

Volcanic carbon cycling in East Lake, Newberry Volcano, Oregon, USA

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ABSTRACT

The carbon cycle in East Lake (Newberry Volcano, Oregon, USA) is fueled by volcanic CO₂ inputs with traces of Hg and H₂S. The CO₂ dissolves in deep lake waters and is removed in shallow waters through largely diffusive surface degassing and photosynthesis. Escaping gas and photosynthate have low δ¹³C values, leading to δ¹³C(DIC) (DIC—dissolved inorganic carbon) as high as +5.7‰ in surface waters, well above the common global lake range. A steep δ¹³C depth gradient is further established by respiration and absorption of light volcanic CO₂ in bottom waters. The seasonal CO₂ degassing starts at >100 t CO₂/day after ice melting in the spring and declines to ~40 t/day in late summer, degassing ~11,700 t CO₂/yr. Thus, volcano monitoring through gas fluxes from crater lakes should consider lacustrine processes that modulate the volcanic gas output over time. The flux contribution of a bubbling CO₂ “hotspot” increased from 20% to >90% of the lake-wide CO₂ flux from 2015 to 2019 CE, followed by a “toxic gas alert” in July 2020. East Lake is an active volcanic lake with a “geogenic” ecosystem driven primarily by hydrothermal inputs.

INTRODUCTION

Some volcanic lakes have active CO₂ inputs (e.g., Chiodini et al., 2012; Christenson and Tassi, 2015), and a few have experienced explosive episodes in the past (e.g., Lake Nyos, Cameroon; Kusakabe, 2017). Volcanic CO₂ negatively impacts some oceanic ecosystems (e.g., Carey et al., 2013; Price et al., 2015), and CO₂ emissions in the volcanic Mammoth Lakes (California, USA) area (e.g., Bergfeld et al., 2006) kill local forests. However, some ecosystems utilize volcanic inputs (Cabassi et al., 2013). The incoming volcanic CO₂ is processed by the lentic ecosystem, but most of it escapes through surface degassing (e.g., Caudron et al., 2012; Andrade et al., 2016). This CO₂ lake flux is commonly used in volcano monitoring (e.g., Rouwet et al., 2014; Mazot and Bernard, 2015; Varekamp, 2015).

Newberry Volcano (Oregon, USA; 43.728°N, 121.210°W) is a Cascade Range back-arc volcano (Carlson et al., 2018), most recently active 1300 yr ago, with two small crater lakes aged ca. 7.5 ka (Jensen and Donnelly-Nolan, 2017). East

Lake is a drowned crater with visual evidence of CO₂ inputs from the lake bottom (Lefkowitz et al., 2017). The CO₂ bubbles carry traces of H₂S and Hg gas but almost no fluids. A bubbly CO₂-Hg-H₂S “hotspot” is found along the southeastern beach (Fig. 1). East Lake has no inlets or outlets, a maximum water depth of 55 m, and a 4.2 km² surface area. It is frozen in winter and thermally stratified in summer (Lefkowitz et al., 2017). This study aims to determine the magnitude of the volcanic CO₂ input in this carbonate-rich volcanic lake based on 5 years of flux measurements and to map the local carbon cycle based on carbon isotope evidence.

METHODS

We measured lake surface CO₂ fluxes using an accumulation float chamber (e.g., Mazot and Bernard, 2015) and a LI-COR CO₂ detector (model LI-6252). Field samples and measurements were collected annually between June and August from 2015 to 2019, with additional data collected in May 2018 shortly after ice melting. The field data points were treated with se-

quential Gaussian simulations (SGSs; Cardellini et al., 2003) to estimate lake-wide CO₂ release rates. Gas samples from the accumulation chamber and ambient CO₂ from air on land and from above the lake were injected into pre-evacuated Exetainer vials for stable isotope analyses. Gas bubbles in the hot-spring pools were collected through water displacement in inverted plastic bottles. Lake depth profiles for pH and temperature were obtained with a YSI Professional Plus five-probe analyzer. Lake water samples were taken at 10 m depth intervals with a Teflon Wildco 2 L water sampler and stored in Exetainers for δ¹³C(DIC) (DIC—dissolved inorganic carbon) analyses after filtration (0.2 μm). Additional details on methods are provided in the Supplemental Material¹.

