RESEARCH ARTICLE

Key Points:
- We integrated lake carbon fluxes in the context of inorganic and organic carbon cycling in a karst catchment from a system prospective
- Significance of lake C fluxes to adjustment of land C budgets depends on carbon origins and may be overrated (ignoring inorganic origin)
- Partitioning C fluxes between organic and inorganic origins is essential to accurately assess the roles of lakes in global carbon cycle

Importance of Considered Organic Versus Inorganic Source of Carbon to Lakes for Calculating Net Effect on Landscape C Budgets

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Abstract Lakes and reservoirs transform, emit, and bury carbon that is exported from land and are thus significant components of terrestrial carbon budgets. Their significance is often assessed by integrating these water bodies into terrestrial primary production. However, the transfer of inorganic carbon (IC) is likely a sticking point for these integrations because IC is not part of net ecosystem production. Here we integrated carbon evasion and organic carbon (OC) burial in a lake in the context of inorganic and OC cycling in a karst catchment from a system perspective. The lake emitted carbon dioxide (CO_2) and buried OC at rates of 1.0 ± 0.2 and 0.9 ± 0.2 g C m^{-2} a^{-1}, respectively, approximately equaling 13% and 11% of catchment net ecosystem production, respectively. These proportions represent significant influences on terrestrial carbon budgets, given an organic origin. However, catchment carbon export is dominated by IC that is derived from carbonates dissolved by soil CO_2. Lake CO_2 evasion accounts for less than 0.1% of soil CO_2 efflux, suggesting little potential in significantly altering terrestrial carbon budgets. This comparison indicates the significance of aquatic CO_2 evasion, requiring an adjustment of terrestrial carbon budgets to recognize their dependence on carbon origins. The significance may be overstated if inorganic origin is ignored. Our study suggests that a careful reassessment of the significance of CO_2 evasion and OC burial in freshwater ecosystems to local and global carbon budgets, with full consideration of their sources, is necessary and pressing.

1. Introduction

Lakes and reservoirs, together with other freshwater systems including streams, rivers, wetlands, and ponds, are thought to play important roles in the global carbon cycle by transforming, emitting, and burying amounts of carbon of similar magnitude to total global terrestrial net ecosystem production (NEP; Barros et al., 2011; Butman & Raymond, 2011; Dean & Gorham, 1998; Einarsdottir et al., 2017; Hotchkiss et al., 2015; Lundin et al., 2015; Regnier et al., 2013; Tranvik et al., 2009; Wallin et al., 2013). However, inland freshwaters were often regarded as passive conduits, and thus, their roles in delivering carbon from land to ocean were rarely explicitly included in traditional global terrestrial-ocean-atmosphere carbon cycle models (Cole et al., 2007; see also Aufdenkampe et al., 2011). Recent studies have demonstrated that inland waters are actually extremely active components, which participate in and facilitate local and global carbon cycling (e.g., Cole et al., 2007; Hanson et al., 2015; Tranvik et al., 2009). Not accounting for carbon emitted and sequestered by aquatic systems could bias efforts to accurately calculate terrestrial NEP and net ecosystem exchange (Cole et al., 2007; Hotchkiss et al., 2015; Kling et al., 1991; Nakayama, 2017; Tranvik et al., 2009;). A "boundless" concept has been proposed to designate all lateral and vertical carbon fluxes to and from freshwater systems (Battin et al., 2009; Regnier et al., 2013). The new model of carbon cycling, including freshwater fluxes, is expected to serve as a primary tool in managing Earth surface carbon cycling to address increasing carbon sequestration and decreasing carbon emission targets (Battin et al., 2009; Regnier et al., 2013). Great progress has been made in improving global estimates of freshwater CO_2 evasion by revisiting three key parameters: the amount of CO_2 in freshwaters, the global surface area of inland waters, and the gas transfer velocity, using some new approaches (e.g., Marx et al., 2017; Raymond et al., 2013).

A remaining obstacle to these efforts is that the partitioning and quantification of sources of freshwater CO_2 remain poorly constrained (e.g., Cole et al., 2007; Hotchkiss et al., 2015; Kayranli et al., 2010; Marx et al., 2017;
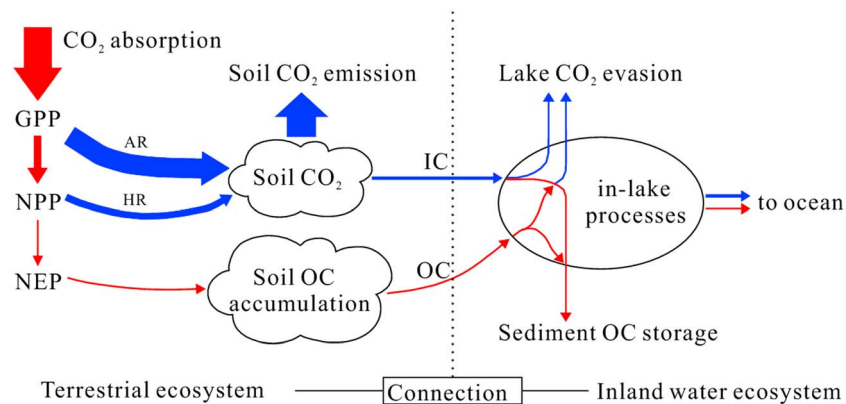
Figure 1. A sketch showing inorganic carbon (blue) and organic carbon (red) pathways of carbon transfer from terrestrial to aquatic ecosystems, which correspond to inorganic and organic origins of lake CO_2 (see text), respectively.

Raymond et al., 2013; Regnier et al., 2013; Xu et al., 2014). As shown in Figure 1, CO_2 in lakes either originates from the input of soil CO_2 (derived from root-related autotrophic respiration [AR] and soil heterotrophic respiration [HR]) through flowing groundwater and rivers (hereinafter referred to as "inorganic origin") or is internally produced by in-lake mineralization of soil organic carbon (OC) from catchments (hereinafter referred to as "organic origin"; Cole et al., 1994) and is likely from both (Buffam et al., 2011). Transport of carbon from land to water is an important factor in controlling the carbon budget in lakes and reservoirs (Cole et al., 1994). Recently, much progress has been made concerning terrestrial carbon export and its coupling with aquatic ecosystems. Ludwig et al. (2011) gave a global average of continental carbon erosion of 0.60 GtC yr^{-1}, of which 0.37 GtC yr^{-1} can be attributed to OC, and the remaining 0.23 GtC yr^{-1} to inorganic carbon (IC; CO_2 as a form of HCO_3^-). Tian et al.'s (2015) modeling results suggested that climate variability is the primary factor in causing interannual variability of OC and IC export. Nakayama (2017) developed an advanced model which coupled hydrology-carbon cycling in terrestrial-aquatic linkages and the interplay between inorganic and OC, and produced horizontal and vertical carbon fluxes for rivers, which are in good agreement with prior research. However, the connections of carbon between terrestrial and aquatic systems in terms of type (IC and OC), strength, spatial extent, and influence (Figure 1) remain poorly constrained (Buffam et al., 2011).

A close relationship between dissolved OC (DOC) concentrations and partial pressures of CO_2 (pCO_2) is often observed in lakes and is considered a key piece of evidence to support the view that carbon evasion in freshwater systems is maintained by the input of labile organic matter (organic origin; see Figure 1; e.g., Ask et al., 2012; Cole & Caraco, 2001; Sobek et al., 2003). The carbon emitted from and stored within lakes is assumed to originate mainly from allochthonous OC delivered from catchments and thus should be included in estimates of terrestrial net primary production (NPP; e.g., Battin et al., 2009; Cole et al., 2007; Tranvik et al., 2009). The scenario that OC sources predominate in lake carbon input occurs, in particular, in boreal and arctic areas, where tundra ecosystems accumulate OC, which can be subsequently supplied to lakes and rivers by erosion (Einarsdottir et al., 2017; Kling et al., 1991; Strock et al., 2017).

Recently, increasing evidence indicates that lacustrine CO_2 production is closely related to IC input (Figure 1). Direct, hydrological input of soil CO_2 is assumed to be an important source of lake CO_2 (Abril et al., 2014; Maberly et al., 2013; McDonald et al., 2013). On the other hand, dissolved IC (DIC, mainly HCO_3^-) derived from catchments is thought to be a primary factor in sustaining CO_2 emissions in many lakes (Jones et al., 2001; Öquist et al., 2009; Wang et al., 2012, 2015; Weyhenmeyer et al., 2015). These findings are in agreement with the results of a statistical analysis of more than 5,000 lakes reported by Marcé et al. (2015), which showed that CO_2 supersaturation in lakes was directly related to carbonate weathering in catchments.

