Available online http://amq.aiqua.it
ISSN (print): 2279-7327, ISSN (online): 2279-7335

Alpine and Mediterranean Quaternary, 33 (1), 2020, 99-106 https://doi.org/10.26382/AMQ.2020.09

# ARE STABLE ISOTOPES OF LACUSTRINE CARBONATE A GOOD TRACER OF LAKE HYDROLOGY AND LAKE LEVEL VARIABILITY? THE LAKE LEDRO CASE (NORTHERN ITALY).

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ABSTRACT: Oxygen and carbon isotopes were analysed on bulk samples and carbonate concretions from Lake Ledro, northern Italy, with the aim to assess their suitability as reliable proxy of the hydrological changes in the lake. For this purpose, isotopic composition of bulk marls and concretions were compared to lake level reconstruction obtained with sedimentological approach. The $\delta^{13}C_{bulk}$ record mostly mirrors the $\delta^{18}O_{bulk}$ record, while carbon and oxygen isotopes from concretions are not correlated. We found that there is a statistically significant correlation between the oxygen and carbon isotopic record of the bulk carbonate and lake level changes, with the positive changes in lake level corresponding to lower values on $\delta^{18}O_{bulk}$ and $\delta^{13}C_{bulk}$ and vice versa. Assuming that most of the bulk carbonate is formed by bio-induced calcite precipitation, this means that this kind of carbonate reflects the local hydrological condition of the lake water better than lacustrine concretions, which instead are more affected by local conditions of precipitation like biological plant activity.

Keywords: stable isotopes carbonate; lacustrine carbonates; lake level; Lake Ledro.

## 1. INTRODUCTION

The hydrological balance of lakes may respond sensitively to changes in net precipitation and humidity, which often are accompanied by change in the lake level. Hence, the lake level is a sensitive proxy that document past changes in lake water budget, and past climate (e.g., Magny, 2013 and reference therein).

In lacustrine settings, stable isotopes of carbonates are a frequently used as proxy to identify and reconstruct terrestrial environmental changes and have been extensively used in Quaternary paleoenvironmental reconstructions (e.g., Eicher & Siegenthaler, 1976; Punning et al., 1984; Lotter et al., 1992; Mayer and Schwark, 1999; Zanchetta et al., 1999; Schwander et al., 2000; Hammarlung et al., 2003; Andresson et al., 2010; Jonsson et al., 2010; Whittington et al., 2015). The oxygen isotopic composition of inorganic lake carbonate is mainly influenced by: (1) the oxygen isotope composition of precipitation in the watershed, (2) the water temperature at time of carbonate precipitation, and (3) the hydrological balance of the lake (Buchardt & Fritz, 1980; Gibson et al., 1993; von Grafenstein et al., 2000; Ito, 2001; Leng & Marshall, 2004; Bernasconi & McKenzie, 2007). In specific conditions, the hydrological balance dominates the oxygen isotopic signal of water lakes and, consequently, that of inorganic carbonates (Roberts et al., 2008). So it should be related to changes in lake level. Despite this argumentation is commonly used as rational for interpreting the isotope signal as proxy of the lake level change (e.g., Leng et al., 2010), there are very few examples of direct comparison between lake level records and oxygen isotope composition of lake carbonate. A comparison between bulk lacustrine carbonate stable isotopes and changes in lake level reconstructed using sedimentological approach (Magny, 1992, 1998) have been performed for instance on the study of Gerzensee Lake, on the Swiss Plateau at 603 m a.s.l., and Lautrey lake, located at 788 m a.s.l. in the Jura mountains (eastern France) (Magny, 2001, 2013; Magny et al., 2006).

