followed observed temporal patterns across years and study lakes (Fig. 2). There was an overall strong goodness of fit for both DOC and DO based on RMSE across lakes (0.73-1.46 mg L-¹; Table 4). The model also accounted for considerably more information than long-term means of DOC based on NSE values across lakes (0.70-0.96). Although the model captured annual and seasonal DOC and DO dynamics, the model did not consistently characterize the magnitude of short-term spikes in DOC (i.e., days to weeks). Nonetheless, long-term model performance indicated the ability to account for lake variability in DOC and DO from seasonal to inter-annual time scales.

Burial\_alloch, which represented the daily proportion of POCalloch buried in sediments, was 1 (i.e., 1=100% burial, 0% leached to DOC) for all lakes (Table 2). Similarly, Burial\_autoch was near 1 for Monona, Vanern and Toolik; however, values were 0 for Harp and Trout, indicating nearly all autochthonous POC was leached to DOC in these two lakes and subsequently mineralized or exported. DOC respiration parameters were lower for DOCalloch than DOCauto across all lakes, indicating that DOCauto was more readily respired than DOCalloch. Respiration\_alloch was relatively similar across lakes, ranging 0.001-0.003, whereas Respiration\_autoch was more variable, ranging 0.015 (Trout) to 0.297 (Toolik).

Modeled DOC (mg L-1) was generally most sensitive to Respiration\_alloch, except for Monona, for which modeled DOC was most sensitive to Burial\_autoch (Fig. 3). The other 4 lakes were minimally affected by changes in this parameter (< 1 mg L-1 difference across the range of parameter values). Changes in Burial\_alloch had consistently minimal effects on the range of modeled DOC across lakes. Harp and Monona were the only lakes with considerable sensitivity to Respiration\_autoch. Overall, parameter sensitivity was greatest for Vanern, Harp and Monona, for which modeled DOC varied as much as 5-6 mg L-1 across the range of parameter values (Fix 3.). Conversely, modeled DOC varied no more than 2 and 3 mg L-1 for Toolik and Trout, respectively. A bootstrapping of model residuals revealed variable uncertainty in parameters between lakes, with Lake Monona being a clear outlier (Fig S1?). Monona had large uncertainties in all parameters, and DOC respiration parameters often fell at the lower and upper boundary constraints. All other lakes had more narrowly constrained parameters that fell within boundary constrains, with the exception of Vanern where the labile DOC respiration and burial factors had large variability within bounds.

### *Summary of fluxes and fates*

With the exception of Trout, OC loads (based on complete years only) were primarily driven by allochthony, underscoring the importance of terrestrially derived OC in overall lake budgets (Table 5). Trout had the smallest OC load of the five lakes (41.5 g m-2 yr-1) and was the only lake driven primarily by autochthony (67.8%) rather than allochthony (32.2%). Trout processed 90.5% of its load, which was the greatest among all lakes (9.5% exported), through greater respiration (37.5 g m-2 yr-1) than burial (1.2 g m-2 yr-1). OC inputs to Harp averaged 71.9 g m-2 yr-1, predominantly driven by allochthony (55.5%) rather than autochthony (44.5%). Harp processed 75.5% of its load (24.5% exported via surface water) through respiration (47.5 g m-2 yr-1), whereas a smaller fraction was processed through burial (5.3 g m-2 yr-1). Monona had the largest OC load of the five lakes (119.0 g m-2 yr-1) and was also driven primarily by allochthony (54.5%) rather than autochthony (45.6%). Monona processed 49.6% of its load (50.4% exported) through greater burial (43.8 g m-2 yr-1) than respiration (1.68 g m-2 yr-1). Vanern had the second lowest OC load (58.3 g m-2 yr-1) and was driven predominantly by allochthony (55.3%) rather than autochthony (44.7%), which were similar ratios to Harp and Monona. Vanern processed 79.2% of its load (20.8% exported) through greater respiration (29.2 g m-2 yr-1) than burial (19.9 g m-2 yr-1). Toolik had the intermediate OC load (76 g m-2 yr-1) and was driven mostly by allochthony (87.1%) rather than autochthony (12.9%), which was a larger ratio than the other lakes. Toolik processed 36.4% of its load, which was the lowest among all lakes (63.6% exported), through greater respiration (23.7 g m-2 yr-1) than burial (6.7 g m-2 yr-1).

