Influence of the ratio of planktonic to benthic diatoms on lacustrine organic matter $\delta^{13}C$ from Erlongwan maar lake, northeast China

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ABSTRACT

Carbon isotope ratio ($\delta^{13}C_{org}$) values of organic matter in lake sediments are commonly used to reconstruct environmental change, but the factors which influence change are varied and complex. Here we report $\delta^{13}C$ values for sediments from Erlongwan maar lake in northeast China. In this record, changes in $\delta^{13}C$ cannot be explained by simple changes in aquatic productivity. Instead, values were likely influenced by differences in the ratio between planktonic and benthic algae, as indicated by the remains of diatoms. This is because the variation of $\delta^{13}C_{org}$ in algae from different habitats is controlled by the thickness of the diffusive boundary layer, which is dependent on the turbulence of the water. Compared with benthic algae, which grow in relatively still water, pelagic algae are exposed to greater water movement. This is known to dramatically reduce the thickness of the boundary layer and was found to cause even more severe $\delta^{13}C$ depletion. In Erlongwan maar lake, low values were linked to the dominance of planktonic diatoms during the period commonly known as the Medieval Warm Period. Values gradually increased with the onset of the Little Ice Age, which we interpret as being driven by an increase in the proportion of benthic taxa, due to effects of the colder climate. The increase in planktonic diatoms at the end of the Little Ice Age, linked to higher temperature and a reduction in ice cover, resulted in a further decline in $\delta^{13}C_{org}$.

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1. Introduction

Carbon isotope ratio ($\delta^{13}C$) values are an important proxy for reconstructing environmental change from lakes, such as changes in temperature, precipitation patterns and evaporation. These changes are all indirectly linked to the carbon cycle (Leng and Marshall, 2004). However, interpretation of $\delta^{13}C$ ($\delta^{13}C_{org}$) from bulk organic matter (OM) in lake sediments is complex and dependent on many factors. It is commonly used in palaeoecological studies to determine OM sources in a lake, e.g. allochthonous vs. autochthonous sources (Meyers, 1994), the source of dissolved carbon used by aquatic plants (Leng et al., 2006), productivity (Leng and Marshall, 2004) and catchment soil respiration (Hammarlund, 1993; Mackay et al., 2012), as well as anthropogenic carbon released into the environment because of fossil fuel combustion (the Suess effect; Keeling, 1979). As far as we are aware, little work has been done to explore the influence of varying ratio of phytoplankton and benthic algae on $\delta^{13}C$ in lake sediments, despite the environmental importance that changes in this ratio represent in terms of lake level variability, hydrodynamics of the water column and habitat availability (France, 1995).

Here we discuss the possible reasons for $\delta^{13}C_{org}$ variability in the sedimentary record from Erlongwan maar lake (EML) over the past 1000 yr, using, proportion of sedimentary total organic carbon (TOCs), TOC flux, C/N ratio, and the ratio between planktonic and benthic algae as indicated by changes in the composition of diatom assemblages and diatom flux.

Diatoms are characterized by siliceous cell walls, which are generally well preserved as sedimentary fossils (Battarbee et al., 2001). In lakes, diatoms are commonly classified as planktonic, i.e. suspended, unattached in open water, or benthic (i.e. living on the bottom of the lake floor)/periphytic (i.e. attached to substrates such as stones, aquatic vegetation and sand). These habitats have different micro-environmental conditions, such as light availability and exposure to water turbulence, which control nutrient availability. We discuss how turbulence may play an important role in controlling carbon isotope fractionation in different diatom habitats.
There are eight maar lakes in the Long Gang Volcanic Field (LGVF) region in Jilin Province, NE China. Our study focused on Lake Erlongwan (EML) 42°18′N, 126°21′E, a dimictic lake, that occupies an area of ca. 0.3 km² at 724 m above sea level (Fig. 1). It is a closed lake, with a small catchment (0.4 km²) and no natural inflow or outflow (Mingram et al., 2004).

The modern natural vegetation in the LGVF is typical of the temperate mixed coniferous–deciduous forest zone (Editorial board for Flora of China, 1995). In this forest, the conifer Pinus koraiensis is dominant. The late Holocene pollen assemblage from EML (Li et al., 2012) also reflects the dominance of a mixed conifer–hardwood and deciduous forest around the EML region. Over the last 1000 yr, Pinus was the dominant conifer, while major broad-leaved deciduous trees included Quercus, Betula, Juglans, Ulmus, Carpinus, Corylus, Tilia and Fraxinus.