Similar questions concerning carbon sources exist for the OC buried in lake sediments (Downing et al., 2008). If the OC is primarily allochthonous and derived from the terrestrial biosphere, then the lake is not a net sink of carbon because there is no net change in the total amount of OC, just a change in the storage location: catchment soils to lake sediments (Mendonça et al., 2012), unless OC could escape mineralization and is stored in sediments over long time scales (thousands to millions years; Battin et al., 2009; Cole, 2013). In

contrast, if the OC is primarily autochthonous and produced by photosynthesis in lake waters, it should not be included in estimates of terrestrial primary production. In this case, lakes may be a net sink of carbon, depending on the sources of CO_2 utilized in photosynthesis.

It is still not easy to accurately assess the relative roles of freshwater ecosystems in local and global carbon budgets (Battin et al., 2009; Cole, 2013), despite advances in global estimates of the fluxes of freshwater CO_2 evasion and OC burial (e.g., Dean & Gorham, 1998; Raymond et al., 2013; Regnier et al., 2013; Tranvik et al., 2009). Duarte and Agusti (1998) suggested that if community respiration exceeds gross primary production (GPP), systems are heterotrophic and thus act as CO_2 sources; otherwise, systems are autotrophic and act as CO_2 sinks. However, measurements of aquatic ecosystem production are difficult to make in real ecosystems (Hanson et al., 2004). Another, more feasible method of assessing the net effect of lakes is quantifying the balance of gaseous carbon evasion and OC burial. If OC burial exceeds carbon evasion, lakes and reservoirs act as net CO_2 sinks; otherwise, they act as net CO_2 sources (Hanson et al., 2004; Mendonça et al., 2012). However, quantitative comparisons of these carbon phases, with no distinctions made of their sources, may lead to inaccurate conclusions.

More important is that freshwater carbon fluxes are significant and require accurate incorporation into terrestrial carbon budgets. In conventional carbon cycling, outgassing from inland waters is blended with fluxes of terrestrial ecosystem respiration. However, if it is terrestrially sourced OC that is respired by heterotrophs in inland waters releasing CO_2, these fluxes should be considered part of terrestrial carbon budgets (Cole et al., 2007). Ecosystem production must be increased from a conventional estimate of 2–3.2 to 4–4.5 Pg C a^{-1} to balance the carbon released (see Aufdenkampe et al., 2011; Battin et al., 2009; Tranvik et al., 2009). Considering the strong and extensive connections between carbon transport from terrestrial to aquatic systems, integrating inland waters into terrestrial carbon cycling has been a widely accepted method to construct a complete carbon budget (Buffam et al., 2011; Christensen et al., 2007; Cole et al., 2007; Jonsson et al., 2007). Butman et al. (2016) were the first to do an integrated assessment of freshwater carbon in terrestrial ecosystem production for the conterminous United States. Such an integration is good for constructing local carbon budgets but may limit our understanding of the roles of lakes due to the bypassing of important details of carbon connections between terrestrial and aquatic systems. In particular, integration methods pay close attention to the transfer of OC that belongs to direct products of ecosystem primary production. The catchments involved in prior integration studies (e.g., Buffam et al., 2011; Christensen et al., 2007; Jonsson et al., 2007) are mostly located in boreal and Arctic/subarctic areas, where OC dominates catchment carbon export. IC is a potential sticking point for this method of integration because IC does not belong to NEP (Chapin et al., 2006; Lovett et al., 2006). Lovett et al. (2006) instead used the concept of net ecosystem exchange, and Chapin et al. (2006) proposed the concept of net ecosystem carbon balance, to differentiate IC from NEP; however, NEP is still the most frequently used term in carbon cycling. Although lake carbon is ultimately derived from atmospheric CO_2 fixation in catchment primary production, organic and inorganic pathways (or origins) of carbon transport from terrestrial to aquatic ecosystems (Figure 1) have different turnover rates and residence times, and as such have different influences on local and global carbon cycling. Ignoring these differences could result in miscalculations of local and global carbon budgets.

In this paper, a small karst catchment in southwestern China, with one of the highest values of IC export of atmospheric CO_2 in the world due to widely distributed carbonates (Ludwig et al., 2011), was chosen for study to address the influence of terrestrial IC transfer on the integration of catchment-lake carbon budgets. Previously, we estimated CO_2 emission fluxes in the small reservoir, Lake Hongfeng, and explored the geochemical processes of aquatic CO_2 production from inorganic origins (Wang et al., 2012, 2015). Later, soil CO_2 concentrations and emission fluxes in the catchment were also reported (Liu et al., 2012). Catchment weathering rates, NPP, and NEP were also estimated. In this study, aquatic CO_2 emissions and carbon input and export fluxes in the catchment were observed again with higher sampling frequency and at more sampling sites. By synthesizing the existing and new data for various carbon phases, we are able to comprehensively compare freshwater carbon fluxes with other vertical and lateral fluxes of OC (GPP, NPP, NEP, and OC export) and IC (soil CO_2 emission, carbonate dissolution by soil CO_2, and IC export) in this catchment. Then, we integrated lake carbon fluxes in the context of production, transport, transformation, exchange, and storage (of both OC and IC) in the catchment from a systems perspective. Finally, we developed a
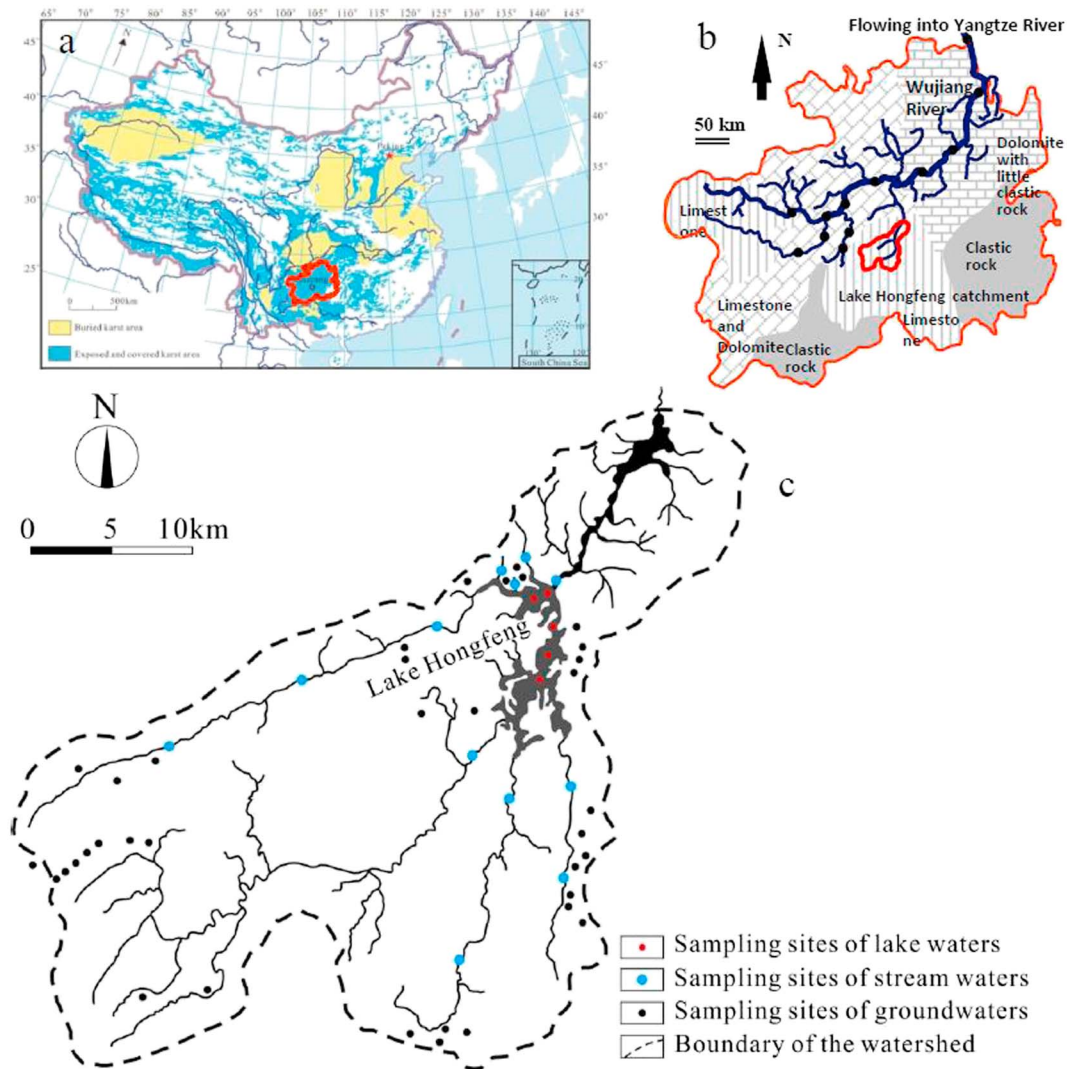
Figure 2. Maps of (a) China, (b) Guizhou province, and the (c) Lake Hongfeng catchment. The distribution of karst in China and the location of Guizhou province (red line) are shown in (a). The Wujiang River, constructed reservoirs (solid circles), the Lake Hongfeng catchment, and the exposed rock types are shown in (b). The sampling sites in the catchment are shown in (c).

method of assessing the significance of lake carbon fluxes to the local carbon budget by considering the origins and coupling of OC and IC.