Seasonal patterns in OC fluxes were consistent across the entire time series, with autochthony and respiration increasing to a summer maximum (Fig. 4) Respiration consistently exceeded burial in all lakes but Monona across all modeled years (Fig. 4, Table 5); on average, respiration accounted for 14.1-90.2% of total OC loads, whereas burial accounted for 7.4-36.8% of total OC loads. Trout and Harp exhibited the largest differences between respiration and burial. Monona was the only lake dominated by burial long-term; on average, burial rates were greater than twice respiration rates across modeled years (Fig. 4, Table 5).

### *Seasonal fates*

As water temperatures increased during the growing season (e.g., May – Aug.), the balance between allochthony and autochthony generally shifted in favor of autochthony due to increases in NPP, whereas the ratio between respiration and burial generally shifted towards respiration (Fig. 5). There was seasonal variability in the dominant fluxes acting in each lake. Trout remained dominated by respiration for the entire year across all sampled years, but respiration increased relative to burial as the growing season progressed. Harp, Toolik and Vanern were dominated by burial early in the growing season, but became dominated by respiration on an annual basis as temperatures warmed. Conversely, Monona remained dominated by burial throughout most of the year and respiration only exceeded burial late in the growing season. Despite this late pulse of respiration, Monona remained dominated by burial on an annual basis (Table 5). Monona and Vanern also showed less of an increase in autochthony as the growing season progressed compared to other lakes, suggesting the importance of continued allochthonous inputs during summer months coinciding with increases in autochthony.

## **DISCUSSION**

### *Capturing lake processes*

Our results indicated that a relatively simple, dynamical model can recreate long-term trends in DOC and represent the set of key biogeochemical, trophic, and landscape processes that combine to determine the fate of OC in lake ecosystems. While postulated previously (Hanson et al. 2011), this is the first demonstration of a dynamical model applied to contrasting lake types. Further, the fluxes we modeled were within the range of other published studies for these lakes based on steady-state models. Using much of the same LTER data, Hanson et al. (2014) provided similar estimates for Trout of allochthony (Hanson estimate: 15.92 g m-2 yr-1, our model estimate: 13.35 g m-2 yr-1), burial and export, but not for respiration, because that study did not account for autochthony. Whalen and Cornwall (1985) modeled Toolik based on a different set of years from our study (1980-1981), but similarly demonstrated that the system contained high allochthony relative to autochthony, low burial and high export. Similar to Hanson et al. (2014), Dillon and Molot’s (1997) estimates for Harp of autochthony, burial (Dillon and Molot: 6.3 g m-2 yr-1, our model estimate: 5.3 g m-2 yr-1) and export (Dillon and Molot: 16.9 g m-2 yr-1, our model estimate: 17.6 g m-2 yr-1), but not respiration (based on field data from 1981-1989), were comparable to our results. Although our results generally agreed with prior studies based on steady-state models, our estimated respiration rates were generally higher than those noted in the literature (Hanson et al. 2014, Dillon and Molot 1997). We offer that dynamical models better represent these processes by accounting for seasonal changes in temperature and chl-*a* concentrations. Therefore, although steady-state models may be sufficient for recreating some key ecological processes, dynamical models are needed for determining the relative magnitudes of OC fates in lake ecosystems, given the importance of autochthony and respiration.

*OC fates and global change implications*

Although prior studies have identified lakes as OC sinks owing to burial in lake sediments, our study showed that burial can be a relatively small component of overall lake OC budgets. We demonstrated how rates of export, respiration, and burial can be constrained by mass balance and the inclusion of both allochthonous and autochthonous sources. A key consideration is that our model buried 100% of POCalloch (Table 2: Burial\_alloch); therefore, any increase in POCalloch would be directly proportional to increases in burial. Owing to lack of observational data, we additionally assumed POCalloch was 10% of DOCalloch. Intense precipitation can increase POC concentration disproportionately to DOC concentration in streams (Jeong et al. 2012, Dhillon and Inamdar 2013), which could temporarily increase POCalloch and thus burial. In addition, our model demonstrated DOC responses to precipitation events when we had corresponding weather and DOCalloch data, but the assumed linear changes in inflow volume and DOCalloch between observation points may also have underrepresented DOCalloch, leading indirectly to underrepresentation of POCalloch. Wet years increase DOCalloch inputs to lakes at regional scales (Rose et al. 2016) and therefore probably also increase POCalloch. Although our burial estimates were somewhat uncertain due to underrepresentation of POCalloch during precipitation events, burial would have to increase substantially over the course of the entire modeling period for burial to dominate over respiration, including three-fold or greater for Harp, Toolik and Trout (Table 5). Therefore, missing POCalloch may constitute noise in the overall OC budget, but is unlikely to account for enough OC to exceed the magnitudes of respiration.