RESULTS

The CO₂ escape rates measured between late June and August were similar in 2015–2019, with average CO₂ fluxes of ~0.2 mol/m²/day. The SGS data treatment provided a mean value of 46 ± 10 t CO₂/day as the typical summer CO₂ evasion rate (Fig. S1 and Table S1 in the Supplemental Material). The bubbly CO₂ “hotspot” increased in size and intensity over 2015–2019 without impacting the overall flux (see the Supplemental Material). The hotspot flux contribution increased from 20% to >90% of the lake-wide CO₂ flux, which culminated in a “gas alert” in July 2020 (USGS, 2020). The mid-May 2018 CO₂ flux was ~95 t CO₂/day, well outside the summer range and with a different spatial flux pattern. More than 50% of the lake area was emitting >0.5 mol/m²/day, whereas the other surveys reached such values over ~15% of the lake area only, largely in the hotspot area.

¹Supplemental Material. Details on the laboratory, data analysis, modeling methods, and equipment specifications, including methods and results of the two box models showing seasonal carbon cycling in East Lake, and raw data from the main text figures. Please visit <https://doi.org/10.1130/GEOL.S.14046902> to access the supplemental material, and contact editing@geosociety.org with any questions.

CO₂ degassing at East Lake

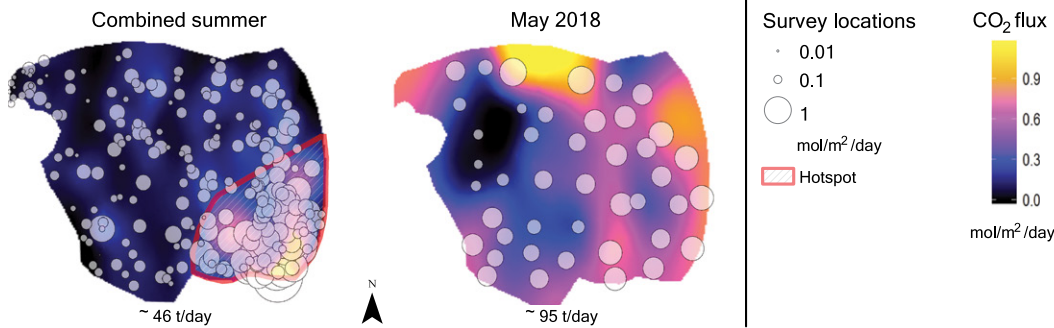


Figure 1. Five-year time-integrated summer CO₂ flux pattern at East Lake (Oregon, USA; 43.7306°N, 121.2098°W), combining 256 flux measurements, compared to the pattern in May 2018. Sample point sizes are scaled to measured flux. Sequential Gaussian simulation color scale is located to the right. Hotspot region is outlined in red. In May 2018, the flux had a different distribution pattern and larger overall flux. There were many zones of high CO₂ flux, and the hotspot no longer stood out.

Gas samples, with 420–800 ppm CO₂, taken from the float chamber during CO₂ accumulation runs, are mixtures of ambient and lake CO₂, with δ¹³C(CO₂) from –8‰ to –17‰ (Fig. 2). The δ¹³C(CO₂) in ambient gas samples (air) ranged from the fully mixed atmospheric value of ~–8.7‰ at ~410 ppm CO₂ (NOAA, 2020)

to values of –17‰ at 700 ppm CO₂. In some experiments, we pumped the air component out of the chamber and after some accumulation time, the chamber contained only lake gas, with δ¹³C(CO₂) values of –9‰ to –15‰. The δ¹³C(CO₂) of hot-spring bubbles ranged from –3‰ to –8‰ (Table S2).

Surface-water pH values varied over the years of record from ~7.4 in May to 8 in September, with pH values down to ~6.4 in the hypolimnion. Lake-water profiles show increasing DIC and conductivity with depth (Lefkowitz et al., 2017), mainly due to CO₂ gain from volcanic CO₂ absorption and respiration at depth and CO₂ loss from diffusive CO₂ escape and photosynthesis in the epilimnion.