2. Materials and Methods

The study lake (Lake Hongfeng) and its catchment have been previously described in detail (Wang et al., 2012). Lake Hongfeng (106°19–28′E, 26°26–35′N) is located in the upper reaches of the Maotiao River, a tributary of the Wujiang River, which is located in the upper reaches of the Yangtze River catchment (Figure 2). It was constructed in 1960 and has a mean water residence time of 0.35 years (Wang et al., 2012). The lake is 16 km long, with a surface area of 32.2 km^2, a catchment area of 1,596 km^2, a maximum depth of 45 m, an average depth of 9.2 m, and a volume of approximately 0.298×10^9 m^3. Lake Hongfeng is mesotrophic to eutrophic due to input of domestic sewage from surrounding towns, with high concentrations of total nitrogen (0.204 mmol L^{-1}), total phosphorus (1.516 μmol L^{-1}), and chlorophyll a (6.2–10.9 mg m^{-3}; Wang et al., 2015).

The Guizhou karst plateau in southwestern China is located in the center of the Southeast Asian Karst Region, where karstification is most developed (Figure 2a). The Lake Hongfeng catchment is located in the center of

the Guizhou karst plateau (Figure 2b). The catchment is 65 km long and 17 km wide, with a southwest to northeast orientation, and an elevation range of 700–1,765 m above msl. Permian and Triassic age limestones and dolomites dominate the bedrock here (Figure 2b). The catchment is characterized by well-developed karst landforms, steep-sloped valleys, and gorges and is sparsely vegetated (mainly with shrubs and limited broadleaf and coniferous forests). A subtropical, monsoon climate prevails here, with a mean annual temperature of 14 °C, and mean annual precipitation of 1,195 mm.

Lake Hongfeng and its surrounding streams, springs, and headwaters were sampled seasonally from 2001 to 2002 and bimonthly in 2004 (Wang et al., 2015). In 2013, lake and stream waters were again collected at these sites with higher sampling frequency (monthly to semimonthly), sampling of spring water in the catchment was extended from 12 to 36 sites (Figure 2c), and sampling frequency was increased from 1 to 10 times (see Appendix Tables S1, S2, and S3 in the supporting information), with the overall objective of reducing uncertainties in the results. Sample analysis methods were described in Wang et al. (2015). pCO_2 was calculated from HCO_3^-, pH, the reaction constant between HCO_3^- and CO_2 (aq), and Henry's law constant for CO_2 (Wallin et al., 2010; Wang et al., 2015).

The exchange flux (F) of CO_2 through the water-air interface was estimated by a molecular diffusion model:

$$F = k \times \Delta C \tag{1}$$

where k = the gas exchange rate (0.75 m d^{-1} for lake surface and 1.9 m d^{-1} for stream surface; Cole et al., 1994; Cole & Caraco, 2001) and ΔC = the difference of gas concentration across the interface. Groundwater CO_2 evasion was estimated by multiplying the CO_2 concentration difference between spring water and river water by the mean annual discharge. Input and export fluxes of carbon species into and out of the lake were estimated by multiplying riverine carbon concentrations by discharge. Export flux of HCO_3^- out of the catchment was estimated by multiplying the HCO_3^- concentration in the groundwater by mean annual discharge. HCO_3^- in groundwater is mainly produced by the weathering of carbonate by soil CO_2:

$$CaCO_3 + CO_2 + H_2O = Ca^{2+} + 2HCO_3^- \tag{2}$$
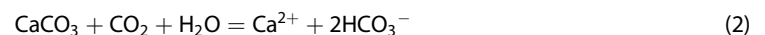
Half of the HCO_3^- in equation (2) is from soil CO_2, and half is from carbonate. On the other hand, sulfuric and nitric acids can enhance carbonate weathering. Han and Liu (2004) reported that 54% of HCO_3^- is from carbonate, and 46% is from soil CO_2. We estimated the CO_2 consumption flux in carbonate dissolution using the empirical ratio 54/46. The deposition and burial fluxes (Fs) of carbon in the sediments were calculated using

$$Fs = a \times Cc \tag{3}$$

where a = sediment mass accumulation rates and Cc = carbon concentrations in sediments in the upper 2 cm (for deposition flux) or below 30 cm (for burial flux; see Appendix Table S4). The emission rate of soil CO_2 was measured monthly from June 2006 to May 2007, at 10 sampling plots in the catchment using a dark static flux chamber (Liu et al., 2012). NPP and NEP were compiled from published data (J. Cao et al., 2011; M. Cao et al., 2003; Dong & Ni, 2011; Gu et al., 2007; Ke et al., 2003; Liu, 2017). GPP, AR, and HR were estimated using

$$NPP = GPP - AR \tag{4}$$

$$NEP = NPP - HR \tag{5}$$

All results are expressed with average ± standard deviation.

3. Results and Discussion

3.1. Aquatic CO_2 Emission and OC Burial in the Catchment

Partial pressures of CO_2 in the surface lake water and emission flux of CO_2 at the lake water-air interface showed clear seasonal patterns, according to the observations that have made with higher sampling frequency at more sampling sites in 2013 (Figure 3) relative to the previous study (Wang et al., 2015). In summer and fall, the surface lake water was undersaturated with respect to atmospheric CO_2 owing to heavy consumption of CO_2 by algal photosynthesis. Therefore, it absorbed CO_2 from the atmosphere and acted as a carbon sink. In winter and early spring, the lake water was supersaturated in CO_2 with high pCO_2 ranging from 1,000 to 2,000 μ atm and emitted more CO_2 into the atmosphere, acting as a carbon source (Figure 3). In comparison, variations of pCO_2 displayed some irregular patterns in the previous observation that had a
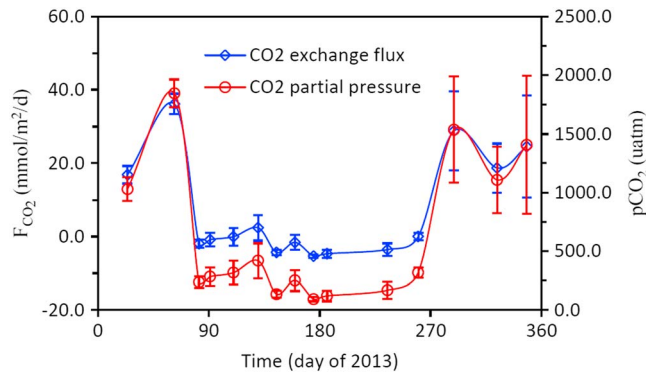
Figure 3. Seasonal variations of CO_2 emission fluxes and pCO_2 in lake surface waters, 2013.