Across all lakes in our study, warm surface temperatures were generally associated with autochthony in productive lakes (Monona, Vanern) and allochthony when the OC pool was dominated by allochthonous inputs (Harp, Trout, Toolik; Fig. 5). In lakes with primarily allochthonous OC pools, the ratio of respiration to burial decreased as allochthony decreased. More productive systems had the opposite response, however, and revealed an increase in the ratio of respiration to burial as allochthonous inputs decreased. Across lakes, warm surface temperatures appear to drive lakes toward OC export rather than burial, though the mechanisms driving this trend likely differ among systems dominated by allochthony versus autochthony. The implications of this finding are that climate warming and associated increases in lake surface temperatures will not have consistent effects across all lakes, but instead may favor net heterotrophy and export of terrestrial OC from some systems, and autochthony and export of labile OC from others. Lakes have become increasingly productive under recent climate warming (Kraemer et al. 2016), which increases autochthony. As climates continue to warm, we would therefore expect lake OC budgets to become more respiration-dominated over time, but ratios between respiration and burial will be mediated by ratios between allocthony and autochthony.

### *On-going research needs and future implications*

Our work is an important advance in terms of quantifying the fates of OC across aquatic ecosystems; however, we encountered constraints associated with current data availability. If necessary data were collected for a larger number of lakes spanning wider environmental gradients (e.g., climate, watershed conditions), contributions of lakes to landscape carbon cycles based on lake function could be estimated at broad spatial scales. Particularly necessary are high-frequency measurements of inflow DOC concentration. Our study generally used data collected at weekly or bi-weekly intervals; although data collection may be expensive and logistically challenging, the increasing availability of automated, high-frequency sensor equipment may alleviate long-term costs associated with sensor deployment and manual data retrieval (e.g., Porter et al. 2009). In addition, relatively little is currently known about POC budgets despite their key interactions with DOC (Einsele et al. 2001); we need more POC observational data for incorporation into dynamical models of OC, particularly in inflows for estimating POCalloch. Such studies would help constrain POC parameters and improve estimates of the fates of POC within overall OC budgets. Thus, a key implication of our study is the need for more observational data, particularly pertaining to surface water DOC concentrations, POC cycling, and burial rates. Lake resource managers should be aware of the importance of allochthonous inputs derived from watershed sources in lake OC budgets; allochthony exceeded autochthony in 4 of the 5 lakes in this study. More broadly, development of a flexible model that quantifies OC fates across heterogeneous lake ecosystems represents an important step in better incorporating lakes into the global C cycle.

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**Tables**

**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 |
| **Lake Area (ha)** | 71 | 1326 | 149 | 1610 | 565000 |
| **Data years** | 1991-2001 | 2003-2014 | 2001-2010 | 2004-2013 | 2001-2013 |
| **zmean (m)** | 12 | 8.3 | 7 | 14.6 | 27 |
| **RT (yr)** | 2.5 | 0.8 | 0.8 | 5.9 | 6.3 |
| **Trophic status** | oligotrophic | eutrophic | oligotrophic | oligotrophic | oligotrophic |
| **Secchi (m)** | 4.3 | 2.7 | 4.7 | 5.3 | 4.5 |
| **Chl-*a***  **(µg 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 |
| **References** | Yao et al. 2011 | NTL LTER | Kling et al. 2000 | Webster et al. 1996, NTL LTER | Kvarnäs 2001 |

zmean = mean depth, RT = hydrologic residence time, Secchi = Secchi depth, Chl-*a* = chlorophyll-a, SW DOC = inflow dissolved organic carbon, Lake DOC = in-lake DOC (mean water column). All values calculated from model calibration data or derived from cited references. NTL LTER = [https://lter.limnology.wisc.edu/](https://lter.limnology.wisc.edu/datacatalog/search). Secchi, Chl-*a*, and DOC measurements are the mean values for the data used in this study.