The δ¹³C(DIC) in the epilimnion varied over the years of the survey from +2.5‰ to +5.7‰, with May 2018 at 3.5‰ and the highest values in late summer (Table S3). Hypolimnial values were lower and ranged from –0.5‰ to +4‰ (Fig. 3). The isotopic depth gradient [Δδ¹³C(DIC)] also differed over the season, with the smallest Δδ¹³C(DIC) values in May 2018 and June 2015–2016 (0.5‰–1‰), and the largest (4‰–5‰) in late summer over the years surveyed (Fig. 3).

The ¹⁴C contents of DIC in surface water and at 42 m depth collected in summer 2015 (Table S4) was 24.7% and 28.8% of modern ¹⁴C, respectively, both much lower values than in lakes in open exchange with the atmosphere (e.g., Taibei et al., 2018).

East Lake sediment carries 2%–12% C_{org} (org—organic) from diatoms, cyanobacteria, and subaquatic vegetation debris (SAV). Cores taken near the CO₂-rich bubbling zone have the highest C_{org} (8%–12%; see the Supplemental Material). The δ¹³C(C_{org}) ranges from –17‰ (rich in SAV) to –24‰, with a mean value of –23‰ in surface sediments (Lefkowitz et al., 2017). Mercury concentrations in cores and grab samples throughout the lake range from 0.5 to 4 ppm Hg (Lefkowitz et al., 2017), whereas cores in and around the hotspot have 3–13 ppm Hg (see the Supplemental Material).

DISCUSSION

The data provide a 5 yr record of carbon cycling in East Lake, with implications for its ecosystem functioning and volcano monitoring. Spring melting of the winter ice cover leads to lake overturn, intense CO₂ degassing