low sampling frequency (Wang et al., 2015). For example, exceptionally high pCO_2 of up to 5,000 μ atm occurred, and sometimes the lake water was under saturated in CO_2 in winter when the lake was vertically mixed (see Wang et al., 2015). As a whole, the seasonal pattern of CO_2 emissions that was observed in this time is in accord with the mesotrophic-eutrophic status of the lake. On the other hand, the seasonal pattern showed that the conversion of the lake between acting as a carbon source and sink could be finished in a short time (Figure 3), indicating that high sampling frequency is essential for acquiring convincing results. Annual averages of emission fluxes of CO_2 at the lake water-air interface ranged from 7.0 to 19.0 mmol m^{-2} d^{-1}, with no clear interannual variation trends (Table 1). The overall average of the means is 11.5 $\pm$ 2.6 mmol m^{-2} d^{-1} (Table 1), which is slightly lower than the mean, global value (16.2 mmol m^{-2} d^{-1}, Cole et al., 1994). This flux is equal to 1.0 $\pm$ 0.2 g C m^{-2} a^{-1} when expressed considering catchment area. The average of pCO_2 values (3,725.1 $\pm$ 1,938.1) at seven catchment stream sampling sites observed in 2001–2001 were consistent with those from rivers from other regions of the world (Butman & Raymond, 2011; Cole & Caraco, 2001; Raymond et al., 2013). The average decreased to 2,730.7 $\pm$ 473.2 μ atm in 2013 (Table 1). Assuming that stream surface area is about 0.19% of the catchment surface, like other low-order streams in headwater areas (Wallin et al., 2013), riverine CO_2 evasion could be estimated to be 1.5 $\pm$ 0.2 g C m^{-2} a^{-1}, which is slightly lower than the results from a boreal headwater catchment (1.8 g C m^{-2} a^{-1}) reported by Leith et al. (2015) but much lower than the value (5.0 $\pm$ 1.8 g C m^{-2} a^{-1}) in DOC-rich streams in northern Sweden documented by Wallin et al. (2013). Including groundwater CO_2 degassing of 1.9 $\pm$ 0.1 g C m^{-2} a^{-1}, total CO_2 evasion from the aquatic environments in this catchment was as high as 4.4 g C m^{-2} a^{-1}, of which the lake accounts for 23%, river water for 34%, and groundwater for 43%.
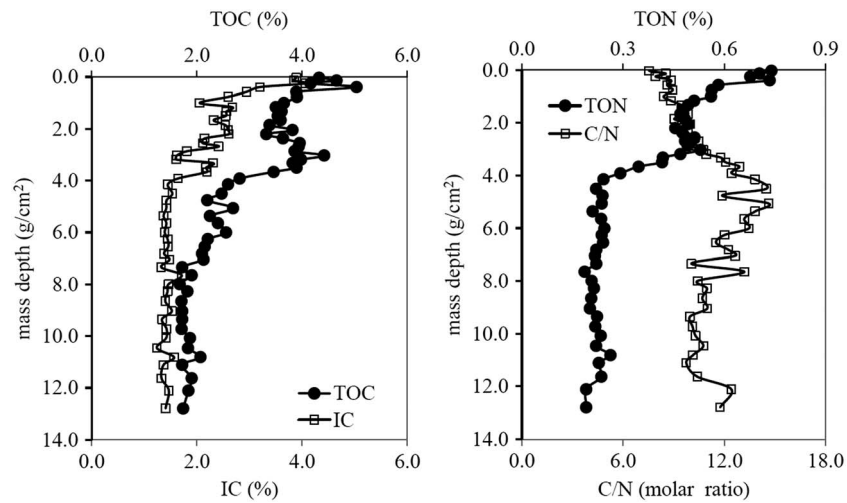
The inorganic and OC concentrations in lake sediments decreased with depth (Figure 4, similar to the results of Wang et al. (2015). Using the sediment mass accumulation rate of 0.17 g cm^{-2} a^{-1} (Wan et al., 1990) and the OC concentrations in the sediment, it was estimated that OC was delivered to the sediment-water interface at a rate of approximately 29.7 $\pm$ 7.3 mmol C m^{-2} d^{-1}, of which ~69% was remineralized during burial (diagenesis), leaving 9.9 $\pm$ 1.6 mmol C m^{-2} d^{-1} to be stored in the sediment (Table 1). The flux was 0.9 $\pm$ 0.2 g C m^{-2} a^{-1} as expressed considering catchment area. Organic N concentrations decreased, and C:N ratios increased with depth in lake floor sediments (Figure 4) due to organic matter decomposition, during which CO_2 was released and further dissolved carbonate. As a result, IC concentrations sharply decreased (Figure 4), and HCO_3^- had high concentrations in sediment pore waters, and thus could diffuse into overlying lake water (Wu et al., 1997).

3.2. Terrestrial OC and IC Fluxes in Relation to CO_2 Exchange

Net primary production in this karst catchment is relatively low, according to the compiled data. Estimates of NPP here varied, as investigators utilized different methods (Table 2). According to Ke et al.'s (2003) results using the Carnegie Ames Stanford Approach (CASA) model, average NPP in the southwestern part of the Yangtze River catchment (1981–2000) was 262 g C m^{-2} a^{-1} (Table 2). Gu et al. (2007) used the Atmosphere-Vegetation Interaction Model to model NPP in the same area and reported 417 g C m^{-2} a^{-1}. Dong and Ni's (2011) results using the CASA model (1999–2003) showed that NPP in Guizhou was <285 g C m^{-2} a^{-1}. This value is in broad agreement with J. Cao et al.'s (2011) results, showing that NPP from 1991 to 2009 in Guizhou ranged from 194.0 to 268.3 g C m^{-2} a^{-1} (mean of 227.5 $\pm$ 23.6 g C m^{-2} a^{-1}). Liu's (2017) result using the Lund-Potsdam-Jena Dynamics Global Vegetation Model suggested that the average NPP of vegetation in Guizhou reached 737.8 g C m^{-2} a^{-1}. It is interesting to note that Ke et al. (2003), Dong and Ni (2011), and J. Cao et al. (2011) all used the process-based CASA model (Table 2) and reported similar, low results for NPP (194.5–285 g C m^{-2} a^{-1}). In comparison, Gu et al. (2007), Liu (2017), and Wang, Yang, and Wang (2007) used land and eco-physiology feedback-based vegetation models and meteorological data to model NPP and all reported higher values (417.0–737.8 g C m^{-2} a^{-1}). Given that vegetation in this catchment is sparse and shrub-dominated, the results from the eco-physiology-based vegetation models