In 2001, a 66.5 cm long core was extracted from the central, deepest region of the lake at a water depth of 36 m. The core was laminated from 0 to 50 cm. The sediments are composed of detrital mud with varying amounts of OM. Between 51 and 66.5 cm the sediments are graded, likely representing a rapidly deposited sediment slump (Frank, 2007). We focus therefore only on the laminated record of the top 50 cm.

The age model was developed by combining results from 210Pb and 14C radiometric dating (Fig. 2). The activity of 137Cs and 210Pb were measured using gamma spectrometry with a low background well-type Ge detector (EGPC 100P-15R) at the Institute of Geology and Geophysics, Chinese Academy of Science, Beijing. The 14C date (Poz-19967) was analysed on a Pinus macrofossil found at the bottom of the core (49.5 cm) using accelerator mass spectrometry (AMS). AMS gave a date of 1045 ± 30 14C yr BP (Fig. 2), which, after two sigma calibration, gave an age range of 900–1030 AD (Wang et al., 2012b). The ages of the samples below those dated with 210Pb and above the radiocarbon date at 49.5 cm were linearly interpolated. Linear extrapolation was also used to extend the age model to the base of the core at ca. 980 AD (Fig. 2).

Diatom samples were prepared according to standard methods (Battarbee et al., 2001; Li et al., 2009). Diatom concentration (valves/g) were calculated by way of addition of divinylbenzene microspheres (Battarbee and Kneen, 1982). Diatom identification followed the guidelines set out by Krammer and Lange-Bertalot (1982). We chose to merge Discostella pseudostelligera, D. woltereki and D. stelligera into “Discostella species” because these three taxa were difficult to distinguish consistently under light microscopy (Haworth and Hurley, 1984; Tanaka, 2007). The diatom data are displayed as relative abundance (%) and total dia-
3. Results

Organic content of the sediments (inferred from TOC flux) is above average for the core (0.012 g/cm²/yr) from 50 to 47 cm (ca. 970–1080 AD) and increased to a peak between 31 and 36 cm (ca. 1070–1280 AD; Fig. 3). Thereafter, TOC flux declined steadily, concurrent with a decrease in diatom flux (Fig. 3). TOC flux values subsequently increased at 10 cm (ca. 1900 AD), as did diatom flux and the proportion of planktonic diatom species (Fig. 3). TOC correlated most significantly with TN flux (0.990; Table 1), and significantly positively with diatom flux (0.747; Table 1; Fig. 4). TOC and diatom flux are highly and significantly correlated (0.422, i.e. increasing abundance of planktonic diatoms (Table 1; Fig. 4). C/N values fluctuated between 10.6 and 15.6. The highest C/N values occurred between 26 and 13 cm (ca. 1500–1780 AD; Fig. 3).

The $\delta^{13}$C values fluctuated between $-29.3\%$ and $-28.0\%$ from 50 cm to 17 cm, but showed a gradual and distinct increase ($3.4\%$) from 33 cm ($-29.0\%$) to 12 cm ($-25.6\%$; ca. 1350–1800 AD; Fig. 3). From 33 cm to 12 cm (ca. 1350–1800 AD), increasing $\delta^{13}$C values occurred as TOC flux, diatom flux and P/B ratio declined (Fig. 3). For the whole profile, $\delta^{13}$C and TOC flux are negatively correlated ($-0.314$; Table 1). Above 12 cm (after ca. 1800 AD), $\delta^{13}$C values rapidly declined to the lowest values in the profile (ca. $-29.8\%$), between 10 cm and 9 cm, coincident with a distinct increase in the proportion of planktonic diatom species. More specifically, the shift in diatom composition appeared to be driven by a rapid decline in Discostella species and an increase in the planktonic species Puncticulata praetermis and Asterionella formosa (Fig. 5). Overall, $\delta^{13}$C values correlated most significantly with P/B ratio ($-0.627$; Table 1; Fig. 4); $\delta^{13}$C values in the uppermost sediments, spanning the last 50 yr (ca. 6 cm), increased up to the time the core was taken (Fig. 3), coincident with the return to dominance of the Discostella species (Fig. 5).