When using the carbon and oxygen isotopes for lake level reconstruction, many complex factors have to be carefully taken into account. Usually, the interpretation of the $^{13}C/^{12}C$ ratio in lacustrine carbonate is more complex than that of $^{18}O/^{16}O$ ratio. Variations of $\delta^{13}C$ values of precipitated calcite in general reflect the $\delta^{13}C$ values of the dissolved inorganic carbon (DIC), which is controlled by the isotopic composition of incoming waters, the exchange of $CO_2$ between lake surface waters and the atmosphere, by photosynthesis and respiration of algae and aquatic macrophytes, and by organic mat-

ter decomposition (McKenzie, 1985; Lee et al., 1987; Meyers & Teranes, 2001; Teranes & Bernasconi, 2005) as well as by carbonate dissolution in the water column. Moreover, the input and the isotopic composition of the DIC from the catchment depends on several factors such as leaching of soil $CO_2$ and dissolution of old carbonates (Leng & Marshall, 2004; Zanchetta et al., 2018).

Several types of lacustrine carbonates can be studied, like bio-induced carbonates (the most common, sometime called authigenic carbonates; Eicher & Siegenthaler, 1976; Hammarlund et al., 1999, 2003), freshwater shells (e.g. Bottger et al., 1998; Zanchetta et al., 1999; Bonadonna et al., 1999; Jones et al., 2002), ostracods (e.g. von Grafenstein et al., 1994, 1999; Lézine et al., 2010) and *Chara* encrustations (Apolinarska & Hammarlund, 2009). However, each different type of carbonate forms in specific conditions and, therefore, may give a different type of information (Leng & Marshall, 2004).

In this work, we further explore the relation between oxygen and carbon isotopes and lake level changes using data on bulk carbonates (mainly bio-induced) and lake concretions in cores recovered in Lake Ledro within a Holocene succession, for which chronology and lake level reconstruction has already been performed (Magny et al., 2009).

## 2. SITE DESCRIPTION

Lake Ledro is located at 652 m a.s.l. on the southern slope of the Alps (N-E Italy, Fig. 1), with a surface area of ca. 2.17 $km^2$, a maximum water depth of 48 m, a mean depth of 35 m, and a volume of ca. 0.75 $km^3$. The catchment area is ca. 131 $km^2$ and is developed on a substratum mainly composed by carbonate rock, including Triassic (dolomite) and Jurassic-Cretaceous limestones covered by Quaternary glacial deposits. The outlet of the lake is Ponale River, which is responsible for the downcutting of the local morainic dam (Beug, 1964) and hydrologically links Lake Ledro to the Lake Garda (Fig. 1). Since 1929, the lake level has been artificially regulated for hydroelectric power supply, modifying consequently the natural hydrological variability. Lake Ledro is fed by many submerged springs and by some minor temporary torrential tributaries: Ponale - Massangla, Assat di Cencei and Sat di Pur, which are dry for most of the year.