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

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| **Parameter** | **Description** | **Harp** | **Monona** | **Toolik** | **Trout** | **Vanern** |
| **General** |  |  |  |  |  |  |
| Perimeter (m) | Lake perimeter | 4000 | 35200 | 8104 | 25900 | 2007000 |
| Mean Depth (m) | Mean water depth | 12 | 8.3 | 7 | 14.6 | 27 |
| Area (m²) | Lake surface area | 7.14E+05 | 13.3E+07 | 1.49E+06 | 1.61E+07 | 5.65E+09 |
| Volume (m³) | Lake volume | 8.32E+06 | 1.10E+08 | 1.06E+07 | 2.35E+08 | 1.53E+11 |
| DOC\_init (g m-3) | Initial in-lake DOC | 3.70 | 7.00 | 5.00 | 2.90 | 4.40 |
| POC\_init (g m-3) | Initial in-lake POC | 0.37 | 0.10 | 0.50 | 0.29 | 0.40 |
| **Allochthony** |  |  |  |  |  |  |
| PropCanopy | Proportion of shoreline with forest | 1.000 | 0.167 | 0.000 | 0.780 | 0.615 |
| PropWetlands | Proportion of shoreline with wetlands | 0.000 | 0.026 | 0.133 | 0.011 | 0.037 |
| WetlandLoad  (g d-1) | Loading rate of POC from wetlands | 1 | 1 | 1 | 1 | 1 |
| DOC\_gw (g m-3) | DOC concentration of groundwater | 10 | 10 | 10 | 10 | 10 |
| PropGW | Proportion of lake inflow as groundwater | 0.00 | 0.00 | 0.00 | 0.19 | 0.00 |
| DOC\_precip\_conc (g m-3) | DOC concentration of precipitation | 2 | 2 | 2 | 2 | 2 |
| AerialLoad (g d-1) | Influx of aerial POC (i.e., leaflitter) | 1 | 1 | 1 | 1 | 1 |
| **Autochthony** |  |  |  |  |  |  |
| *Respiration\_alloch (d*-*1)* | *Decomposition rate of allochthonous DOC in heterotrophic respiration* | *0.002* | *0.001* | *0.003* | *0.001* | *0.001* |
| *Respiration\_autoch (d-1)* | *Decomposition rate of autochthonous DOC in heterotrophic respiration* | *0.092* | *0.003* | *0.297* | *0.015* | *0.069* |
| R\_autotroph | Proportion of GPP autotrophically respired | 0.8 | 0.8 | 0.8 | 0.8 | 0.8 |
| **Burial** |  |  |  |  |  |  |
| *Burial alloch* | *Proportion of allochthonous POC buried in sediments* | *1.000* | *1.000* | *1.000* | *1.000* | *1.000* |
| *Burial\_autoch* | *Proportion of autochthonous POC buried in sediments* | *0.000* | *1.000* | *0.968* | *0.000* | *1.000* |
| Observed\_MAR (g m² yr-1) | Observed sediment mass accumulation rate | 78 | 249 | 153 | 27 | 186 |
| POClc\_alloch | Proportion of allochthonous POC leached to DOC (1-Burial\_alloch) | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 |
| POClc\_autoch | Proportion of autochthonous POC leached to DOC (1-Burial\_autoch) | 1.000 | 0.000 | 0.032 | 1.000 | 0.000 |