4. Discussion

All the samples have $\delta^{13}$C values below $-25\%$ (Fig. 3), indicating the dominance of $C_3$ plants as the principal carbon source to EML. $C_3$ plants utilize RubisCO (ribulose-1,5-bisphosphate carboxylase) to fix atmospheric CO$_2$, which results in $\delta^{13}$C values between $-22\%$ and $-33\%$ (O’Leary, 1981). This photosynthetic pathway is the most common among plants and is used by most trees, shrubs, aquatic vegetation and algae. Pollen evidence from the sediments highlights the idea that extensive pine and deciduous forest cover has persisted throughout the past 1000 yr (Mingram et al., 2004).

The $\delta^{13}$C values of bulk sediments cannot be used to distinguish between algae and terrestrial $C_3$ plants (Meyers and Lallier-Vergès, 1999), so C/N ratio in lake sediments is commonly used to assess the relative importance between allochthonous and autochthonous OM sources (Meyers, 1994); algae generally contain only a small amount of cellulose or lignin (both carbon rich; Leng et al., 1999), so C/N ratio of lake sediments are commonly used to assess the relative importance between allochthonous and autochthonous OM sources (Meyers, 1994); algae generally contain only a small amount of cellulose or lignin (both carbon rich; Leng et al., 2006). Fig. 3 shows that all the samples have C/N ratio < 16, with an average value of 12.8. It is likely that the plant OM in EML is derived from a mixture of sources, especially algae (with relatively low C/N ratio 5–10; Meyers and Lallier-Vergès, 1999) and aquatic vegetation (both submerged and floating) with relatively high C/N ratio (ca. 20 to 30; Fellerhoff et al., 2003) and only a small contribution from terrestrial vegetation (C/N between 20 and 160; Meyers and Lallier-Vergès, 1999).

OM input from terrestrial vegetation is likely to be low because TOC and diatom flux are highly and significantly, correlated (0.747; Table 1 and Fig. 4), which indicates that algae are a major component of OM in the core. Secondly, the highest C/N values between 28 and 12 cm (1500–1800 AD; Fig. 3) are unlikely caused by...
increased in-wash of terrestrial vegetation to the lake, because pollen concentration data suggest a period of relatively drier summers and a less dense vegetation cover (Mingram et al., 2004; Li et al., 2012) during that interval. Previous studies of EML diatom data (Wang et al., 2012b) and historical documents relating the frequency of snow events in NE China (Chu et al., 2008) shows that a cold climate prevailed for this interval. Third, although the abnormal changes in $^{210}$Pb and the high sediment accumulation rate at the top of the core (5–0 cm; Fig. 2) indicate that there was an increase in run-off input to the lake during the most recent part of the core, the values for the C/N ratio are still low (Fig. 3), indicating that the proportion of allochthonous input to the lake has remained small.

Relatively high C/N values during the cold period (1500–1800 AD) do coincide with an increase in benthic taxa and a concomitant decline in P/B ratio, which were controlled by the duration of ice cover. Longer ice cover leads to a restriction in the development of planktonic communities during cold periods. Unlike planktonic taxa, benthic taxa, such as the fragilarioid species, increase between 28 and 12 cm in EML (Wang et al., 2012b).

### Table 1

Pearson product moment correlation coefficients.

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<th>$\delta^{13}$C</th>
<th>C/N</th>
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<th>Diatom_flux</th>
<th>TN_flux</th>
<th>TOC_flux</th>
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<td>Diatom_flux</td>
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<td>$-0.154$</td>
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<td>$-0.369^{b}$</td>
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<td>$-0.256$</td>
<td>$0.422^{b}$</td>
<td>$0.747^{a}$</td>
<td>$0.990^{a}$</td>
<td>1</td>
</tr>
</tbody>
</table>

$^a$Correlation significant at 0.01 level (2-tailed).

$^b$Correlation significant at 0.05 level (2-tailed).

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**Fig. 4.** Scatter plots showing strong correlation between (a) TOC flux and $\delta^{13}$C, (b) diatom flux and TOC flux, and (c) planktonic to benthic (P/B) ratio and $\delta^{13}$C.