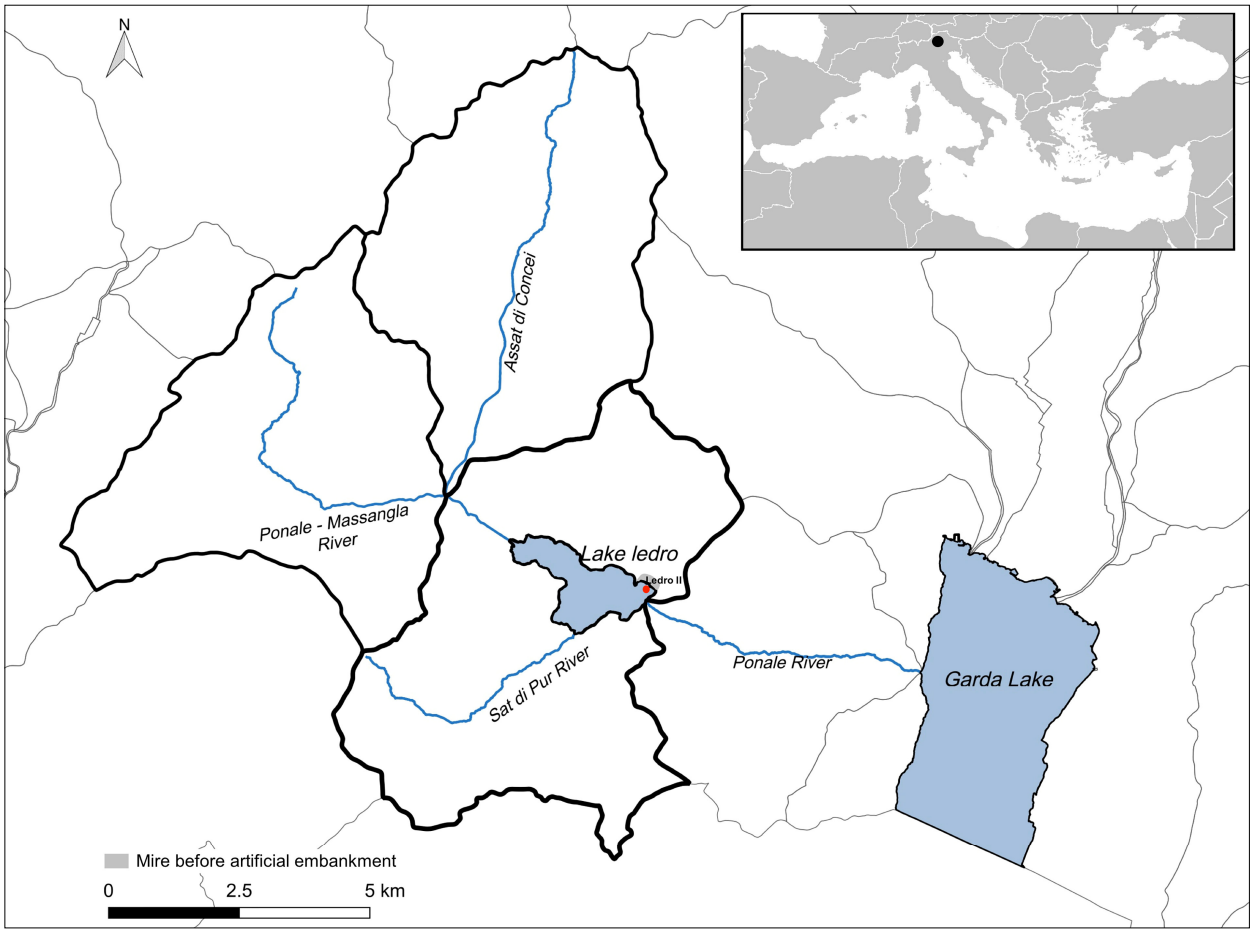


Fig. 1 - Location of Lake Ledro catchment with an outline map showing the study site in Northern Italy. The circle marks the core site (core Ledro II). The thick black line indicates the sub-catchment of the different inlet rivers.

The climate of Ledro Valley is influenced by the near Garda Lake (65 m a.s.l.), characterized by particularly mild climatic conditions. The mean temperature, recorded at Bezzecca weather station (710 m a.s.l.), is ca. 0 °C in the coldest month (January) and ca. 19 °C in the warmest month (July). The mean annual precipitation is ca. 1220 mm, with seasonal maxima in spring and autumn.

Lake Ledro waters can be classified as Ca-bicarbonate and are stratified during summer period (Corradini & Flain, 2000).

The yearly mean isotopic composition of precipitations collected from five stations from 1992 to 2001 around the Garda area and reported by Longinelli & Selmo (2003) is -7.47 ±0.91 ‰ for $\delta^{18}O$ and -49.3 ±7.3 ‰ for $\delta^{2}H$. The weighed mean isotopic values of precipitations measured at Riva del Garda from February 2007 to January 2008 (Longinelli et al., 2008) is -7.25 ±0.70 ‰ for $\delta^{18}O$ and -52.1 ±5.9 ‰ for $\delta^{2}H$.

## 3. MATERIALS AND METHODS

The present study was carried out on the core retrieved in April 2005 at the site Ledro II, located on the north-eastern shore of the lake (Fig. 1). Here, a ~ 3 m long core was collected by means of a Russian peat corer (Magny et al., 2009). The sedimentary succession, under the superficial peat layer, shows an alternation of carbonate lake-marl and organic-rich layers. Detailed core description, chronology, pollen and lake-level reconstructions are extensively discussed by Magny et al. (2009, 2012), to which the readers are referred for any details. The recovered sediments, dated by means of radiocarbon, represent ca. the last 7000 yr BP.