Table 1
Carbon Exchange Fluxes in the Lake Hongfeng Catchment

Processes	Compartment /C types	Time	pCO$_2$ (μ atm)/OC or IC contents (%)	Exchange fluxes of carbon Mmol m^{-2} d^{-1} per lake surface area	gC m^{-2} a^{-1} per catchment area	Sampling frequencies	References
Aquatic CO$_2$ emissions	Surface lake water	2001	1,185.3 ± 557.1	19.0 ± 13.7	1.7 ± 1.2	5 Times at 3 Sites	Wang et al. (2015)
		2002	675.4 ± 254.0	7.6 ± 12.8	0.7 ± 1.1	2 Times at 3 Sites	Wang et al. (2015)
		2004	876.7 ± 361.7	12.2 ± 8.6	1.1 ± 0.8	6 Times at 4 Sites	Wang et al. (2015)
		2013	617.9 ± 275.0	7.0 ± 6.5	0.6 ± 0.6	15 Times at 5 Sites	This study
		Average	838.8 ± 118.5	11.5 ± 2.6	1.0 ± 0.2		
	River	2002	3,725.0 ± 1,938.1	201.5 ± 114.5	1.7 ± 1.0	4 Times at 7 Sites	Wang et al. (2015)
		2013	2,730.7 ± 473.2	142.8 ± 28.0	1.2 ± 0.2	10 Times at 12 Sites	This study
		Average	3,086.9 ± 232.9	172.2 ± 19.2	1.5 ± 0.2		
	Groundwater	2002	10,207.0 ± 7,749.0		1.9 ± 1.8	1 Time at 12 Sites	Wang et al. (2015)
		2013	8,798.4 ± 1,046.7		1.8 ± 0.2	10 Times at 36 Sites	This study
		Average	9,502.7 ± 996.0		1.9 ± 0.1		
Carbon exchange at the sediment-water interface	OC deposition	January 2002	11.1 ± 0.8	43.4 ± 2.3	3.8 ± 0.3		Wang et al. (2015)
		August 2002	6.9 ± 0.8	27.4 ± 2.3	2.4 ± 0.3		Wang et al. (2015)
		May 2015	4.5 ± 0.2	18.3 ± 0.1	1.5 ± 0.1		This study
		Average	5.7 ± 1.7	29.7 ± 7.3	2.6 ± 1.1		
	OC burial	January 2002	2.9 ± 0.1	11.4 ± 0.5	1.0 ± 0.03		Wang et al. (2015)
		August 2002	2.9 ± 0.4	11.4 ± 2.3	1.0 ± 0.1		Wang et al. (2015)
		May 2015	2.0 ± 0.3	6.8 ± 2.3	0.7 ± 0.1		This study
		Average	2.6 ± 0.5	9.9 ± 3.2	0.9 ± 0.2		
	IC deposition	January 2002	8.2 ± 5.5	32.0 ± 20.5	2.8 ± 1.9		Wang et al. (2015)
		August 2002	10.9 ± 0.6	43.4 ± 2.3	3.7 ± 0.2		Wang et al. (2015)
		May 2015	3.9 ± 0.1	16.0 ± 0.5	1.3 ± 0.03		This study
		Average	7.4 ± 4.9	30.5 ± 7.6	2.6 ± 1.2		
	IC burial	January 2002	1.7 ± 0.3	6.8 ± 2.3	0.6 ± 0.1		Wang et al. (2015)
		August 2002	1.4	4.6	0.5		Wang et al. (2015)
		May 2015	1.4 ± 0.1	4.6 ± 0.5	0.5 ± 0.03		This study
		Average	1.5 ± 0.2	5.3 ± 0.8	0.5 ± 0.1		
Terrestrial carbon exchanges	NPP				204.6 to 274.3		See Table 2
	NEP				−8 (0 to −20)		See Table 2
	Soil CO$_2$ efflux	2006/2007			1,392.0 ± 122.6	12 Times at 10 Sites	Liu et al. (2012)
	CO$_2$ consumption in carbonate erosion	2001			12.6		Wang et al. (2015)
		2013			10.5		This study
		Average			11.6 ± 1.5		
Carbon input into the lake from the catchment	CO$_2$	2001			0.8		Wang et al. (2015)
		2013			0.4 0.6 ± 0.13		This study
		Average					
		2001			18.0 (8.4 from soil CO$_2$)		Wang et al. (2015)
	HCO$_3^-$	2013			15.0 (6.9 from soil CO$_2$)		This study
		Average			16.5 ± 2.1 (7.7 ± 1.1 from soil CO$_2$)		
		2001			0.15		Wang et al. (2015)
	CO$_3^{2-}$	2013			0.15		This study
		Average			0.15		
	DIC	2001			19.0		Wang et al. (2015)
		2013			15.5		This study
		Average			17.3 ± 2.5		
	DOC	2001			0.9		Wang et al. (2015)
	POC	2001			0.2		Wang et al. (2015)
Lake carbon export downstream	DIC	2001			16.2		Wang et al. (2015)
		2013			14.4		This study
		Average			15.3 ± 1.3		
	DOC	2001			1.4		Wang et al. (2015)
	POC	2001			0.15		Wang et al. (2015)

Figure 4. Profiles of total organic carbon, inorganic carbon, total organic nitrogen, and C:N ratios in lake floor sediments.

likely reflect the NPP of vegetation and forest ecosystems but overestimate overall NPP for the entire catchment. Thus, we adopted the confidence interval (204.6–274.3 g C m^{-2} a^{-1}) from the CASA model at 95% confidence level as the most likely range of NPP for this catchment (Table 1). As a whole, these values are relatively low. In contrast, modeling results show that the relatively high temperature and precipitation here produced high respiration rates in the catchment (J. Cao et al., 2011; Liu, 2017). As a result, NEP in the area was consistently negative (0 to −20 g C m^{-2} a^{-1}) as documented by M. Cao et al. (2003). This was confirmed by the modeling results reported by Liu (2017), which indicate that NEP is negative in 85% of the Guizhou Province, with an average of −8 g C m^{-2} a^{-1} (Table 1).

Comparatively, soil CO_2 efflux in this catchment is high. The vertical CO_2 efflux from soils reached an average value of 1,392 ± 122.6 g C m^{-2} a^{-1} (Liu et al., 2012). This value is in agreement with J. Cao et al.'s (2011) observation showing that soil respiration in Guizhou was 1,487.7 ± 63.3 g C m^{-2} a^{-1}. These emission rates of soil CO_2 in carbonate areas are similar to results from noncarbonate areas of China, in spite of the relatively low NPP. Mo et al. (2005) gauged the soil CO_2 efflux in evergreen broadleaf forest, pine forest, and pine and broadleaf mixed forest in the subtropical, monsoonal Dinghushan area (23°10′N, 112°34′E) in southern China at 1,550, 1,419, and 1,103 g C m^{-2} a^{-1}, respectively. According to estimates of NPP and NEP (Table 2) and equation (5), HR in this catchment should range from 200 to 290 g C m^{-2} a^{-1}. If soil CO_2 is further assumed only to originate from autotrophic and HR, and not from abiological sources, AR should range between 1,100 and 1,200 g C m^{-2} a^{-1}. These estimates indicate that ~80% of soil CO_2 was derived from root respiration, and the remaining 20% was derived from heterotrophic decomposition of soil organic matter. Even though the abiological contribution to soil CO_2 is taken into account (15–35%; Wang et al., 2015), the amount of CO_2 derived from root respiration still surpassed that from HR. It must be noted that the simple estimates of contributions of heterotrophic and AR to soil CO_2 presented here contain large errors, since partitioning of soil CO_2 between heterotrophic and AR is a complicated task (see Hanson et al., 2000;

Table 2
A Comparison of the Compiled NPP and NEP Data

References	Region of modeling	Period of modeling	Model	NPP (gC/m^2/a)	NEP (gC/m^2/a)
Ke et al. (2003)	The Yangtze Basin	1981–2000	CASA	262	
Dong and Ni (2011)	Guizhou	1999–2003	CASA	<285	
J. Cao et al. (2011)	Guizhou	1991–2009	CASA	227.5 ± 23.6 (194.0–268.3)	
Gu et al. (2007)	Southwest China	1981–2000	AVIM2	417.0	
Wang et al. (2007)	Guizhou	2001	Light use efficiency model	407.2	
M. Cao et al. (2003)	Guizhou	1988–1998	CEVSA and GLO-PEM		0 to −20
Liu (2017)	Guizhou	2000–2010	LPJ-karst	737.8	−8
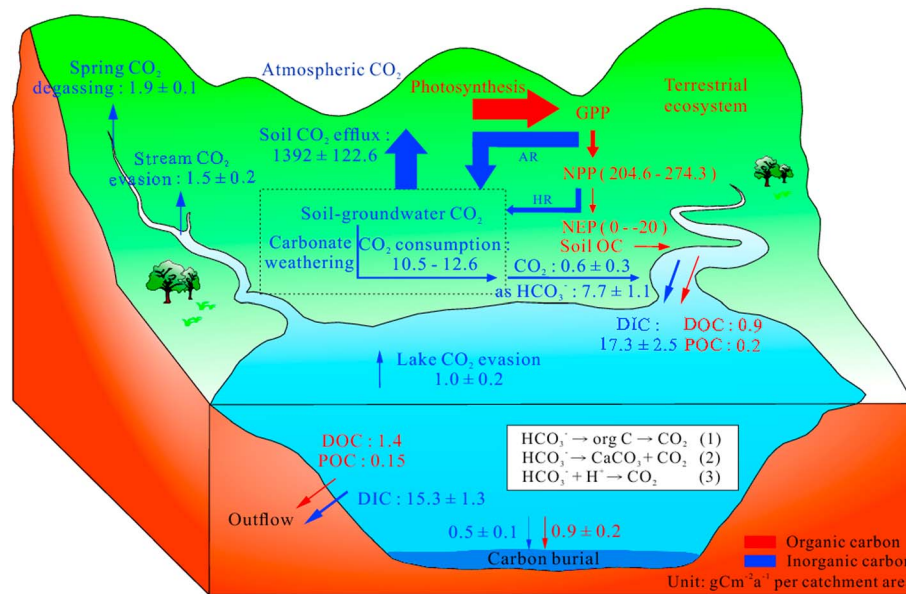
Figure 5. Pathways and vertical and lateral fluxes of organic carbon and inorganic carbon transfer in the soil (vegetation)—carbonate—water systems in the Lake Hongfeng catchment. Size of arrows reflects relative magnitudes (for data see Table 1).