**Fig. 5.** (a) $\delta^{13}$C, (b) ratio of planktonic to benthic diatoms, (c) proportion (%) of Discostella species complex, and (d) combined relative abundance of *P. praetermissa* and *A. formosa*. 
et al., 2011; Mackay et al., 2012). In EML we suggest that the de-
crease in P/B ratio and higher C/N values (Fig. 3) are due to persistent
growth of aquatic vegetation with C/N values between 20 and 30
along the littoral zone of the lake, leading to the increase in the rel-
ative abundance of OM derived from aquatic vegetation.

Primary production in lakes can influence the changes in OM
$\delta^{13}$C values. Primary production through time can be inferred using
TOC (Leng and Marshall, 2004). A long decline in TOC flux occurs
between 33 and 12 cm (ca. 1350 to ca. 1800 AD; Fig. 3), indicating that
the productivity of algae and aquatic vegetation decreased.
TOC can also vary due to changing sedimentation rate. Here we
only can provide a constant sediment accumulation rate from 50
to 20 cm because of only one $^{14}$C date anchoring the base of the
core (Fig. 2), so we are not able to resolve this issue directly from
the data. However, a previous Holocene core taken from EML
was reported to be varved, with a relatively constant sedimenta-
tion accumulation rate of ca. 0.06 cm/yr over the past 1000 yr
(You et al., 2008), which is very similar to the sedimentation rate
estimated in this study (0.05 cm/yr). In addition, it is possible to
exclude the impact of sediment rate on TOC for the radiometric-
dally dated top of the core between 20 and 0 cm. The data also
show that the changes in TOC flux are very similar to changes in
diatom flux, likely indicating a link between the two (Fig 3). Be-
tween 50 and 12 cm, TOC flux is also almost concurrent with the
change in diatom flux and their correlation over the whole
1000 yr record is high (0.747). It is therefore likely that the changes
in TOC flux are real and not caused by changing sedimentation rate,
and therefore that they can be used as indicator of aquatic
productivity.

Enhanced productivity in lakes results in dissolved CO$_2$ becom-
ing limited; algae can adapt by switching to the utilisation of HCO$_3$^-
as carbon source, resulting in an increase in $\delta^{13}$C. However, it is
noticeable in our record that the sustained decline in TOC flux and
diatom flux is concurrent with an equally sustained increase in
$\delta^{13}$C values of ca. 3.7‰. It is unlikely that increasing $\delta^{13}$C values
in EML sediments were caused by declining aquatic productivity.

Other factors which may cause increasing $\delta^{13}$C values include
gradual maturation of lakes and changes in catchment vegetation.
As lakes mature, $^{13}$C-enriched OM is transported to bottom sedi-
ments (Meyers and Lallicher-Vergès, 1999). However, the changes
in EML are quite abrupt, e.g. at 11 cm and, moreover, the increase
in values only occurs during a specific timeframe, and so is unlikely
to be due to lake maturation. In the catchment, the development of
stable mature soils can result in increased soil respiration and the
supply of $^{13}$C-depleted dissolved CO$_2$ to lakes, leading to lower $^{13}$C
values (Hammarlund, 1993). Conversely, increasing $^{13}$C values may
be interpreted to be due to forest retreat and subsequent reduction
in catchment soil respiration. At EML, there is evidence for a de-
cline in tree pollen concentration during the period of increasing
$^{13}$C values (Li et al., 2012), but almost no change in the composition of
pollen assemblage took place. This suggests that the vegetation
cover in the region did not change to such an extent that it could
lead to an increase in $\delta^{13}$C-depleted dissolved CO$_2$.