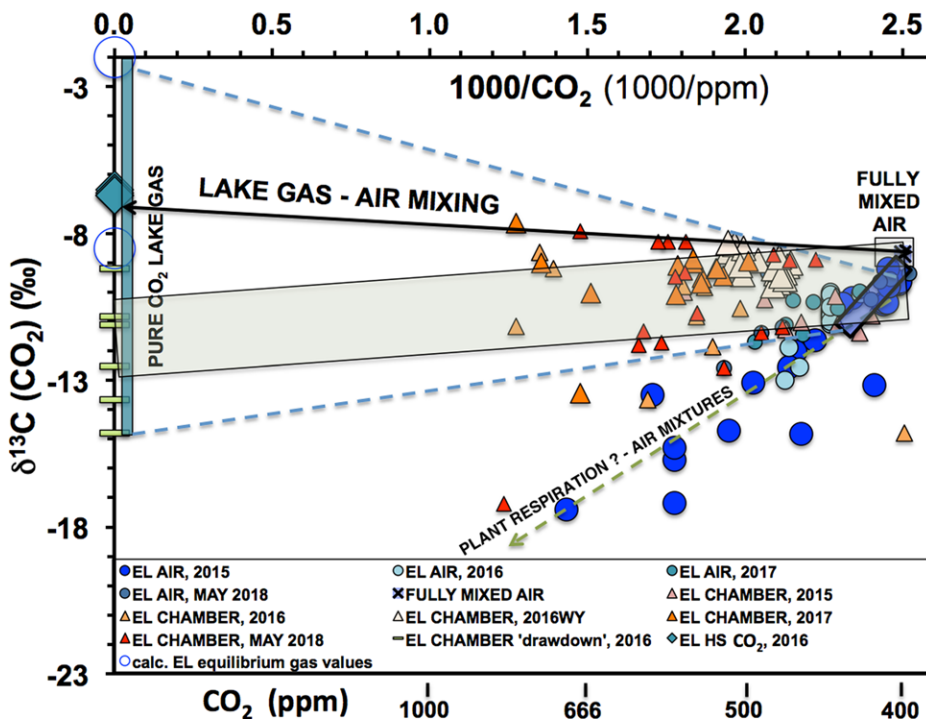


Figure 2. Keeling mixing diagram with concentrations and δ¹³C values of CO₂ in ambient air (filled circles) and in accumulation chamber gas (triangles) from our sampling campaign at East Lake (EL; Oregon, USA; 43.7306°N, 121.2098°W). δ¹³C values of “pure lake gas” plot at zero on the x-axis. Air plots on the right-hand side of diagram (~400 ppm), with blue bar indicating the range of δ¹³C values from air samples taken 5–15 km upwind from the lake. Air data array represents mixing between common air and plant respiration, boat exhaust gas, lake gas above the lake surface, and possibly forest-fire CO₂. Large blue open circles represent predicted equilibrium isotope compositions of lake gas. Light-green “drawdown” symbols (flat bars) stem from experiments where ambient CO₂ had been pumped out of the chamber. Chamber data represent mixtures of ambient CO₂ and possible lake gas compositions (vertical teal bar). The 2016WY data were analyzed at the University of Wyoming (USA), all other data are from the University of California–Davis (USA); HS samples refer to CO₂ bubbles collected from hot springs. Likely mixing array that covers ~75% of the data is shown with a pale green band, indicating mixing of slightly contaminated air with “pure lake gas” that has δ¹³C values of –10‰ to –12.5‰. Thick black line represents mixing between standard fully mixed atmospheric CO₂ with an equilibrium lake gas (HS sample); dashed light-blue mixing lines represent the possible extremes of air-lake gas mixing; dashed green line is the air CO₂–biogenic CO₂ mixing line.

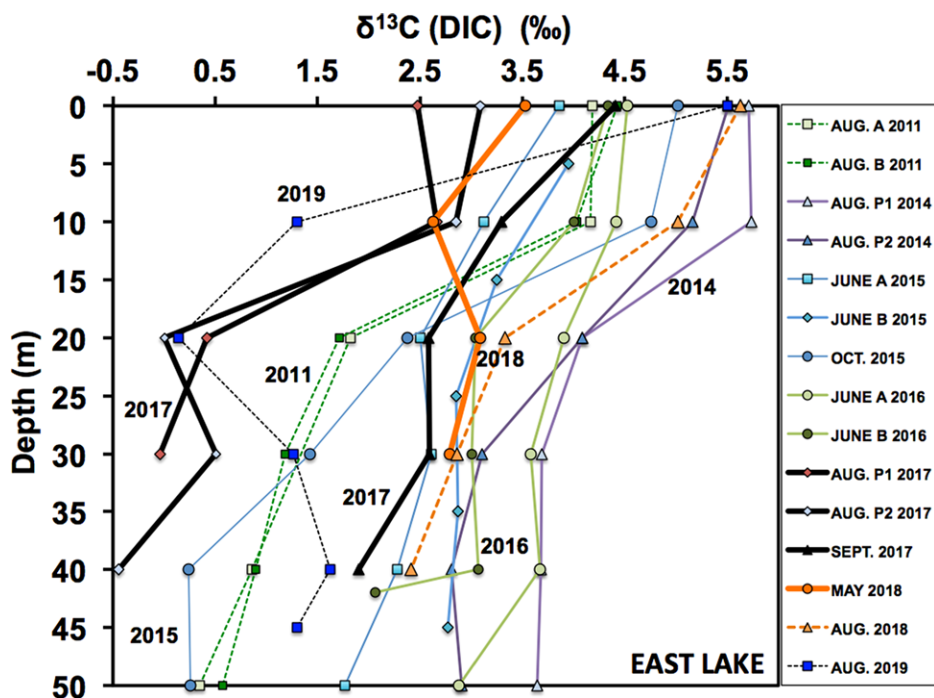


Figure 3. Depth versus $\delta^{13}\text{C}(\text{DIC})$ (DIC—dissolved inorganic carbon) trends at East Lake Oregon, USA (43.7306°N, 121.2098°W) over several years, taken in different months (May–August). A, B, P1, and P2 refer to different locations of depth profiles taken in the same sampling year. Slopes are variable: almost vertical in May, and stronger gradients in August. Thick black and orange lines are discussed in the text.