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| **Table 3. Built model with equations** | | |  |  |
| **Allochthonous DOC and POC** | **Units** | **Reference** | | |
| 1a) DOC SW = surface water concentration \* surface water inflow rate \* 86400 | g d-1 | Hanson et al. 2014 | | |
| 1b) Daily precipitation = rainfall \* 0.001 \* Area | m3 d-1 | Hanson et al. 2014 | | |
| 1c) DOC precipitation = DOC\_precip \* Daily precipitation | g d-1 | Hanson et al. 2014 | | |
| 1d) DOC Wetland = PropWetland \* WetlandLoad \* Perimeter | g d-1 | Hanson et al. 2014 | | |
| 1e) DOC GW = groundwater concentration \* groundwater inflow rate \* 86400 | g d-1 | Hanson et al. 2014 | | |
| 1f) POC canopy = PropCanopy \* AerialLoad \* Perimeter | g d-1 | Hanson et al. 2014 | | |
| 1g) Inflow load DOC = DOC Wetland + DOC GW + DOC SW + DOC precipitation | g d-1 | Hanson et al. 2014 | | |
| 1h) Internal load POC = (DOC Wetland + DOC SW) \* 0.1 | g d-1 | Hanson et al. 2014 | | |
| 1i) Inflow load POC = POC Aerial + Internal load POC | g d-1 | Hanson et al. 2014 | | |
| 1j) POC Aerial = AerialLoad \* Perimeter | g m-1 d-1 | Hanson et al. 2014 | | |
| **Autochthonous DOC and POC: primary production** |  |  | | |
| 2a) GPP rate = 10^(1.18 + (0.92 \* log10(chl-*a*\* photic depth)) + (0.014 \* epilimnion temperature)) | mg m-2 d-1 | Morin et al. 1999 | | |
| 2b) GPP percent DOC = 71.4 \* (chl-*a* \* photic depth)^(-0.22) | % | Pace and Prairie 2005 | | |
| 2c) GPP DOC rate = GPP rate \* (GPP percent DOC/100) | mg m-2 d-1 |  | | |
| 2d) GPP POC rate = GPP rate \* (1-(GPP percent DOC/100)) | mg m-2 d-1 |  | | |
| 2e) NPP DOC\_autoch = GPP DOC\_autoch \* 0.2 \* Area / 1000 | g d-1 |  | | |
| 2f) NPP POC\_autoch = GPP POC\_autoch \* 0.2 \* Area / 1000 | g d-1 |  | | |
| 2g) Autochthonous Respiration = GPP DOC rate \* Respiration\_autoch(1.08^(epilimnion temp - 20)) | g m-3 |  | | |
| 2h) Allochthonous Respiration = GPP DOC rate \* Respiration\_alloch(1.08^(epilimnion temp - 20)) | g m-3 |  | | |
| **Burial** |  |  | | |
| 3a) MAR\_alloch = POC mass \* Burial\_alloch \* 365/Area | g m-2 yr-1 |  | | |
| 3b) MAR\_autoch = POC mass \* Burial\_autoch \* 365/Area | g m-2 yr-1 |  | | |
| 3c) POC Burial\_alloch = MAR\_alloch \* (1/365) \* Area | g d-1 |  | | |
| 3d) POC Burial\_autoch = MAR\_autoch \* (1/365) \* Area | g d-1 |  | | |
| 3e) POC\_alloch leached out = POC\_alloch concentration \* POClc\_alloch \* Volume | g d-1 |  | | |
| 3f) POC\_autoch leached out = POC\_autoch concentration \* POClc\_autoch \* Volume | g d-1 |  | | |
| 3g) DOC\_alloch leached in = POC\_alloch leached out | g d-1 |  | | |
| 3h) DOC\_autoch leached in = POC\_autoch leached out | g d-1 |  | | |
| **NEP and Oxygen Flux** |  |  | | |
| 4a) NEP (as O2) = (NPP - DOCrespired) \* 32/12 | g m-3 d-1 |  | | |
| 4b) Fatm = 0.7 \* (DOconc - DOsat)/Zmix | g m-3 d-1 |  | | |
| 4c) DO(t+1) = DOconc + NEP - Fatm | g m-3 d-1 |  | | |

MAR = mass accumulation rate, Fatm = atmospheric flux rate, Zmix = mixed depth (m)

**Table 4. Model goodness of fit**

|  |  |  |
| --- | --- | --- |
| **Lake** | **RMSE\*** | **NSE\*\*** |
| Harp | 1.22 | 0.86 |
| Monona | 1.46 | 0.70 |
| Toolik | 1.19 | 0.79 |
| Trout | 0.80 | 0.95 |
| Vanern | 0.73 | 0.96 |
| \* Root mean square error (mg L-1) | | | |
| \*\* Nash-Sutcliffe efficiency | | | |

Table 5. Summary of annual mass balances (g m-2 y-1)

|  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- |
| **Lake** | **Alloch** | **Autoch** | **Total Load** | **Processed** | **Resp** | **Burial** | **Export** |
| **Annual means** | |  |  |  |  |  |  |
| Harp | 39.88 | 32.03 | 71.91 | 54.31 | -47.46 | -5.30 | -17.60 |
| Monona | 64.74 | 54.23 | 118.97 | 59.02 | -16.76 | -43.81 | -59.95 |
| Toolik | 76.19 | 11.27 | 87.45 | 31.86 | -23.68 | -6.67 | -55.59 |
| Trout | 13.35 | 28.17 | 41.52 | 37.56 | -37.45 | -1.19 | -3.95 |
| Vanern | 32.25 | 26.08 | 58.32 | 46.20 | -29.20 | -19.85 | -12.12 |
| **SD of annual means** | | |  |  |  |  |  |
| Harp | 10.71 | 7.99 | 11.42 | 9.89 | 9.42 | 0.95 | 4.86 |
| Monona | 24.16 | 18.05 | 21.81 | 15.38 | 1.99 | 12.85 | 20.58 |
| Toolik | 64.78 | 3.36 | 62.91 | 11.08 | 3.52 | 5.55 | 53.53 |
| Trout | 2.68 | 4.59 | 4.41 | 3.88 | 5.98 | 0.18 | 0.88 |
| Vanern | 5.81 | 4.91 | 5.86 | 4.97 | 2.35 | 3.14 | 1.84 |
| **Proportion of total load** | |  |  |  |  |  |  |
| Harp | 0.55 | 0.45 | 1.00 | 0.76 | -0.66 | -0.07 | -0.24 |
| Monona | 0.54 | 0.46 | 1.00 | 0.50 | -0.14 | -0.37 | -0.50 |
| Toolik | 0.87 | 0.13 | 1.00 | 0.36 | -0.27 | -0.08 | -0.64 |
| Trout | 0.32 | 0.68 | 1.00 | 0.90 | -0.90 | -0.03 | -0.10 |
| Vanern | 0.55 | 0.45 | 1.00 | 0.79 | -0.50 | -0.34 | -0.21 |