Another potential influence on sedimentary $^{13}$C values is the difference in the proportion of phytoplankton and benthic
microalgae (France, 1995), assuming much of the sedimentary
OM is derived from the algae. France (1995) collated literature data
on $^{13}$C for marine and freshwater benthic and planktonic algae
and found that the average value for benthic algae in lakes was
$-26\%\pm 3$, whereas it was lower at $-32\%\pm 3$ for planktonic algae,
an average difference of ca. 6‰. Doi et al. (2009) also found
significant differences between $\delta^{13}$C values of plankton (lower)
and benthic (higher) microalgae sampled simultaneously across a
range of lakes. These differences may be explained by the diffusive
boundary layer effect (Jørgensen and Revsbech, 1985). The bound-
ary layer is caused by the viscous properties of water and is com-
posed of a film of water that sticks to the surface of the solid (e.g.
sediment, plants, biofilm or detritus) and does not participate in
the general circulation of the surrounding water. Its thickness
can vary from 0.2 to >1 mm and water turbulence above the
sol–water interface has a major influence on the thickness of the
boundary layer (Jørgensen and Revsbech, 1985). The diffusive
boundary layer constitutes a transfer resistance for fluxes of dis-
solved organic compounds, inorganic ions and gases such as CO$_2$
and O$_2$ across the solid–water interface, and is especially limiting
for plants and algae at high uptake rate (Jørgensen and Revsbech,
1985). A thick boundary layer, because it leads to an entrapment
of otherwise expelled $^{13}$C, results in more positive $\delta^{13}$C values
for the plants or algae (Jørgensen and Revsbech, 1985; Doi et al.,
2009).

In the EML sediments, diatoms fall into three important
functional groups, related to their preferred habitat. Benthic and
periphytic diatoms, growing on the lake bottom or attached to a
sub-stratum are generally less exposed to turbulent water, and so
likely have a thick boundary layer. By contrast, planktonic
diatoms, as they grow floating and unattached in the open water,
have a thinner boundary layer. Among planktonic diatoms, small
species such as D. stelligera and D. pseudostelliger, grow well in
strongly stratified water with weak turbulence (Wang et al., 2008,
2012a, b), whereas large planktonic diatoms, such as P. praetermissa
and A. formeda (Rioual et al., 2009; Wang et al., 2012b), need more
turbulence to enable their suspension in the photic zone. These
three functional groups have different $\delta^{13}$C values on the basis of
different diffusive boundary layers. Their rank in order of increas-
ing boundary layer thickness is large planktonic < small
planktonic < benthic.

In cold conditions with extensive ice cover, the productivity
of benthic communities in a lake is proportionally less affected than
that of the planktonic communities, because the littoral zone
may still melt and open up during summer months (Douglas and
Smol, 1999). From ca. 1350 to 1800 AD, diatom data (Wang et al.,
2012b) suggests the prevalence of a cooler climate, possibly poss-
current with the Little Ice Age. A cooler climate seems the most
likely cause for the observed decline in TOC and diatom flux, and
the decline in planktonic diatom biovolume accumulation rate
(Wang et al., 2012b). At this time there is also a marked decline
in P/B, indicative of increased proportion of benthic diatoms in
the sedimentary record. As discussed by France (1995) and
Doi et al. (2009), benthic algae are $^{13}$C-enriched vs. planktonic algae,
which may also contribute to the sustained increase in $\delta^{13}$C values
at this time.

The same process could also explain the abrupt decline in $\delta^{13}$C
values by ca. 4.2‰ after ca. 1800 AD that occurred simultaneously
with an increase in the proportion of planktonic diatoms (Fig. 5).
The decline in $\delta^{13}$C values at EML then is also coincident with the
onset of the industrial revolution and release of greater $^{13}$C
to the atmosphere due to fossil fuel combustion and deforestation.
This has resulted in the relative decline of $^{13}$C (and $^{14}$C) in atmosphe-
ric CO$_2$ over the past ca. 200 yr, which in turn has resulted in
their decline in other reservoirs, including oceans and lakes.
The process is referred to as the Suess effect (Keeling, 1979), and
is apparent in $\delta^{13}$C records from several lake sediment records
(e.g. Verburg, 2007; Castaneda et al., 2011; Jiang et al., 2011). Be-
tween ca. 1840 and 2000 AD, the decline in atmospheric $^{13}$C val-
ues was ca. 1.6‰ (Verburg, 2007). Therefore, the magnitude of
change in EML during this period cannot be attributed solely to
the Suess effect. Moreover, the Suess effect cannot explain increas-
ing $\delta^{13}$C values observed in EML during the past 50 yr. Furthermore, the sharp decline in $\delta^{13}$C values cannot be explained by a decline in algal productivity, because TOC and diatom flux increase after ca. 1800 AD (Fig. 3).