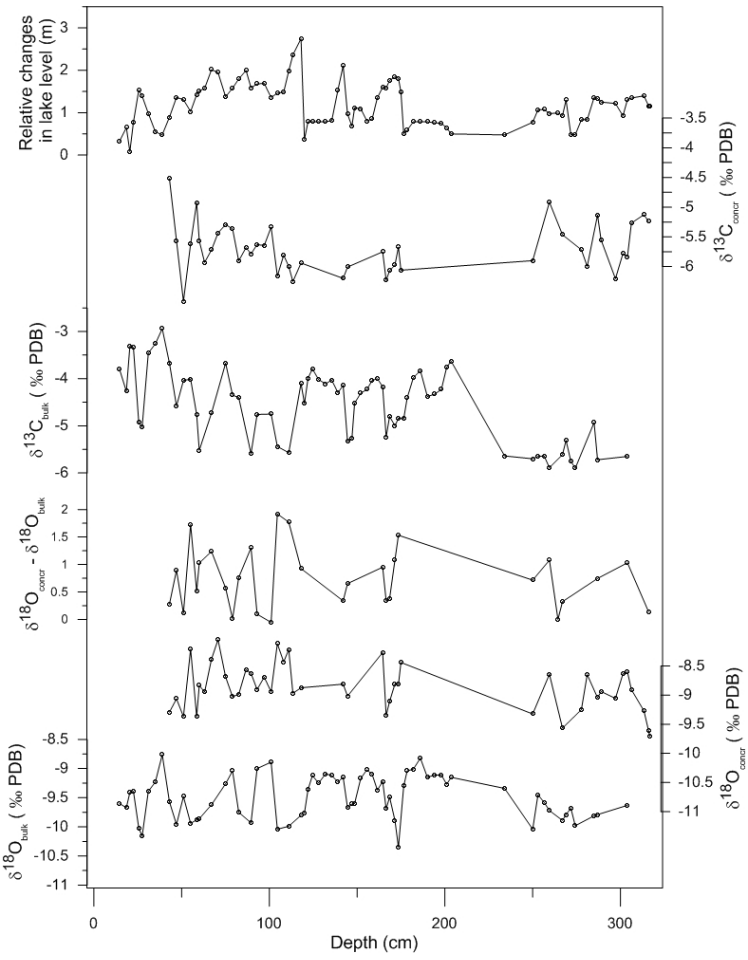


Fig. 2 - Stable carbon and oxygen isotope records of different carbonate components, together with lake level changes, plotted against core depth.

The carbonate lake-marl samples and carbonate concretions were sampled for oxygen and carbon isotopes on bulk carbonate about every 3-4 cm. Sediments were sieved at 200 μm. The term "bulk carbonate" is here used for the fine-grained inorganic lake marl (less than 200 μm), mostly devoid of shell and ostracods remains. The carbonate concretions where hand-picked under a binocular microscope from the coarser fraction (larger than 200 μm). This latter material corresponds to the original material selected by Magny et al. (2009) for defining lake level oscillations based on different typology of lacustrine concretions (Magny, 2004). Concretions were cleaned ultrasonically with distilled water to separate adhering particles. The sediment samples were dried at 40°C, powdered and analysed for isotopic composition with no further pre-treatment considering the high carbonate content (Manella et al., 2020).

All the carbonate samples were reacted with 100% phosphoric acid at 70 °C for 1 h under vacuum to release $CO_2$. The released $CO_2$ was then cryogenically-cleaned in a vacuum line and analysed in the mass spectrometry Varian Mat 252. The results were expressed with the well-known δ-notation and normalized to the Vienna Pee Dee Belemnite scale (PDB in *per mil*) using internal working standards of carbonate- MS, MAB (Carrara Marble) and MOM, which were cross-checked against the international standard NBS18. The precision (as standard deviation) of the analyses was better than 0.2‰ for $\delta^{13}C$ and 0.2‰ for $\delta^{18}O$.