(>100 t CO₂/day), and the onset of algal productivity. In the fall, the CO₂ flux diminishes to <40 t/day, then the lake again turns over, followed by freezing of surface waters with the cessation of photosynthesis and CO₂ surface degassing. During the winter, the lake accumulates dissolved volcanic CO₂.

We fitted a power-law function to the CO₂ flux data (Fig. 4) to model seasonal CO₂ escape rates (Supplemental Material), showing that excess winter CO₂ is “blown off” from April through late August. In late summer, the lake evolves toward a steady state with volcanic CO₂ output roughly equaling the volcanic

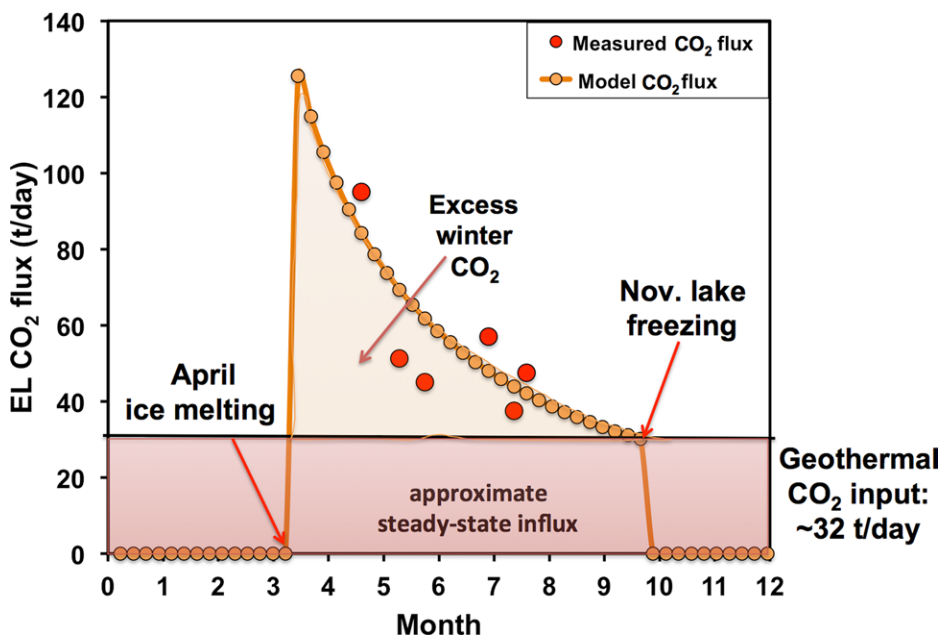


Figure 4. Seasonal CO₂ degassing trend at East Lake (EL; Oregon, USA; 43.7306°N, 121.2098°W) with best fit power-law trend to lake-wide average flux.

input, estimated at ~32 t CO₂/day year-round (Supplemental Material). The lake CO₂ loss of ~11,700 t/yr is comparable to those from other similarly sized volcanic lakes (Pérez et al., 2011). Over these 5 yr of study, the hotspot growth was most likely caused by a drop in lake level, driven by higher ambient temperatures and lower precipitation levels (NIDIS, 2020). A lower lake level created a larger area across which bubbles could reach the surface and discharge directly into the atmosphere. The 2020 “gas alert” thus was not driven by deep-seated volcanic degassing processes (USGS, 2020).

East Lake has high epilimnial $\delta^{13}\text{C}(\text{DIC})$ values compared to typical global $\delta^{13}\text{C}(\text{DIC})$ lake values (−20‰ to 0‰; Bade et al., 2004) but similar to some other CO₂-degassing carbonate-rich volcanic lakes (e.g., Mazot et al., 2014). High $\delta^{13}\text{C}(\text{DIC})$ occurs in nonvolcanic lakes with extensive methane generation (e.g., Gu et al., 2004) or strong seasonal algal blooms (e.g., Oren et al., 1995), which are both absent in East Lake. $\Delta\delta^{13}\text{C}(\text{DIC})$ broadly increases over the season as a result of increasing $\delta^{13}\text{C}(\text{DIC})$ in surface waters and decreasing $\delta^{13}\text{C}(\text{DIC})$ in deeper waters. $\delta^{13}\text{C}(\text{DIC})$ in the epilimnion can increase by ~2‰ (e.g., between May and August 2018), while deep-water $\delta^{13}\text{C}(\text{DIC})$ can decrease by several per mil after spring homogenization. $\Delta\delta^{13}\text{C}(\text{DIC})$ is created by photosynthesis and CO₂ degassing in the epilimnion and by respiration and addition of low- $\delta^{13}\text{C}$ volcanic CO₂ in the hypolimnion. The $\delta^{13}\text{C}(\text{DIC})$ depth profiles show the fully developed gradient in August 2017, a lesser gradient in September (storm-related lake mixing), and then near fully mixed conditions in May 2018 after ice melting, indicative of lake turnover (Fig. 3, thick black and orange lines).