The late decline in $\delta^{13}$C to the lowest values between 1900 and 1950 AD is concurrent with the increase in planktonic diatoms which, as discussed above, have lower $\delta^{13}$C values, specifically *P. praetemissa* and *A. formosa* (Fig. 5), both of which have been shown to thrive during periods of spring and autumn turnover, when turbulence in the lake is particularly high (Morabito et al., 2002; Ptacnik et al., 2003; Rioual et al., 2009). For example, in the neighbouring Lake Shihailongwan, 18 km east of Lake Erlongwan, analysis of sediment trap samples shows that *P. praetemissa* blooms seasonally in spring and autumn, when windy conditions cause turnover and the water column is well mixed (Rioual et al., 2009). Detailed experimental studies of oligotrophic lakes showed that *A. formosa* is also abundant under conditions with high water turbulence (Morabito et al., 2002; Ptacnik et al., 2003).

The increased abundance of these two large planktonic diatoms indicates that the climatic conditions changed from cold during the Little Ice Age to warm in the 20th Century. This latest warm period is, however, not identical to the Medieval Warm period as the two differ in the relative length of the seasons (Wang et al., 2012b). During the MWP the duration of summer was longer, while the spring and autumn were shorter than in the 20th Century. During the 20th Century, long spring and autumn seasons, with strong turbulence due to strong wind, favoured blooms of large planktonic diatoms (*P. praetemissa* and *A. formosa*; Wang et al., 2012b). In such turbulent conditions, cells of these planktonic diatoms would be expected to have thinner diffusive boundary layers and more depleted $\delta^{13}$C values than those of *Discostella* species. We also acknowledge that, during periods with strongly turbulent conditions, atmospheric exchange could also contribute to the decline in $\delta^{13}$C values of the sediment because of relatively high dissolved carbon concentration in lake water leading to further discrimination against $\delta^{13}$C by the algae.

By contrast with large planktonic species, cells of *Discostella* species, a group of planktonic species which blooms in summer months when the water column is thermally stratified and water turbulence is weak (Sorvari et al., 2002; Smol et al., 2005; Rühland et al., 2008; Wang et al., 2008, 2012a) would have less depleted $\delta^{13}$C values than those of *P. praetemissa* and *A. formosa*, but more depleted $\delta^{13}$C values than benthic algae. Therefore, over the last five decades, the EML sediment record shows that $\delta^{13}$C values increased slowly with increasing proportions of *Discostella* species (Fig. 5). The increase in the relative abundance of small planktonic *Discostella* species and the concurrent decrease in large diatoms (*Asterionella* and *Punctulata*) are consistent with increased temperature, leading to strong thermal stratification of the water column (Wang et al., 2012b). The increase in *Discostella* (and other small central diatoms of the genus *Cyclotella*) with global warming has been reported for the recent sediments of numerous lakes throughout the northern Hemisphere (Sorvari et al., 2002; Rühland et al., 2003, 2008, 2010; Smol et al., 2005; Wang et al., 2012b), including nearby Xiaolongwan Lake (Panizzo et al., 2012).

We have shown that changes in the carbon isotopes signal of a lake sedimentary record can be interpreted by way of climatic changes (warm and cold) and their control of the duration of ice cover and associated variation in the ratio of planktonic and benthic algae.

In our opinion, this new approach has potential for interpreting carbon isotopes as proxy records for climate change and should be considered in future studies. However, the interpretation is useful only if due attention is paid to the following points: (i) the lake sedimentary OM should be derived almost exclusively from algae and aquatic vegetation, (ii) as water level change also cause variation in the ratio between planktonic and benthic diatom, it may also affect the carbon isotopes signal, (iii) eutrophication, as it often leads to large increase in the abundance of planktonic algae, may also alter the carbon isotope record and should also be taken into account when interpreting the record. Besides the usefulness of this new approach for interpreting carbon isotope records, it is interesting to note that diatom oxygen isotope values are also likely to be influenced by the effect of turbulence on the boundary layer thickness.

### 5. Conclusions

Interpretation of $\delta^{13}$C values in lakes sediments is complex and necessitates a multiproxy approach to unpick changes in lake productivity, the dominant producers and changes in the lake catchment. To our knowledge this study is the first to suggest that the difference in the ratio of planktonic and benthic diatoms helps interpret some of the $\delta^{13}$C variation in lake sediments. In future studies, when trying to explain variations in lacustrine organic $\delta^{13}$C, more attention should be given to the difference in isotope signatures between phytoplankton and benthic microalgae, especially when major shifts between these two groups are evident.

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