## 4. RESULTS

Figure 2 shows isotopic composition of the bulk and carbonate concretions, compared to lake level data vs depth. The $\delta^{18}O$ values of bulk sediment ($\delta^{18}O_{bulk}$) vary in a range of ca. 1.6 ‰ from ca -10.4 - to -8.8 ‰, very similar to the range (ca. 1.7‰) of $\delta^{18}O$ values of concretions ($\delta^{18}O_{concr}$) but the maximum and minimum $\delta^{18}O_{concr}$ values are lesser negative, ranging between -9.7 and -8.0 ‰. Indeed, considering each couple of $\delta^{18}O_{bulk}$ and $\delta^{18}O_{concr}$ at the same depth, the $\delta^{18}O_{concr}$ has higher oxygen isotope composition than $\delta^{18}O_{bulk}$. This means that the difference between $\delta^{18}O_{concr}$ and $\delta^{18}O_{bulk}$ is usually positive (Fig. 2) with a mean value of 0.8 ‰ and a range from 1.9 to -0.1‰.

The $\delta^{13}C$ values of bulk sediment ($\delta^{13}C_{bulk}$) fluctu-

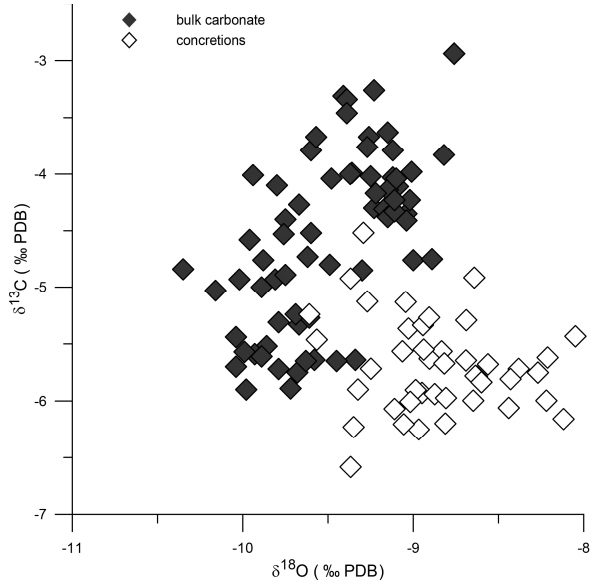


Fig. 3 - $\delta^{18}O$ versus $\delta^{13}C$ diagram for the core record of both bulk and concretions.

ate in a wider range of ca. 3 ‰, i.e., from ca. -2.9 to -5.9 ‰ (Fig. 2). The carbon isotopes of concretions are offset from the corresponding values of bulk carbonate of -0.8 ‰ on average with a range from -2.5 to +0.5 ‰.

The $\delta^{18}O_{bulk}$ record is characterized by a relatively low variability from the bottom to around 190 cm, from which frequent and short fluctuations start and minimum $\delta^{18}O$ values are reached at around 173 cm, 147-144 cm, 118-104 cm, 89 cm, 55 cm, 47 and 22 cm. The $\delta^{18}O$ of concretions shows similar fluctuations toward more negative values at 173 cm, 144 cm and 104 cm, even if in general there is not a statistically significant correlation between the two records (r=0.22, n=32).

The $\delta^{13}C_{bulk}$ record mostly mirrors the $\delta^{18}O_{bulk}$ one with a good correlation (r=0.59, n=67), in contrast to the carbon and oxygen values of concretions that are not significantly correlated (Fig. 3). This is probably related to the presence of different kind of concretions, but also to the different, local, nature of the environment where lacustrine concretions forms compared to bio-induced carbonates.