Kuzyakov, 2006). Generally, the contribution of root respiration to soil CO_2 emissions varies over a large range (<10% to >90%), with an overall mean of 48% as reported by Hanson et al. (2000), who compiled data from 50 studies covering large areas of the globe. Ding et al. (2010) found that the contribution of root respiration to soil CO_2 emissions averaged >80% in densely vegetated parts of the evergreen broad lead forest, and the contribution was >90% in the upper 20 cm of soil, and decreased to 32% below 20 cm at sparsely vegetated sites in the Dinghushan area in southern China, geographically near to the study area. The phenomenon reported here, that root respiration contributed a large proportion of soil CO_2 in the catchment, is consistent with the observation that plants in karst areas tend to allocate more biomass in roots (Ni et al., 2015).

The lateral flux of soil CO_2 export into streams in this catchment is a small proportion of the soil CO_2 budget, <1% of the vertical emission of soil CO_2 to the atmosphere. This flux consists of two parts: free CO_2 in waters and the HCO_3^- that is produced by chemical weathering of rock by soil CO_2 (excluding carbon from carbonate). The latter has been estimated to be 8.4 g C m^{-2} a^{-1} in 2001 (Wang et al., 2012) and 6.9 g C m^{-2} a^{-1} in 2013 (Table 1), using catchment stream discharge and HCO_3^- concentrations. In comparison, J. Cao et al. (2011) determined the mass loss of carbonate tablets that were placed into soils in the field to gauge the rate of carbonate dissolution at 13.8 g C m^{-2} a^{-1} in the Pearl River catchment of China, which geographically borders the study site. It is understandable that our results are lower than those from J. Cao et al.'s (2011) experiment because carbonate precipitation and CO_2 evasion, likely as well photosynthesis, reduced DIC concentrations from 3.99 ± 1.17 mmol L^{-1} ($n = 318$) in groundwater to 2.64 ± 0.80 mmol L^{-1} ($n = 115$) in streams. It could be concluded that the flux of soil CO_2 consumption in the catchment due to carbonate erosion might range from 10.5 to 12.6 g C m^{-2} a^{-1} (Table 1). Gombert (2002) gave an average global karst dissolution rate of 10.8 g C m^{-2} a^{-1}. The results within Guizhou are slightly larger than the global average, due likely to the wet and hot climate.

3.3. Process-Based Integration Shows That the Lake CO_2 Originates From Soil CO_2 in the Catchment

By synthesizing existing and new data (Table 1), the catchment-scale mass balance of carbon, coupling with NPP, NEP, soil CO_2 emissions, carbonate dissolution by soil CO_2, and aquatic CO_2 emissions, was constructed (Figure 5). The lake received a considerable amount of carbon input from the catchment, of which IC was 17.3 ± 2.5 g C m^{-2} a^{-1} and OC was 1.1 g C m^{-2} a^{-1} (Figure 5). Catchment carbon (hereinafter excluding carbon from carbonate) export from terrestrial to aquatic systems in the form of DIC species (8.3 g C m^{-2} a^{-1}) is about 8 times as much as in the form of OC species (1.1 g C m^{-2} a^{-1}). Such high

export loading of DIC from this catchment undoubtedly results from chemical weathering of carbonate. Due to high dissolution rates, carbonate dissolution rapidly and constantly consumes soil CO_2 to form HCO_3^- and exports HCO_3^- to streams, and then to the lake (Figure 5). By this mechanism, carbonate dissolution could markedly accelerate the processes of catchment carbon transfer from terrestrial to aquatic ecosystems and thus strengthen the IC connection between the two systems. The export of DIC takes part in carbon cycling in the lake, and then a portion of the DIC is exported downstream, and another portion is released to the atmosphere as CO_2, or is converted into OC that is buried in the sediment (Figure 5), highlighting the significance of catchment carbonate dissolution in terms of terrestrial carbon export.

At the catchment scale, the lake is a net producer of OC. Comparison between lake CO_2 evasion and OC input fluxes indicates that OC provided by the catchment is not likely the primary source of lake CO_2. DOC export from the catchment ($0.9 \, g \, C \, m^{-2} \, a^{-1}$) was much larger than POC ($0.2 \, g \, C \, m^{-2} \, a^{-1}$). However, DOC concentrations were low in stream waters, spring waters, and groundwater in the catchment (Wang et al., 2012). As a result, the low export load of OC cannot maintain the carbon emission and carbon budget of the lake. In comparison, the amount of DOC provided to the lake was slightly less than the lake CO_2 evasion and was also less than the downstream export of the lake ($1.55 \, g \, C \, m^{-2} \, a^{-1}$). Considering OC burial of $0.9 \pm 0.2 \, g \, C \, m^{-2} \, a^{-1}$ in the sediment, the lake is actually a net producer of OC, while simultaneously also emitting considerable amounts of CO_2 to the atmosphere.

Dissolved IC influx sustains gaseous CO_2 evasion and OC burial in the lake. Mass balance shows that the difference between DIC input and export in the lake ($2.0 \, g \, C \, m^{-2} \, a^{-1}$) approximately equals the total amount of CO_2 emitted ($1.0 \, g \, C \, m^{-2} \, a^{-1}$), plus OC buried in the sediment ($0.9 \, g \, C \, m^{-2} \, a^{-1}$), and the difference between OC export and input ($0.45 \, g \, C \, m^{-2} \, a^{-1}$; Figure 5). Small differences between the total carbon input to and exported from the lake are likely a product of data uncertainty and/or due to some carbon input that was not considered. As a whole, DIC input is likely a crucial factor in supporting CO_2 emission and OC burial in the lake. In a previous study, we identified three geochemical processes in the lake (Wang et al., 2015), by which HCO_3^- could be converted to CO_2 (equations (1)–(3) in Figure 5). In fact, there have been many studies over the past decade (Finlay et al., 2009; Jones et al., 2001; Marcé et al., 2015; McDonald et al., 2013; Weyhenmeyer et al., 2015) which argued that a portion of lake CO_2 was derived from catchment DIC input. As shown in Figure 5, it is clear that the DIC input to the lake originates from soil CO_2 (excluding carbon from carbonate). Therefore, it could be concluded that CO_2 in some lakes does not originate primarily from the input of soil organic matter from catchments, but from soil CO_2. The CO_2 emissions in this kind of lake are one way that soil CO_2 returns to the atmosphere, although there are often small differences in isotopic compositions and ages of CO_2 between that evaded from waters and that evaded directly from soils (Leith et al., 2014) and between that produced in surface versus deep soils (Ding et al., 2010). This view is in accord with the conventional carbon cycle that blends outgassing from aquatic systems with terrestrial respiration (see Battin et al., 2009).

3.4. Lake CO_2 Emissions Derived From Organic and Inorganic Origins Have Different Impacts on the Adjustment of Local Carbon Budget

The lake CO_2 derived from organic and inorganic origins has different sources and cycling pathways (Table 3) and thus requires different methods to integrate them into landscape carbon cycles (Figures 6a and 6b). If the lake CO_2 is produced by in-lake mineralization of soil organic matter input from the catchment, it belongs to exported ecosystem production and thus should be included in the estimate of terrestrial NEP (Figure 6a), as done in previous studies (Bastviken et al., 2011; Cole et al., 2007; Tranvik et al., 2009). Including groundwater and stream CO_2 evasion, the amounts of aquatic CO_2 emitted reached $4.4 \, g \, C \, m^{-2} \, a^{-1}$. This is equal to 55% of the catchment NEP (assuming an average NEP of $-8 \, g \, C \, m^{-2} \, a^{-1}$, Table 1). In the lake, the sum of CO_2 emission and OC burial equals 24% of the catchment NEP. These percentages indicate that aquatic CO_2 emission, given an organic origin, might account for a large proportion of catchment NEP, possibly altering the estimate of carbon balance of the terrestrial ecosystem. Release to the atmosphere of amounts of carbon equaling 13% of NEP (Figure 6a) is enough to exert influence on change of atmospheric CO_2 concentrations, as the result is extrapolated to the entire globe.