The ¹⁴C(DIC) data provide apparent water ages of >10,000 yr (Table S4), but East Lake is only 6500 yr old. Water-budget modeling suggests a water residence time of ~20 yr (Lefkowitz et al., 2017). The CO₂-degassing surface waters do not equilibrate with atmospheric CO₂, and the large flux of “dead” volcanic CO₂ strongly dilutes the atmospheric ¹⁴C input that presumably enters the lake largely through precipitation.

The measured $\delta^{13}\text{C}(\text{CO}_2)$ values from the accumulation chamber range from −8‰ to −17‰ and cannot be explained as binary mixtures of lake CO₂ gas in equilibrium with lake DIC and fully mixed atmospheric ambient CO₂ (−8.7‰). Equilibrium $\delta^{13}\text{C}(\text{CO}_2)$ values for lake gas were calculated from isotope mass-balance statements, temperature-dependent intra-species fractionation factors (DeVries et al., 2001), and speciation calculations using the program CO₂Sys (Pierrot and Wallace, 2006). The epilimnial $\delta^{13}\text{C}(\text{DIC})$ values (+2.5‰ to +5.7‰), temperature (0–18 °C), and pH (7–8.2) provided $\delta^{13}\text{C}(\text{CO}_2)$ equilibrium values of −2.5‰ to −8.5‰.

Binary mixing of standard ambient air with an equilibrium lake gas composition of -7‰ $\delta^{13}\text{C}(\text{CO}_2)$ would create a mixing line at the upper section of the data array (Fig. 2, thick black line). Our analyses of local ambient air show a range between fully standard air and a low- $\delta^{13}\text{C}$ component, such as forest and/or ground respiration CO_2 at $\sim -27\text{‰}$ (Bowling et al., 2002; Chiodini et al., 2008, 2011). Newberry summer nights can be below freezing, leading to the formation of a nocturnal atmospheric boundary layer. Soil and forest CO_2 emissions trapped in the atmospheric ground layer may create the observed low $\delta^{13}\text{C}$ values in CO_2 -enriched air (Fig. 2). In addition, low- $\delta^{13}\text{C}$ CO_2 may have been contributed by the common regional forest fires and powerboat exhaust gases, and air collected above the lake may have a lake gas component. The range in $\delta^{13}\text{C}(\text{CO}_2)$ of potential pure lake gas samples (Fig. 2, teal green bar) spans the calculated equilibrium gas values and the “pure lake gas” chamber samples (light green bar symbols). The latter may be explained with a kinetic isotope effect relative to the calculated equilibrium values. Mixing between the “pure lake gas” samples and slightly contaminated air covers the majority of data points (pale green band), but this solution is not unique.

Thus, the escaping CO_2 has low $\delta^{13}\text{C}$ values, with $\delta^{13}\text{C}(\text{CO}_2)$ a function of lake composition and a degree of kinetic fractionation that usually depends on the wind speed. The loss of this CO_2 gas is one driver for the heavy $\delta^{13}\text{C}(\text{DIC})$ in shallow waters and the depth gradients that build up over the season.