The $\delta^{13}C_{bulk}$ values are generally $^{13}C$-depleted from the bottom of the record to 230 cm, when the values start to increase reaching the maximum of -3.8 ‰ at around 200 cm. It has to be noted that from 250 to 175 cm there are not samples of concretions analysed because there was not sufficient material, in accord by the fact that the core from 240 to 190 cm is characterized by peat and gyttja and that from 230 cm to 190 cm there are only organic remains without concretions (Magny et al., 2009). From 200 cm to the top of the record, $\delta^{13}C_{bulk}$ is characterized by frequent fluctuations with the lowest and highest values corresponding to the lowest and highest values on the oxygen record of bulk carbonates, respectively.

## 5. DISCUSSION

$\delta^{18}O$ of inorganic carbonates in small/medium open lakes, like Lake Ledro, dominantly reflects the $\delta^{18}O$ of the water during calcite precipitation (Ito, 2001; Leng & Marshall, 2004; Teranes et al., 1999). Authigenic calcite inorganically precipitates in late spring/early summer in the epilimnion because of changes in lake water chemistry (increased temperature, algal activity and pH changes). However, inorganic carbonates may also be formed as an extra-cellular by-product during photosynthesis. For example, *Characeae* and other micro- and macrophytes actively remove bicarbonate from the lake system, leading to the formation of inorganic calcite, which typically encrusts the plants (von Grafenstein et al.,

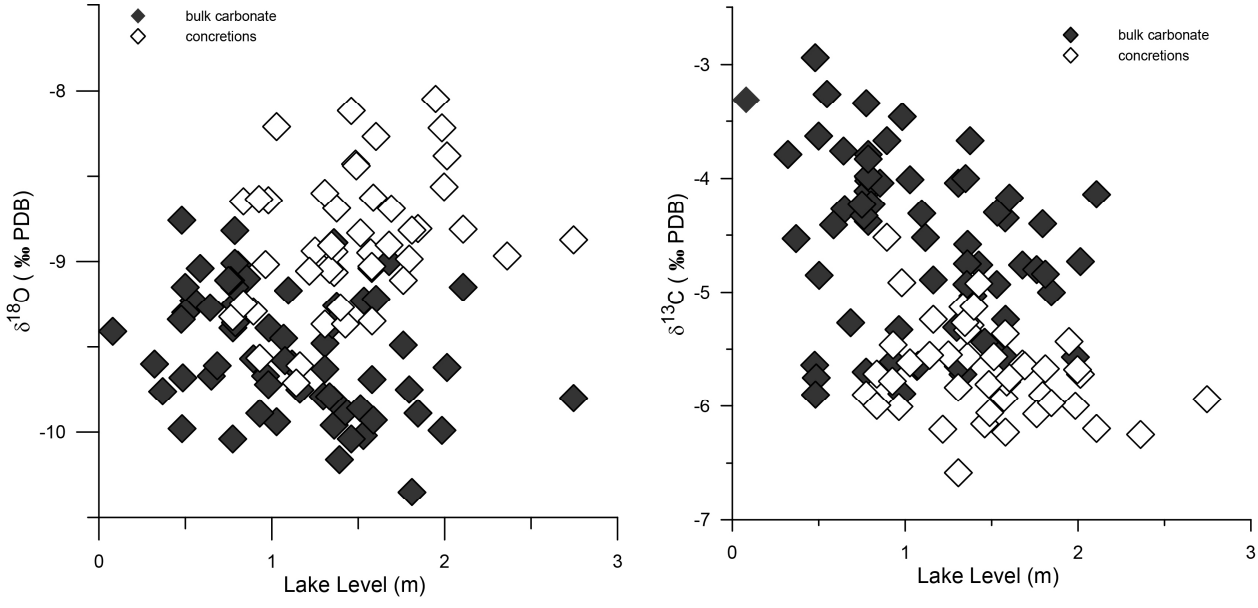


Fig. 4 - Stable carbon and oxygen isotope records of different carbonate components plotted against lake level changes.