In comparison, lake CO_2 derived from inorganic origin has a completely different nature from that of organic origins (Table 3). Essentially, the CO_2 derived from inorganic origins belongs to soil CO_2 (as shown in Figure 5),

Table 3
Comparisons in Nature, Contribution, and Assessment of Lake Carbon Between Organic Origins and Inorganic Origins

For lake CO_2		Organic origins	Inorganic origins
CO_2 emission	Sources	Soil OC	Soil CO_2
	Attributes	NEP	Soil AR and HR
	Turnover rates in systems	Slow	Fast
	Residence times in systems	Long	Short
	Impact on adjusting terrestrial C budget	Significant impact	No significant impact
	How to integrate in landscape carbon cycling	Included in NEP	Included into soil CO_2 efflux
	Contribution on atmospheric CO_2 change	CO_2 source	No significant effect
For lake OC		Allochthonous OC	Autochthonous OC
OC b burial	OC mineralization	Carbon source	No significant effect
	OC escape mineralization (C sequestration)	Carbon sink	Net carbon sink
Net effect of lakes/reservoirs		Depending on the ratio of CO_2 efflux to OC burial	Depending on the difference between catchment NEP and OC burial

which is produced by autotrophic and HR in soils. NEP is restricted to OC balance and excludes IC (Chapin et al., 2006; Lovett et al., 2006). Therefore, lake CO_2 emissions derived from inorganic origin have no effect on the terrestrial NEP. Aquatic CO_2 evasion accounts for a very small percentage ($<0.1\%$) of soil CO_2 efflux (Figure 6b). With so small a proportion, lake CO_2 evasion has little potential to significantly alter the concentration of atmospheric CO_2. The comparison (Figures 6a and 6b) indicates that whether lake CO_2 evasion requires an adjustment of the terrestrial carbon budget, and alters atmospheric CO_2 concentrations, depends on carbon origins, even though the amounts of CO_2 emission are equal. The difference is caused by cycling rates of IC and OC in the catchment—lake system (Table 3). IC has a much faster turnover rate and far shorter residence times, relative to OC (Raich & Schlesinger, 1992). Especially, the carbon from root respiration has the fastest turnover rate and shortest residence time, lasting a few minutes to a few hours in the plant-soil system (see Hanson et al., 2000; Kuzyakov, 2006). Because of this, carbon used for root and rhizo-microbial respiration has no effect on long-term carbon balances (Kuzyakov, 2006). The significance of lake CO_2 evasion to local and global carbon budgets might be overstated if the CO_2 that has short residence time and little influence is regarded as the one that has long residence time and large potential to alter landscape carbon budgets (Table 3). Therefore, partitioning the
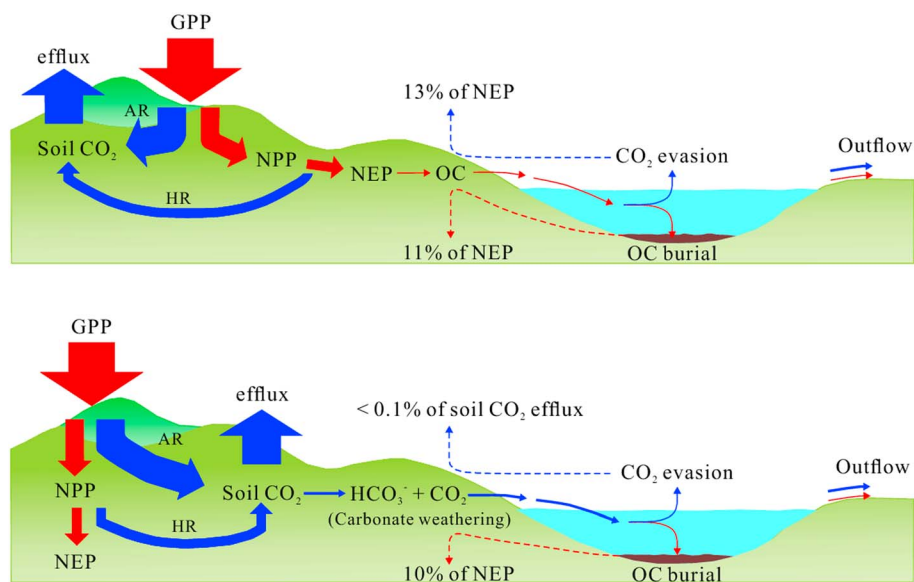
Figure 6. Comparisons of transfers and effluxes of organic (red arrows) and inorganic carbon (blue arrows) between the soil-air interface and the lake water-air interface, on the basis of (a) organic and (b) inorganic origins of lake carbon. The size of the arrows reflects relative magnitudes. The broken lines exhibit the carbon source/sink strength of the reservoir.

contribution of autotrophic and HR in soils and in-lake mineralization of soil OC is a crucial step to assess the effect of CO_2 emission on local carbon cycling.

### 3.5. OC Burial in the Lake Sediment Is a Net Sink

Allochthonous OC is a part of terrestrial primary production, while autochthonous OC stored in lake sediments might comprise a net carbon sink if atmospheric CO_2 is utilized in OC production by photosynthesis (Figure 6b). According to results from the nearby Puding reservoir, C/N ratios of allochthonous organic matter are approximately 45.0, and C/N ratios of autochthonous organic matter are about 5.0. If these ratios are regarded as end-members, it could be estimated using simple mixing models that ~85% of OC stored in the lake sediment was produced by photosynthesis within the lake, and just 15% was supplied from input of terrestrially derived OM (Figure 4). It is the HCO_3^- supplied from the catchment that was utilized during photosynthesis as a carbon source to form the autochthonous organic matter (Wang et al., 2015). This HCO_3^- originated from input of soil CO_2 dissolving carbonate, most of which was from root respiration with a very short residence times (Figure 6b). Burial of autochthonous OC in the lake sediment greatly extends the residence time of carbon, from hours to decades to thousands of years. Therefore, OC sequestration in the lake sediment could be a net sink of carbon. Expressed per catchment area, ~0.77 g m^{-2} a^{-1} of autochthonous OC was finally stored in the sediment as a net sink (Figure 6b), which is slightly lower than the lower range of values (1–4 g C m^{-2} a^{-1}) of atmospheric carbon withdrawal rates in lakes reported by Einsele et al. (2001).

3.6. Accurately Assessing the Roles of Lakes and Reservoirs in the Context of Organic and Inorganic Carbon Cycling

The methods of assessing the net environmental effect of lakes in terms of carbon source/sink strength differ depending on the origins of lake carbon. In previous studies, the ratio of gaseous carbon evasion to the atmosphere to OC burial in sediments was generally viewed as a value that characterizes the effect to be negative ($>$1), positive ($<$1), or neutral (~1) (Hanson et al., 2004; Mendonça et al., 2012). However, it should be noted that sources of aquatic carbon were not taken into consideration in estimating the ratio values.

In the scenario of a primarily OC origin (Figure 6a), CO_2 evasion in the lake would function as a source of atmospheric carbon, if stable OC in soils is decomposed at a faster rate. OC burial is not a sink of atmospheric CO_2 unless the decomposable OC in soils escape mineralization and is stored in the sediments for a long time. Simply, the ratio of CO_2 evasion to OC burial represents the vertical allocation of exported OC between decomposition and deposition and thus is able to reflect the role of lakes in deciding the fate of OC from the catchment (Hanson et al., 2004). The comparison between catchment NEP and aquatic CO_2 emission that has often been done in previous studies could reflect the ability of lakes to convert exported OC into gaseous carbon; therefore, these parameters could approximately reflect the source/sink strength of lakes, given the lake CO_2 is of organic origin.

In contrast, in the scenario of a primarily IC origin, aquatic CO_2 is produced by processes other than OC decomposition (equations (1)–(3) in Figure 5). CO_2 evasion could not be regarded as an effective source of atmospheric carbon, but OC burial is a net sink for atmospheric carbon. The ratio of gaseous carbon evasion and OC burial may not be an appropriate parameter for assessing the source/sink strength of the lake. We argue that the differences between NEP and OC burial might be a more reasonable proxy to denote the net environmental effect of the lake on a catchment scale (Figure 6b), given that lake CO_2 is primarily of inorganic origin. At Lake Hongfeng, the ratio of CO_2 evasion to OC burial is ~1.11 (1.1/0.9), suggesting that the lake acts as a weak net source of atmospheric carbon on the basis of prior standard (Hanson et al., 2004; Mendonça et al., 2012). However, the autochthonous OC burial rate of 43.4 g C m^{-2} a^{-1} (9.9 mmol m^{-2} d^{-1}, Table 1) in the lake sediment is larger than NEP (-20.0 to 0 g C m^{-2} a^{-1}) in the catchment, meaning that the reservoir has much greater carbon sequestration than the catchment (as long as the calculation is done on a per area basis). The rate of lake CO_2 emission sharply decreases relative to soil CO_2 efflux, and carbon sequestration markedly increases relative to NEP after impoundment (Figure 6b). Therefore, the reservoir plays an important role as a net sink of atmospheric carbon, if little or none of the emitted CO_2 came from mineralization of soil-derived OC in situ. This comparison demonstrates that different methods are needed to assess the source/sink strength of lake emissions and OC burial depending on carbon origins.
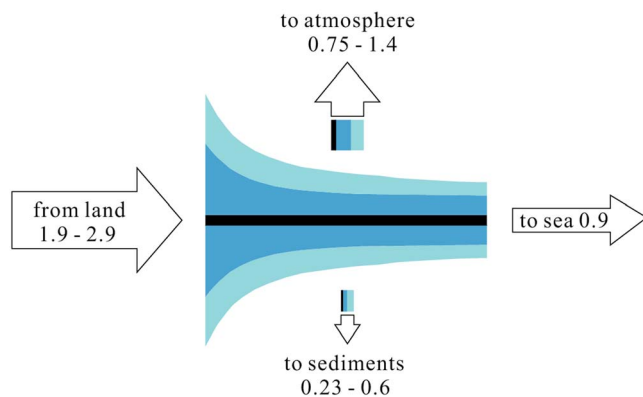
Figure 7. Partitioning aquatic carbon fluxes between organic and inorganic origins. The "active pipe" hypothesis was advanced by Cole et al. (2007) and revised by Aufdenkampe et al. (2011), Tranvik et al. (2009), and others (units: Pg C a^{-1}. The black color denotes carbon from carbonate, the dark gray color denotes carbon from inorganic origin, and the light gray color denotes carbon from organic origin. For data see Aufdenkampe et al. (2011), Cole et al. (2007), Nakayama (2017), Regnier et al. (2013), and Tranvik et al. (2009).