Positive fluxes represent OC inputs to lakes, whereas negative fluxes represent OC removal. Processed OC is the amount of the total load not exported. Positive net values represent OC sources. Proportions of allochthony and autochthony sum to 1. Proportions of respiration, burial, and export also sum to 1. SD = standard deviation.

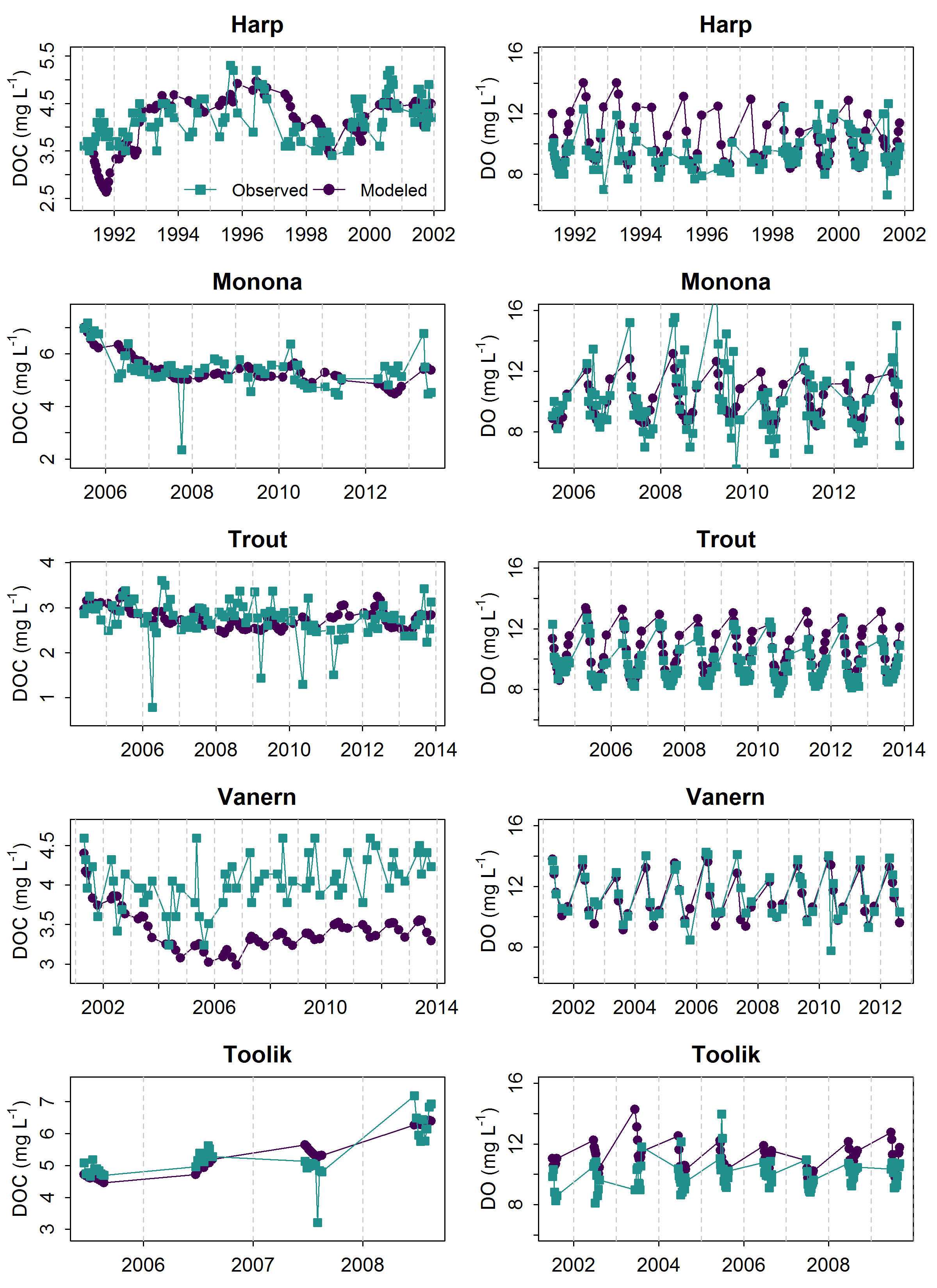
**FIGURE CAPTIONS**

**FIGURES**



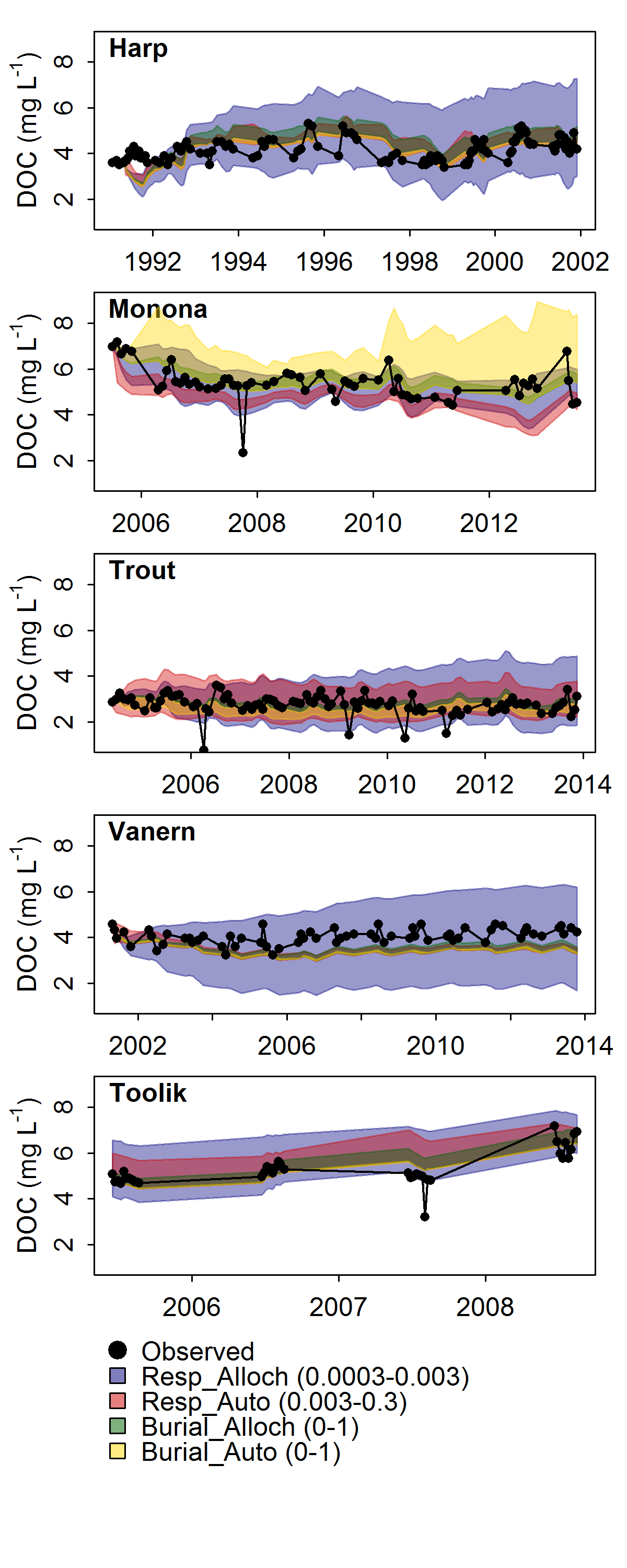
**Figure 1.**

Figure 1. Conceptual diagram of the organic carbon lake model depicting fluxes based on allocthonous (externally derived) and autochthonous (NPP) inputs of organic carbon, long-term burial, leaching of particulate organic carbon (POC) to dissolved organic carbon (DOC), respiration of DOC to CO2, and export via outflow. Four parameters (Respiration\_auto, Respiration\_alloch, Burial\_auto, and Burial\_alloch) are treated as free parameters and calibrated for each individual lake. Parameters and equations are defined in Tables 2 and 3.