2000). For the Lake Ledro record, encrustations and macroscopic components were isolated and used for lake level reconstruction (Magny et al., 2009) and isotopically analysed together with the isotopic characterization of the fine-grained carbonate matrix, which could be supposed to be composed by a mix of authigenic calcite and disaggregated calcitic concretions, as evidenced for other marl lakes (von Grafenstein et al., 2000; Apolinarska & Hammarlund, 2009). However, it is reasonable to expect that fine-grained matrix bulk carbonate is mostly related to the general water condition in the upper part of the lake water, where algae photosynthesis occurs driving bio-induced calcite precipitation, whereas concretions are more related to local, plant driven, conditions.

In general, in hydrologically open lakes with fast turnover, temperature and the isotopic composition of precipitation entering the lake will be the most important drivers of final $\delta^{18}O$ values of carbonates. However, in lacustrine system evaporation influences the isotopic composition of lake water in relation to the residence time and to the morphometric characteristic of the lake itself and local climate (Gonfiantini, 1986). In particular, in closed lakes, with long residence times, the effect of evaporation will usually be far more important (Li & Ku, 1997; Leng & Marshall, 2004; Leng et al., 2006). Some lakes nearby Lake Ledro, with different morphometric parameters and elevation, show a seasonal variation on the isotopic composition of their waters, especially in the epilimnetic values, due to evaporation and, in general, their isotopic composition of water is related to hydrological processes (Baroni et al., 2006; Perini et al., 2009; Natale et al., 2013; Flaim et al., 2013).

Lake Ledro turnover time is less than 1 year (0.8 years; Casellato & Caneva, 1994) but considering that the rivers feeding the lake are dry for most of the year, we can hypothesize that the water isotopic composition is mainly controlled by the rate of evaporation, and the supply of local meteoric precipitation and spring waters. In particular, the rate of evaporation would be important in summer. In this situation, we can assume that the oxygen isotopes of lacustrine carbonates should be mainly interpreted as changes in the hydrological budget of the lake rather than changes in temperature (e.g. Roberts et al., 2008; Zanchetta et al., 2012). Higher $\delta^{18}O$ values of lake water, especially in the epilimnion, and thus higher $\delta^{18}O$ of carbonates, should correspond to phases of reduced precipitation with lower contribution from the torrential tributaries and/or increase of evaporation rate (i.e., drier period), while lower $\delta^{18}O$ values should correspond to an increase in the amount of precipitation and/or reduction of the evaporation of lake water (i.e., wetter period). However, this can be complicated by temperature changes and by the changes in the isotopic composition of meteoric precipitation, which act as additional factors that ultimately can either amplify or partially suppress the isotopic signal linked to changes in the rate of evaporation (Dansgaard, 1964; Rozanski et al., 1992; Fricke & O'Neil, 1999). Because lake carbonates (in particular algal bio-induced precipitation) occur mainly in summer, their isotopic composition is skewed toward this season, even if the winter recharge is important in defining the final isotopic budget (Bini et al., 2019). Overall, we can then assume that higher $\delta^{18}O$ values of carbonates correspond to drier period, in which lake level also decreases.

Considering the lake level reconstructed by Magny et al. (2009) from Ledro core, the data showed in Figure 2 highlight that there is a good correspondence between the oxygen and carbon record of the bulk carbonate, with the positive changes in lake level corresponding to lower values on $\delta^{18}O_{bulk}$ and $\delta^{13}C_{bulk}$ and, vice versa, with negative changes in lake level corresponding to higher values on $\delta^{18}O_{bulk}$ and $\delta^{13}C_{bulk}$. This behaviour is evident also for the carbon record of concretions, with higher $\delta^{13}C_{concr}$ values corresponding to lower level lake, whereas $\delta^{18}O_{concr}$ shows in some case opposite trends compared to lake level.

In fact, considering all the record there is a statistically significant (at 0.05 significance level) negative correlation between the lake level changes with the $\delta^{18}O_{bulk}$ (r=-0.33, n=67) and the $\delta^{13}C_{bulk}$ values (r=-0.30, n=66) (Fig.4). The correlation with the concretions and the lake level changes is also negative considering the carbon isotopes record (r=-0.31, n=44), whereas there is a positive correlation with the oxygen isotope record of concretion (r=+0.32).