The conclusion drawn here has important implications concerning the significance of inland waters fluxes to the global carbon budget. While this research is based on study of only a single lake, the reality that lake carbon is, or can be, of predominantly inorganic origin is a likely condition in many catchments and lakes around the world. In carbonate areas, carbonate weathering drives soil CO_2 to transform into HCO_3^-, and that IC makes up a larger percentage of catchment carbon export. In fact, even where carbonates comprise as little as 5% of exposed bedrock, carbonate weathering still has an important influence on water chemistry (Barth et al., 2003). Tian et al.'s (2015) modeling shows that DIC flux exceeds the sum of DOC and POC fluxes in eastern North America. In many lakes around the world, input loading of DIC exceeds that of OC (Hanson et al., 2004; Tranvik et al., 2009), indicating that the influence of DIC on aquatic carbon balances cannot be neglected. NEP is restricted to the imbalance between GPP and ecosystem respiration; represents the total amount of OC in an ecosystem available for storage, export, and consumption by nonbiological oxidation; and explicitly excludes IC (Chapin et al., 2006; Lovett et al., 2006). The widely accepted "active pipe" hypothesis concerning inland waters advanced by Cole et al. (2007) and revised by Aufdenkampe et al. (2011) and Tranvik et al. (2009) does not include CO_2 derived from inorganic sources. But, in actual observations, the contribution of CO_2 from inorganic sources to aquatic CO_2 efflux must exist. Therefore, there must be a discrepancy between theoretical models and observational data.

One method of improving the conceptual model is to partition the aquatic carbon fluxes between organic and inorganic origins (Figure 7). The flux of carbon transferred from terrestrial ecosystems to sea, efflux of aquatic CO_2, and the flux of OC burial could be divided into three types: the carbon from carbonate (due to erosion), from inorganic origin, and from organic origin (Figure 7). Many studies have given the terrestrial export fluxes of these three kinds of carbon (e.g., Ludwig et al., 2011; Nakayama, 2017; Tian et al., 2015). However, their contribution to CO_2 efflux and OC burial is not well constrained.

The mechanisms controlling aquatic CO_2 emissions and OC burial remain poorly constrained (e.g., Barros et al., 2011; Cole, 2013; Marx et al., 2017; Nakayama, 2017; Raymond et al., 2013; Regnier et al., 2013; Tian et al., 2015). On average, just 28% of river CO_2 is from the mineralization of terrestrial OC input (Hotchkiss et al., 2015). In lakes, this proportion is likely to change over a large range. In subtropical areas where carbonates are widely distributed (Ludwig et al., 2011), like at Lake Hongfeng, the CO_2 sourced from inorganic origin is dominant. In boreal and Arctic/subarctic areas which have high DOC loads, the contribution of allochthonous OC may be as high as 85% (Hanson et al., 2004). Cole et al. (2002) and Hanson et al. (2004) found that 40–52% of in-lake respiration is supported by allochthonous OC. Combining the results from groundwater, stream water, and lake water, it is likely that <50% of the aquatic CO_2 evasion flux belongs to catchment NEP. Only this fraction of the flux should be considered in the estimate of catchment NEP. The carbon sourced from inorganic origin could be further divided into heterotrophic and autotrophic fractions. The heterotrophic fraction is derived from decomposition of soil OC; thus, this part of the flux could be considered in the estimate of terrestrial NPP, while the autotrophic fraction is derived from root-related respiration in soil and has no effect on long-term carbon cycling. A conservative estimate would be that ~ 50% of soil CO_2 is derived from root respiration (Hanson et al., 2000). This means that just a small fraction of the fluxes of aquatic CO_2 evasion could have a large influence on medium- and long-term changes of atmospheric CO_2 concentrations and on terrestrial carbon balances. A careful reassessment of the significances of CO_2 evasion and OC burial in freshwater ecosystems to local and global carbon budget, with full consideration of their sources, is necessary and pressing.

4. Conclusions

Lake Hongfeng emitted CO_2 and simultaneously sequestrated OC into the sediments at nearly the same rate. Overall, the rate of CO_2 emission across the lake water-air interface ranged from 7.0 to 19.0 mmol m^{-2} d^{-1},

with an average of 11.5 ± 2.6 mmol m^{-2} d^{-1} (1.0 ± 0.2 g C m^{-2} a^{-1}, per unit catchment area). Including groundwater and stream water CO_2 degassing, total aquatic CO_2 evasion in this catchment was as high as 4.4 g C m^{-2} a^{-1}. At the sediment-water interface, 69% of the OC delivered to the interface was remineralized during burial, and the remainder was stored in the sediment at a rate of 0.9 ± 0.2 g C m^{-2} a^{-1} (per unit catchment area).

Integrating the CO_2 evasion and OC burial in the lake in the context of inorganic and OC cycling in the catchment, we constructed a complete carbon budget in the catchment-lake system including all vertical and horizontal fluxes of IC and OC. Results show that carbon export from the catchment in the form of DIC species (8.3 g C m^{-2} a^{-1}) is about 8 times that in the form of OC species (1.1 g C m^{-2} a^{-1}). Carbonate dissolution rapidly and constantly consumed soil CO_2 to form HCO_3^-, which was subsequently delivered to waters, and accelerated the processes of catchment carbon transfer. Comparisons of carbon fluxes in the catchment suggest that the lake carbon primarily originated from soil CO_2 dissolving carbonate, rather than from OC from the catchment.

Carbon pathway-based analysis shows that aquatic CO_2 derived from organic and inorganic origins have different significance in terms of the terrestrial carbon budget. The sum of CO_2 emissions and OC burial in the lake equaled 24% of catchment NEP, meaning that lake carbon fluxes could alter the estimate of carbon balance of the terrestrial ecosystem if the carbon was derived from organic origin. However, lake CO_2 originated from soil CO_2 (and HCO_3^-), which accounts for a small percentage (<0.1%) of the total soil CO_2 efflux. Therefore, it has little potential to significantly alter the terrestrial carbon budget and atmospheric CO_2 concentration in medium and long term. This comparison indicates the importance of aquatic CO_2 evasion and the need to adjust the catchment carbon budget considering explicitly the origins of carbon.

Based on the ratio of gaseous carbon evasion to OC burial, Lake Hongfeng is a weak carbon source. In the scenario of a primarily IC origin, CO_2 evasion from the lake is not a significant source of atmospheric carbon, but OC burial is a net sink for atmospheric carbon. The differences between NEP and OC burial, instead of the above ratio, may be a more reasonable proxy to denote the source/sink strength of lakes. The autochthonous OC burial rate of 43.4 g C m^{-2} a^{-1} in the sediment is far larger than NEP (−20.0 to 0 g C m^{-2} a^{-1}) in the catchment, meaning that the reservoir actually plays an important role in serving as a net sink of atmospheric carbon. The conclusion suggests that aquatic carbon derived from different origins must be considered independently in understanding the role of lakes and reservoirs to local and global carbon budget.

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