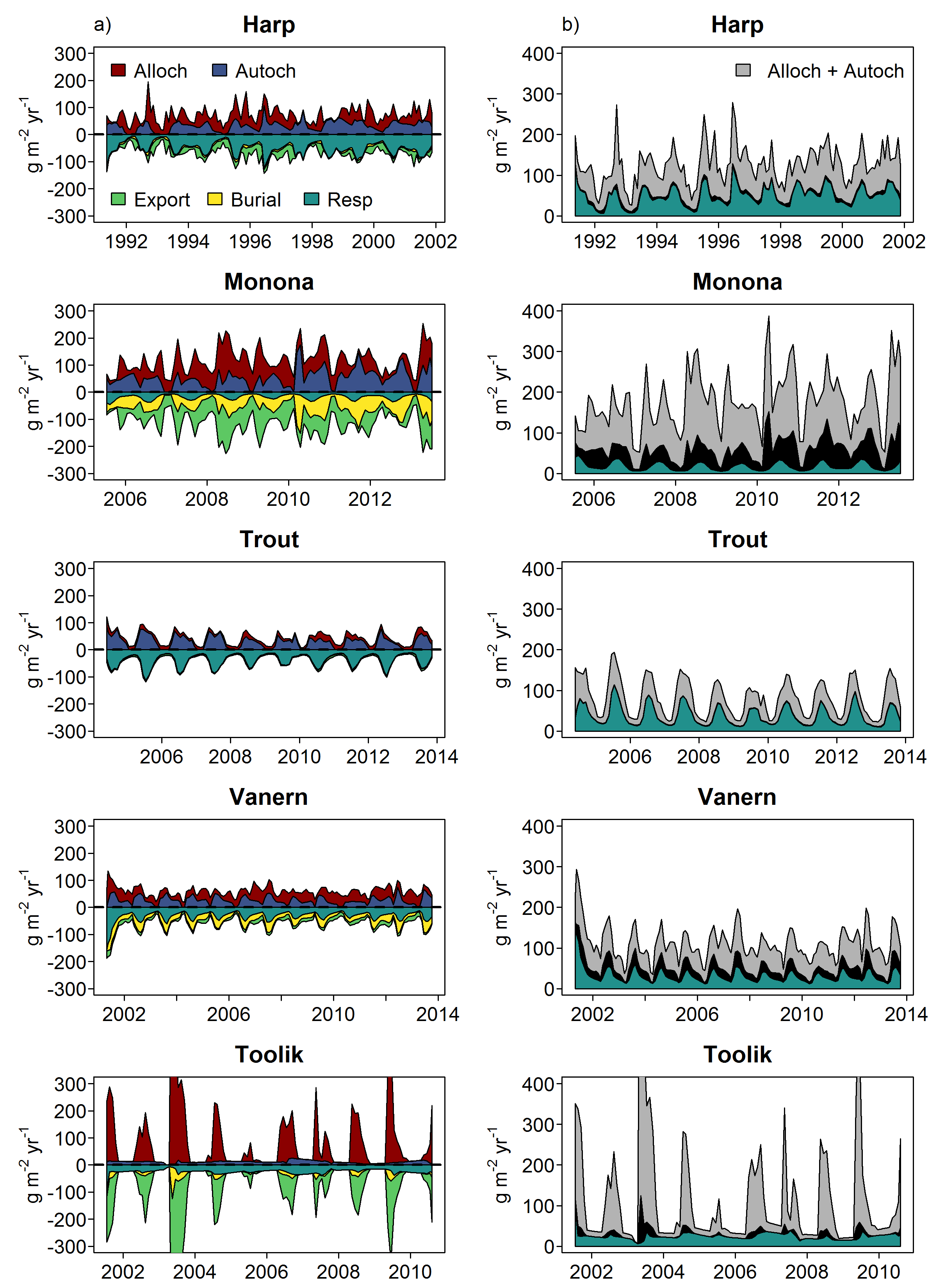
**Figure 2.**

Figure 2. Observed dissolved organic carbon (DOC) and dissolved oxygen (DO) concentrations in all lakes (blue circles) compared with modeled concentrations (red squares) on the same date. For some lakes, years differed between DOC and DO based on availability of observed data (S2).

****

**Figure 3.**

Figure 3. Sensitivity of calibrated model parameters. Each parameter was varied across a given range (Resp\_Alloch 0.0003-0.003, Resp\_Auto 0.003-0.3, Burial\_Alloch 0-1, Burial\_Auto 0-1) while the other three parameters remained fixed at their calibrated values. Shaded areas represent the range of modeled dissolved organic carbon (DOC) concentrations as each parameter is varied. Black circles represent the observed in-lake DOC concentrations.



**Figure 4.**

Figure 4. Time series of organic carbon fluxes and fates. a) Colored areas represent relative magnitudes of input fluxes (allochthonous and autochthonous) and output fluxes (export, burial, and respiration). All lines are stacked to show cumulative magnitudes. b) Absolute values of burial and respiration, and input fluxes.



**Figure 5.**

Figure 5. Relationship between log10-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 (either predominantly a respiration or burial, and either predominantly allochthonous driven or autochthonous driven) associated with each lake.

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