Hence, for Lake Ledro the $\delta^{18}O_{bulk}$ record correlates quite well with lake level changes, with $^{18}O$-depleted values corresponding to phases of lake level increasing (Figs 2 and 4). On the contrary, lowering of lake level matches with higher $\delta^{18}O_{bulk}$ values. This confirms that $\delta^{18}O_{bulk}$ is can be considered as a reasonably good proxy for lake hydrological changes and lake level, in particular for lakes with simple hydrological budge.

$\delta^{13}C$ of carbonate is dependent on carbon isotope composition of the DIC, controlled by several processes (e.g., Leng & Marshall, 2004) including: exchange with atmospheric $CO_2$, respiratory activity of aquatic plants, mineralization of organic matter and DIC isotopic composition of the inflowing water, influenced by the dissolution of carbonate rocks from the catchment and leaching of soil $CO_2$. However, $\delta^{18}O_{bulk}$ and $\delta^{13}C_{bulk}$ of Lake Ledro show a good correlation (Fig. 3), which could indicate that data are controlled by the same external factors. Period of lower lake level and higher $\delta^{18}O_{bulk}$ and $\delta^{13}C_{bulk}$ can indicate a decrease of contribution of soil-derived $CO_2$ by input water and higher equilibration with atmospheric $CO_2$ (Zanchetta et al., 2018). Whereas wetter period can increase the flush of soil $CO_2$ and increasing of recycling of organic matter in a deeper lake (Whittington et al., 2015; Zanchetta et al., 2018). On the other hand, as Horton et al. (2016) pointed out, evaporation causes $^{18}O$ enrichment in water and $^{13}C$ enrichment in DIC. They suggested that evaporation induces increases in alkalinity (i.e. increased bicarbonate concentration) and drives DIC $^{13}C$-enrichment through isotopic equilibrium exchange with atmospheric $CO_2$ (ca. 8‰ at 25°C in the $HCO_3$-$CO_2$ system; Mook et al., 1974). Hence, the highest $\delta^{13}C_{bulk}$ values are originated by $^{13}C$-enriched DIC due to equilibration with atmospheric $CO_2$ (e.g, Leng et al., 1999), enhanced by $CO_2$ outgassing linked to strong evaporation (Talbot, 1990) and the enhanced residence time. In warm conditions, there is also a strong biological activity leading to enhanced $^{13}C$-enrichment in the water (Siegenthaler & Eicher, 1986), if

not compensated by recycling. The possible influence of detrital materials could be neglected as supported by the near absence of lithoclasts found in the samples analysed (Magny et al., 2009).

On the other hand, the absence of correlation between $\delta^{18}O$ and $\delta^{13}C$ of the concretions could be explained by the different origin of the bio-induced carbonates that form the encrustations. Each of these encrustations, differentiated by Magny in several morphotypes, should be characterized by different "vital offset", spaces and times in which they formed. This could lead a strong variability of the different isotope compositions, with no clear and apparent correlation with environmental variables. On the contrary, isotopes of bulk carbonates (in particular the fine fraction) better average the lake conditions and, hence, should be good tracers of hydrological changes of lake itself.

## CONCLUSIONS

In this work, we used the oxygen and carbon isotope signatures of different type of carbonate (bulk and concretions) from Lake Ledro to test their suitability in detecting lake-level oscillations and, then, whether they are good indicators of hydrological changes of this lacustrine system. Our data confirm that different carbonate types have statistically significant different compositions and reflect hydrological changes in different way. In particular, the isotopic composition of bulk carbonate (grain size below 200 μm) shows a correlation with lake level data, allowing us to consider this geochemical signal a good proxy for lake hydrological changes. On the contrary, the concretion isotope values show high variability and did not correlate with lake level changes. This study further indicates that grain size separation would be an efficient and fast method for improving quality of samples in shallow lake deposit rich in concretions.

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*Ms. received: July 13, 2020* *Revised: October 31, 2020*
*Accepted: November 1, 2020* *Available online: November 30, 2020*