The second carbon sink from the epilimnion is the photosynthetic flux, which we constrain by the C_{org} burial rate. Only a fraction of the photosynthetic flux is buried; the rest is recycled in the hypolimnion through respiration and oxidation (Cole et al., 1994). Primary organic productivity thus also depletes the DIC in ^{12}C in epilimnial waters. We calculated the C_{org} burial rate from C_{org} data, the mean sediment mass accretion rate ($\sim 0.05 \text{ g/cm}^2/\text{yr}$) based on core-top ^{210}Pb ages (Lefkowitz et al., 2017) and a volcanic ash age deeper in one core (ca. 1300 yr B.P. Paulina Lake ash flow layer; Jensen and Donnelly-Nolan, 2017), and bulk dry sediment density data. The mean lake C_{org} burial rate is $\sim 3.3 \text{ mg C/cm}^2/\text{yr}$, translating into a lake-wide C_{org} burial rate of $140 \pm 30 \text{ t C/yr}$.

A preliminary two-box model using the measured carbon fluxes and calculated equilibrium isotope fractionation factors shows that the calculated $\delta^{13}\text{C}(\text{DIC})$ values and isotope gradients broadly match the whole observed $\delta^{13}\text{C}(\text{DIC})$ spectrum (Supplemental Material). The data and modeling indicate that diffusive CO_2 degassing strongly contributes to $\Delta\delta^{13}\text{C}(\text{DIC})$. A carbon-isotope depth gradient is thus not a precise indicator of primary lake productivity

(e.g., McKenzie, 1985) if surface CO_2 degassing occurs as well.

The CO_2 -rich hot-spring bubbles possibly contain primary volcanic CO_2 as suggested by He mantle isotope values (ratio relative to the atmospheric value, $R_a = 7.6\text{--}8.3$; Graham et al., 2009). The $\delta^{13}\text{C}(\text{CO}_2)$ in discrete bubbles was determined at -3‰ to -8‰ , and the box modeling (see the Supplemental Material) demands an input value of $\sim -6\text{‰}$ to -7‰ , close to general mantle CO_2 values (Deines and Gold, 1973).

The phosphorus for photosynthesis and silicon for diatom frustule construction are supplied by the geothermal fluids, and fixed nitrogen is supplied by diazotroph cyanobacteria (Lefkowitz et al., 2017). The high levels of CO_2 may stimulate the local primary productivity (Hamdan et al., 2018), and the CO_2 -rich hotspot area has the highest C_{org} contents (as much as 12%; see the Supplemental Material).

Consequently, the lake ecosystem benefits from hydrothermal inputs and is highly productive ($6\text{--}12\%$ C_{org} in ash-poor sediment). The high sedimentary Hg levels (Lefkowitz et al., 2017) have no major deleterious impact on the ecosystem, although plankton tows yielded 5 ppm Hg and fish with low parts-per-million Hg values (see the Supplemental Material).

CONCLUSIONS

East Lake is not a North American version of Lake Nyos in Cameroon, where CO_2 accumulated over decades until a catastrophic release took place (e.g., Kusakabe, 2017). At East Lake, accumulated winter CO_2 is released every year during spring and summer. Seasonal ice cover blocking diffusional CO_2 escape occurs in many high-latitude and high-altitude lakes (e.g., Cole et al., 1994). This delay in the release of winter CO_2 may mute the seasonal oscillation in atmospheric CO_2 (Tranvik et al., 2009). Volcano monitoring through gas flux measurements in volcanic lakes must account for lacustrine processes that modulate the gas flux, especially when ice cover occurs. During low-wind periods, the lake is most likely degassing CO_2 with low $\delta^{13}\text{C}(\text{CO}_2)$ as a result of kinetic fractionation during degassing, whereas during windy periods, the CO_2 flux increases (see the Supplemental Material) and $\delta^{13}\text{C}(\text{CO}_2)$ becomes close to the equilibrium isotopic composition. The accumulation-chamber data are not an exact replica of natural CO_2 degassing because of the protected environment inside the chamber.

East Lake has a “geogenic” ecosystem that is almost entirely fed by volcanic nutrients, including CO_2 . Its waters have high $\delta^{13}\text{C}(\text{DIC})$ values due to CO_2 degassing and photosynthesis. East Lake’s system is dualistic in nature with a hydrothermal supply of good nutrients and harmful toxins, where the good far outpaces the bad for the ecosystem.

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